Spin Dynamics in an Antiferromagnet

Diploma thesis
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Chapter 1

Introduction

The antiferromagnetic state of matter is invariably associated with the name of Louis Néel. In 1932, Néel introduced the concept of two interleaved sublattice magnetisations which are coupled by a—at that time revolutionary—negative molecular field. This allowed to explain the anomalous strength and temperature dependence of the magnetic susceptibility of PtCo alloys [1, 2]. The discovery of this new magnetic state of matter and its experimental observation via neutron diffraction in 1949 [3] paved the way for the awarding of the Nobel prize in physics to Néel in 1970.

The 1970s and 80s also saw the birth of a new magnetic field of research, spintronics. This acronym, composed of spin, transport and electronics, describes effects that exploit and manipulate the spin of an electron in order to transport information. The most prominent representative of spintronics is the giant magnetoresistance (GMR) effect which enables the efficient read out of magnetic information in modern hard disks. It was discovered by Peter Grünberg [4] and Albert Fert [5] in 1988, who were then awarded the Nobel prize in physics in 2007.

In current spintronic devices, antiferromagnets mostly play a static supporting role, e.g. in GMR-type spin valves by enhancing the magnetic hardness of one ferromagnetic layer through the exchange-bias effect [6, 7]. Yet, there is a growing interest in magnetoresistive effects controlled solely by antiferromagnets [8, 9, 10]. Spin transfer torque [11, 12], the manipulation of magnetism through spin-polarized currents, is e.g. an effect for which antiferromagnets might prove advantageous. Due to the absence of shape anisotropies, the expected critical current densities to switch the antiferromagnetic magnetisation orientation are expected to be one or two orders of magnitude smaller than in ferromagnets [8, 13]. This would reduce the heating of spintronic devices based on antiferromagnets.

In this thesis, we want to investigate the influence of antiferromagnets on another central research topic of spintronics: the creation of pure spin currents. A spin current is the transport of angular momentum without an accompanying charge current. In exchange coupled systems, spin currents can be completely decoupled from the drift of conduction electrons. In this way, spin currents are not bound to metallic systems and may also flow in magnetic insulators [14]. Furthermore, spin currents are predicted to be dissipationless [15], which would reduce accompanied
heat production and enable a further minimization of microelectronic devices. Naturally, this has awakened a great interest to detect and characterize potential sources of spin currents.

In the last decade, several spin current creation mechanisms have been reported, such as the spin Hall effect [16] and the spin Seebeck effect [17, 18]. One of the most rigorously validated spin current generation concepts is spin pumping: The magnetisation of a ferromagnet is driven into resonant precession by microwave irradiation, and relaxes towards equilibrium by the emission of a spin current into an adjacent metal. The concept has extensively been studied and was validated for isolating, semiconducting and metallic ferromagnets as well as isolating ferrimagnets [19, 20].

The present thesis investigates whether spin current creation via spin pumping is achievable also in antiferromagnets. Antiferromagnets are attractive in this context, as in contrast to ferromagnets, they possess no net magnetisation, but still exhibit long range magnetic order to which conduction electron spins may couple. Furthermore, the spin flop transition of antiferromagnets enables the induction of a magnetic moment by means of an external control parameter, the external magnetic field. High enough field strengths will thus transform an antiferromagnet into a canted ferromagnet-like state. It is the central question of this thesis, if spin currents may be generated by spin pumping with antiferromagnets in either the antiparallel magnetisation configuration or the spin flop state—or in both.

Our antiferromagnet of choice is MnF$_2$. Being thoroughly investigated [21, 22, 23, 24], MnF$_2$ shows narrow resonance line widths which imply low intrinsic damping [25]. Moreover, the crystalline anisotropy of MnF$_2$ is adequately described by a uniaxial easy axis anisotropy [26], which turns MnF$_2$ into an excellent model antiferromagnet.

This thesis is structured as follows:

Chapter 2 introduces the theoretical concepts behind spin pumping. After briefly summarizing the properties of MnF$_2$, a review of electron spin and ferromagnetic resonance is given, followed by a thorough discussion of antiferromagnetic resonance (AFMR). Thereafter, the spin pumping concept and the detection principle of spin currents are introduced. The chapter ends with a discussion of the recently discovered spin Hall magnetoresistance.

In Chapter 3 we present our experimental AFMR results of MnF$_2$ single crystals. The measured resonance dispersion is insufficiently described by the state of the art AFMR theory. Based on the Landau-Lifshitz equation, we thus develop a numerical AFMR simulation, which is able to determine the resonance frequency, line width and magnetisation motion in arbitrary field configurations. The comparison of simulated and experimental resonance dispersion yields excellent agreement and enables the determination of the crystal alignment in the external field. Finally, the
measured resonance line width and the temperature dependence of the AFMR are discussed.

Chapter 4 turns towards the main goal of this thesis, antiferromagnetic spin pumping. The observed voltage signal, however, does not exhibit the properties of a spin current injected into a normal metal, leaving the spin pumping mechanism undetectable in our experiments. A discussion of the observed signal reveals that it is likely to be caused by microwave rectification, which requires an anisotropic magnetoresistance in our MnF$_2$|Pt layers—an effect so far not reported.

Therefore, presented in Chapter 5, we investigate the resistance of our MnF$_2$|Pt heterostructures as a function of external field. We find a variation of the resistance at the spin flop field, which is to our—knowledge—the first report of a resistance dependence of a normal metal on the magnetic configuration of an adjacent, electrically isolating antiferromagnet.

The thesis ends with a summary of the results in Chapter 6 and a discussion of viable experiments to investigate the angular dependence of the observed antiferromagnetic resistance variation.

Unrelated to the experiments with antiferromagnets, we have also investigated the high frequency behaviour of the recently discovered spin Hall magnetoresistance (SMR) in yttrium iron garnet | platinum structures. The measured impedance resembles the DC SMR effect for frequencies < 200 MHz, while above this frequency, the SMR effect size increases. The results of these experiments are presented in Appendix C.
Chapter 2

Theory

This Chapter introduces the main theoretical concepts of this thesis. First an introduction to general properties of model antiferromagnets is given. Then magnetic resonance phenomena of single electrons and ferromagnetic systems are studied, followed by a thorough discussion of antiferromagnetic resonance. Thereafter, spin currents and their generation by spin pumping in ferromagnets is examined. Finally, a recently discovered magnetoresistive effect of isolating ferromagnet|normal metal bilayers is introduced: the spin Hall magnetoresistance.

2.1 General Properties of Antiferromagnets

Antiferromagnets consist of coupled magnetic moments (spins), whose exchange integral is, in contrast to ferromagnets, negative. Therefore spins at neighbouring sites align antiparallelly, giving rise to a long range magnetic ordering, but not to net magnetisation. Simple antiferromagnets (like MnF$_2$) are effectively described by two sublattice magnetisations $M_1$ and $M_2$, which are coupled via a Weiss molecular field $H_E$ [27, 2], aligning them antiparallelly. Analogous to ferromagnets, the exchange interaction introduces an ordering temperature below which the thermal energy is smaller than the magnetic energy, resulting in long range order. This ordering temperature depends on the ratio of coupling strength and $k_B$ and is called the Néel temperature $T_N$ [28]. Above $T_N$, the thermal energy dominates, the long range order is destroyed and the magnetic moments behave paramagnetically.

Apart from the mentioned exchange interaction and the always present Zeeman interaction, crystalline anisotropies play an important role in antiferromagnets. The form and strength of the anisotropies determine the equilibrium orientation of the sublattice magnetisations below the spin flop field (cf. Section 2.1.2). Crystalline anisotropies exist in various kinds (e.g. uniaxial, cubic), but in the following we restrict the discussion to the simplest form, the uniaxial anisotropy. MnF$_2$, which we consider as model antiferromagnet in this thesis, is adequately modelled using an easy axis anisotropy only [24].
Chapter 2  Theory

Free Energy Term | Effective Field | Conversion
--- | --- | ---
External Field | $-\mu_0 H_0 \cdot M_i$ | $H_0$ | $-\mu_0 H_0 = \frac{2K_{uni}}{\mu_0}$
Uniaxial Anisotropy | $-K_{uni}(c \cdot m_i)^2$ | $H_A$ | $\mu_0 H_A = \frac{2K_{uni}}{\mu_0}$
Exchange Interaction | $\lambda \mu_0 M_1 \cdot M_2$ | $H_E$ | $\mu_0 H_E = \lambda M$

Table 2.1: List of important free energy contributions of an antiferromagnet and the respective effective fields. In addition, the conversion formulas between the effective field strength and the corresponding free energy terms are given. $K_{uni}$ is the uniaxial anisotropy constant, which is positive for a crystalline easy axis. $c$ is the anisotropy axis' unit vector, and $m_i$ and $M$ are the unit vector and magnitude of the $i$th magnetisation vector, respectively. $\lambda$ represents the molecular field strength and is positive for antiferromagnetic alignment.

In the same way as the exchange interaction is represented by an effective molecular field $H_E$, the effect of anisotropy may be expressed by an effective magnetic field, too—the anisotropy field $H_A$. The direction of $H_A$ coincides with the axis of uniaxial anisotropy, while its free energy contribution $-\mu_0 H_A \cdot M_i$ ideally should resemble the original, uniaxial anisotropy free energy term. Often however, this is impossible, since the uniaxial anisotropy has a more elaborate dependence of $M_i$ than a Zeeman-type energy term $H_A \cdot M_i$ can capture. The advantage of the effective field concept, however, is the intuitive description of the acting torques in a qualitative picture. Its drawback is the reduced accuracy compared with the free energy description.

Throughout the course of this thesis, both the free energy description and the concept of effective fields are used. Table 2.1 summarizes these terms for the important contributions of a model antiferromagnet like MnF$_2$. In addition, conversion formulas between the strength of the effective field and the corresponding free energy values are given.

2.1.1 Susceptibility

The uniaxial easy axis anisotropy introduces a specific direction for the alignment of the sublattice magnetisation. Due to this breaking of symmetry, the susceptibility tensor is not isotropic. Instead, we have to distinguish between the parallel susceptibility $\chi_{||}$, if the external field is applied along the anisotropy axis and the perpendicular anisotropy $\chi_{\perp}$, if the field is applied in the plane perpendicular to $H_A$.

At $T = 0$ K, an external field parallel to the uniaxial anisotropy increases the free energy of one sublattice, while reducing the other’s. In total the energy changes cancel each other out and the magnetisation configuration with antiparallel magnetisations $M_i$ along the axis of uniaxial anisotropy $H_A$ is still energetically favorable. Thus, the net magnetisation remains zero and $\chi_{||} = 0$ [29]. In contrast, with $H_0$ perpendicular to $H_A$ both magnetisations can reduce their free energy, by tilting
slightly towards the applied field, at only small anisotropy costs. Thus for \( H_0 \perp H_A \) a net magnetisation is induced and \( \chi_\perp > 0 \). The value of \( \chi_\perp \) at \( T = 0 \) K is determined by the molecular field strength \[30\]:

\[
\chi_\perp = \lambda^{-1}
\]

(2.1)

With finite temperature, the effect of thermal fluctuations increases the parallel susceptibility, while the perpendicular susceptibility remains at a constant value. At \( T = T_N \), \( \chi_\parallel \) resembles the value of \( \chi_\perp \) and the susceptibility tensor is isotropic for \( T \geq T_N \) \[28, 30\].

### 2.1.2 Spin Flop

Probably the most unique feature of antiferromagnets is the change of magnetic configuration at a a certain critical field \( H_C \), also known as spin flop field. As mentioned, the magnetisations align parallelly with \( H_A \) for weak external fields \( H_0 \), as depicted in Figure 2.1 (a). At a high enough field strength \( H_0 = H_C \), the former magnetic configuration is no longer energetically favourable. Instead the magnetisations turn to a position perpendicular to \( H_A \) and bend slightly towards the external field, gaining Zeeman energy (Figure 2.1 (b)). At these field strengths, the gained Zeeman energy outweighs the loss of anisotropy energy and the (small) loss of exchange energy. For even higher fields, the magnetisations further bend towards \( H_0 \) gaining both Zeeman and crystalline anisotropy energy at the cost of exchange energy.

The value of \( H_C \) depends on the strength of the crystal anisotropy field and the molecular field \[22, 25\]:

\[
H_C = \sqrt{H_A (H_A + 2H_E)}
\]

(2.2)
2.2 Magnetic Resonance

This section addresses the theory of spin resonance phenomena and its adoption to antiferromagnets. Starting with the quantum mechanical treatment of an electron spin in an external magnetic field, the steps and equations necessary to describe electron spin resonance (ESR) and ferromagnetic resonance (FMR) are laid out. The section ends with a thorough discussion of antiferromagnetic resonance (AFMR), based on the theoretical work of Keffer and Kittel in the early 1950s [31].

2.2.1 Electron Spin Resonance

Before dealing with exchange coupled systems, we first consider the problem of a single magnetic moment $\mu$ in an external magnetic field $H_0$. The Hamiltonian is given by the Zeeman energy term $H_{\text{Zeeman}} = -\mu \cdot \mu_0 H_0$. The corresponding eigenstates are quantized by the magnetic quantum number $m_j$ and given by

$$E_j = g\mu_0 \mu_B m_j H_0$$

(2.3)

Thus, as depicted in Figure 2.2, the energy levels split with increasing field. For each field strength, there is a (microwave) frequency allowing to excite transitions between states $i$ and $k$. For these magnetic dipole transitions $\Delta m = m_i - m_k = \pm 1$ and

$$\Delta E = E_i - E_k = \hbar \omega_{\text{res}} = g\mu_B \mu_0 H_0$$

(2.4)
Each electromagnetic multipole transition requires a specific polarisation of the electromagnetic wave. Here, for magnetic dipole M1 transitions with $\Delta m = \pm 1$, the polarisation of the microwave magnetic field $\mathbf{H}_{\text{rf}}$ has to be perpendicular to $\mathbf{H}_0$, meaning that spin resonance can only be excited, if the polarisation of the electromagnetic wave possesses components perpendicular to the static magnetic field.

Interestingly\(^1\), the same resonance condition can be obtained using classical mechanics. Starting again with a magnetic moment $\mu$ in an magnetic field $\mathbf{H}_0$, the torque $\mathbf{T}$ acting to align the moment with the magnetic field is \[^{29}\]

$$\mathbf{T} = \mu \times \mathbf{B} \quad (2.5)$$

Using Newton’s second law of motion for angular momentum $\frac{d\mathbf{L}}{dt} = \mathbf{T}$ and exploiting the relation between magnetic moment and angular momentum $\mu = -\gamma \mathbf{L}$, with the gyromagnetic ratio\(^2\) $\gamma = \frac{e \hbar}{2 m_e}$, \[^{2.5}\] can be rewritten to

$$\frac{d\mu}{dt} = -\gamma \mu \times \mu_0 \mathbf{H}_0 \quad (2.6)$$

This is the equation of motion of a magnetic moment in an external magnetic field.

The solution of Eq. \[^{2.6}\] yields a precessional movement, analogous to the motion of a spinning top in a gravity field (cf. Figure \[^{2.2}\] (b)). The precession frequency is called Larmor frequency, given by $\omega_L = \gamma \mu_0 H_0$ \[^{32}\]. Radiation with this frequency is absorbed, which leads to a resonant enhancement of the precession cone angle $\Theta$ and eventually to a complete spin reversal \[^{29}\]. Notice the similarity to the spin flip in the quantum mechanical picture. With the definition of $\gamma = g \mu_B / \hbar$, we also see that the resonance condition derived classically is the same as the condition $\hbar \omega = g \mu_B \mu_0 H_0$ \[^{2.4}\], derived quantum mechanically.

### 2.2.2 Ferromagnetic Resonance

Now we turn to the resonance condition in ferromagnetic systems. As the exchange interaction is large, all magnetic moments align in parallel and we can use a macro spin description for the magnetisation $\mathbf{M} = \sum_V \mu / V$, the sum of all magnetic moments $\mu$ divided by the sample volume $V$. In this limit of large quantum numbers, the previously derived classical description is well justified. However, in ferromagnetic samples, internal fields $\mathbf{H}_{\text{int}}$, such as crystalline anisotropy fields or demag-

\[^{1}\]In the introduction to his book on ferromagnetic resonance \[^{32}\], Vonsovskii stresses that this is not mere coincidence. As the $\hbar$’s can be eliminated from the quantum mechanical resonance condition, the correspondance principle suggests that the quantum mechanical result can be derived classically.

\[^{2}\]As we only consider electrons in this thesis, $\gamma$ is regarded positive and the minus sign of the negative electron charge is transferred to the equations.
netization fields, usually alter the external field $H_0$, giving rise to an effective field $H_{\text{eff}} = H_0 + H_{\text{int}}$. The crucial field is thus no longer $H_0$, but $H_{\text{eff}}$.

Another issue in the description of magnetic resonance in real material systems is the treatment of relaxation processes. While an isolated moment will precess infinitely around the field axis, a magnetic moment in a solid will interact with its environment, dispense energy to the lattice and relax to its equilibrium position parallel to the $H_{\text{eff}}$\(^3\). An appropriate relaxation term for ferromagnetic resonance was added to (2.6) by Landau and Lifshitz, so that the magnetisation dynamics of a ferromagnet can be described by the Landau-Lifshitz (LL) equation\(^{32}\):

$$\frac{dM}{dt} = -\gamma \left[ M \times \mu_0 H_{\text{eff}} \right] - \alpha \frac{\gamma}{M} \left[ M \times \left[ M \times \mu_0 H_{\text{eff}} \right] \right]$$

(2.7)

Here, $\gamma$ again is the gyromagnetic ratio and $\alpha$ represents a dimensionless damping parameter\(^4\). The LL equation conserves the magnitude of $M$, as the strong exchange interaction keeps the ferromagnetic moments aligned. Figure 2.2(c) depicts the precession of $M$ after its excitation, when the microwave radiation is turned off. The magnetisation precesses around $H_{\text{eff}}$, while the damping term, trying to align $M$ with $H_{\text{eff}}$, leads to a spiral movement towards the equilibrium position.

Under microwave excitation, the magnetic field component $h_{\text{rf}}$ excites $M$. The deflection angle $\Theta$ of $M$ from the equilibrium increases until the excitation force and damping force are balanced. $M$ precesses with constant cone angle $\Theta$, which is in the small angle regime, i.e., a few degrees at most.

### 2.2.2.1 Smit & Suhl Free Energy Ansatz

The fact that ferromagnetic resonance is a small angle precession permits an approach to determine the resonance condition using the free energy of the system. This method was first proposed by Smit and Suhl\(^{34,35}\). The basic idea is as follows:

As the Landau-Lifshitz equation conserves the magnitude of $M$, it is natural to rewrite it in spherical coordinates, hence eliminating the radial coordinate (cf. Ch. 3.3.1):

$$\dot{\theta} = \gamma \mu_0 \left( H_\varphi + \alpha H_\theta \right); \quad \dot{\varphi} \sin \theta_0 = -\gamma \mu_0 \left( H_\theta - \alpha H_\varphi \right)$$

(2.8)

with the spherical angles as defined in Figure 2.3 and $\theta_0$ as the azimuthal angle of the magnetisation’s equilibrium position. In this equilibrium position the magnetisation\(^3\) Except for thermodynamic excitations of spin waves.

\(^4\)The letter $\alpha$ is often also used as parameter of the damping term in the Landau-Lifshitz-Gilbert equation. Although it may be shown that these two damping parameters are related\(^{32}\), we may stress that $\alpha$ in this thesis describes the strength of the Landau-Lifshitz damping term.
Figure 2.3: Illustration of Smit and Suhl’s free energy approach. The magnetisation equilibrium orientation is determined by minimizing the free energy $F$. In the vicinity of this equilibrium position, the derivatives of $F$ give the effective fields $H_\varphi$ and $H_\theta$, which are used to solve the Landau-Lifshitz equation in spherical coordinates.

is parallel to the effective field and the transverse components $H_\varphi$ and $H_\theta$ vanish. Due to the excitation with microwave frequency, the magnetisation is deflected from its equilibrium and $H_\varphi$ and $H_\theta$ act as restoring forces. Their strength is determined by the slope of the free energy \[32\]:

$$
\mu_0 H_\theta = -\frac{1}{M} \frac{dF}{d\theta}, \quad \mu_0 H_\varphi = -\frac{1}{M \sin \vartheta_0} \frac{dF}{d\varphi},
$$

As mentioned, at the equilibrium position $\frac{dF}{d\varphi}$ and $\frac{dF}{d\theta}$ are zero, so Taylor expansion of $\frac{dF}{d\varphi}$ and $\frac{dF}{d\theta}$ determines the restoring force with respect to the displacement $\delta \varphi$ and $\delta \theta$. Plugging this, together with (2.9), into the equation of motion (2.8) results in a set of linear differential equations. An harmonic ansatz $\delta \varphi(t), \delta \theta(t) \propto \exp(i\omega t)$ solves these equations and yields the eigenfrequency $\omega_{\text{res}}$ as a function of the free energy derivatives at the equilibrium position \[32\]:

$$
\omega_{\text{res}} = \gamma (1 + \alpha^2) \frac{1}{2} \left( \frac{\partial^2 F}{\partial \varphi^2} \bigg|_{\varphi_0} + \frac{\partial^2 F}{\partial \theta^2} \bigg|_{\theta_0} - \left( \frac{\partial^2 F}{\partial \varphi \partial \theta} \bigg|_{\theta_0 \varphi_0} \right)^2 \right) \frac{1}{2}
$$

The absorption line width is given by

$$
\Delta \omega = \gamma \alpha \frac{1}{M} \left( \frac{\partial^2 F}{\partial \theta^2} \bigg|_{\theta_0} + \frac{1}{\sin^2 \theta_0} \frac{\partial^2 F}{\partial \varphi^2} \bigg|_{\varphi_0} \right).
$$

The advantage of the Smit $S$ Suhl concept is that (2.10) is applicable to a wide range of magnetic systems with different free energy expressions $F$. 
2.2.2.2 Polder Susceptibility Tensor

For a complete description of the ferromagnetic spin dynamics, the microwave magnetic field \( h_{rf} \) should be included into \( H_{\text{eff}} \) of the Landau-Lifshitz equation (2.7). Then it may be shown [36], that the magnetic response in small excitation fields, i.e., for small precession cone angles \( \Theta \), is determined by the Polder susceptibility tensor \( \hat{\chi} \):

\[
m(t) = \hat{\chi} h_{rf}(t),
\]

with \( m(t) \) as the time-dependent component of \( M \). \( \hat{\chi} \) is a complex tensor which fully describes the response of the magnetisation to the microwave excitation. The two eigenvectors of \( \hat{\chi} \) are circularly polarized microwave fields, rotating either in the sense marked by \( H_{\text{eff}} \) (Larmor excitation) or in the opposite sense (anti-Larmor excitation). For the Larmor precession, the complex eigenvalue \( \chi \) of \( \hat{\chi} \) is given by [33]:

\[
\chi(\omega) = \frac{\omega M (\omega_{\text{res}} - \omega - i\alpha \omega)}{(\omega_{\text{res}} - \omega)^2 + \alpha^2 \omega^2},
\]

where \( \omega_M = \gamma \mu_0 M \) and \( \omega_{\text{res}} \) is the resonance frequency, determined by the frequency of the eigenmotion of the unperturbed system, (2.10). As depicted in Figure 2.4, the real part of \( \chi \) shows a dispersive response, while the imaginary part of \( \chi \) possesses an absorption line shape. The full width at half maximum (FWHM) of the absorption, \( \Delta \omega \) is [33]:

\[
\Delta \omega = 2\alpha \omega_{\text{res}}
\]

Thus, the line width is proportional\(^5\) to the dimensionless damping parameter \( \alpha \) introduced in (2.7) and the frequency of the magnetisation motion \( \omega_{\text{res}} \). Therefore, the damping term is also referred to as \textit{viscous damping term}. The width of the resonance along the magnetic field \( \Delta H \) axis may be calculated from the width of

\(^5\)Often, viscous damping is not the only contribution to the line width. In most experiments a zero frequency offset to the line width is found upon plotting \( \Delta H \) against the resonance frequency.
the absorption in the frequency domain $\Delta \omega$ as \[33\]:

$$\Delta H = \frac{\partial H_0}{\partial \omega_{res}} \Delta \omega,$$ (2.15)

Using this, and an implicit relation $\omega_{res}/\gamma_{\text{eff}}(H_0) = \mu_0 H_0$ between external field and resonance frequency, where $\gamma_{\text{eff}}(H_0)$ is determined by (2.10), the susceptibility $\chi(\omega)$ may be expressed in terms of applied magnetic field $H_0$ as:

$$\chi(\mu_0 H_0) = \frac{\mu_0 M(\mu_0 H_{\text{res}} - \mu_0 H_0 - i \mu_0 \Delta H)}{(\mu_0 H_{\text{res}} - \mu_0 H_0)^2 + (\mu_0 \Delta H)^2}$$ (2.16)

### 2.2.3 Antiferromagnetic Resonance

The treatment of antiferromagnetic resonance (AFMR) is more subtle, as both sublattice magnetisations $M_1$ and $M_2$ have their own equation of motion, which are linked by the exchange interaction. The exchange interaction can be modelled by a Weiss molecular field $H_{\text{exch,1,2}} = \lambda M_{2,1}$. Additionally, as discussed in Chapter 2.1, the crystalline easy axis anisotropy plays an important role and can be modelled by an effective field $H_A$. Neglecting damping and following the theory of Keffer and Kittel [31], the equations of motion of the two sublattice magnetisations are:

$$\frac{dM_1}{dt} = \gamma \left[ M_1 \times \mu_0 H_{\text{eff}} \right] = \gamma \left[ M_1 \times \mu_0 \left( H_0 + H_A - \lambda M_2 \right) \right]$$ (2.17)

$$\frac{dM_2}{dt} = \gamma \left[ M_2 \times \mu_0 H_{\text{eff}} \right] = \gamma \left[ M_2 \times \mu_0 \left( H_0 - H_A - \lambda M_1 \right) \right]$$ (2.18)

The sign difference of the uniaxial anisotropy term is due to the fact that the anisotropy acts in positive z-direction for $M_1$ and in the opposite direction for $M_2$. Assuming both $H_0$ and $H_A$ parallel and in $\hat{z}$ direction, one obtains four resonance frequencies from (2.17) and (2.18) [37]:

$$\omega_{\text{res}} = \pm \gamma \mu_0 H_0 \pm \gamma \mu_0 \left[ H_A (2H_E + H_A) \right]^{\frac{1}{2}}$$ (2.19)

The relevant region of the field-frequency\(^6\) diagram (positive frequencies) is depicted in Figure 2.5. The most striking difference to ferromagnetic resonance is the zero field splitting. Even without an applied external field, there is a finite resonance frequency, due to the effective field of the exchange and anisotropy fields.

The motion of the magnetisation at zero external field is governed by four normal modes, which can be grouped into two modes as depicted in Figure 2.6. The

\(^6\)We use the term “magnetic field” rather imprecisely in this thesis for both the magnetic induction with unit (T) and the magnetic field intensity with unit (A/m).
Figure 2.5: The antiferromagnetic resonance dispersion for parallel alignment of external field and uniaxial anisotropy axis, as given by Kittel (2.19), using parameters characteristic for MnF$_2$ ($\mu_0 H_E = 53$ T, $\mu_0 H_A = 0.85$ T, cf. Tab. [3.1]). With increasing field, the degeneracy at zero field breaks up, resulting in a higher frequency branch (green) and an lower frequency branch (blue). Above the spin flop field $H_C \approx 9.5$ T (dashed vertical line), (2.19) is no longer applicable. Instead, the resonance frequency in the spin flop configuration is depicted in orange, as given by Nagamiya (2.20). Only positive frequencies are depicted.

magnetisations rotate around the axis of the effective field, which is the axis of uniaxial anisotropy for both magnetisations (since $H_0 = 0$). Looking along positive z direction, both magnetisations in (a) rotate either clockwise (first mode) or counter-clockwise (second mode). Figure 2.6 (b) depicts the other two modes with again both clockwise and counter-clockwise precession. The difference between the motions depicted in plane (a) and (b) is the cone angles ratio $\eta = \theta_2/\theta_1$, which is smaller than 1 for (a) and larger than 1 for (b).\footnote{\textit{\eta} is a material dependent parameter [31], given by $\eta = \left[H_A + H_E + (H_A (2H_E + H_A))^{1/2}\right] / H_E$}

An external field breaks the symmetry of the fundamental modes. The resonance frequency of one mode is increased by $\gamma \mu_0 H_0$, hence referred to as “higher frequency mode” throughout this thesis, while the other mode’s frequency is reduced by $\gamma \mu_0 H_0$ and named “lower frequency mode”. This can be explained in an intuitive picture as follows: In the higher frequency mode, the top magnetisation $M_1$, i.e. the one parallel to $H_0$, is “driving” the motion, while $M_2$ is rather just following the molecular field of $M_1$. For $M_1$ the effective field, composed of molecular and anisotropy fields is parallel to $H_0$. Hence, an increase of $H_0$ results in a higher effective field, which increases the resonance frequency of $M_1$ and hence the frequency of the whole
system is higher. In the lower frequency mode $M_2$ is leading the motion. However, the molecular and anisotropy fields of $M_2$ are antiparallel to $H_0$. An increase of $H_0$ thus leads to a decrease of the effective field of $M_2$ and the resonance frequency of the lower frequency mode.

Caution is required above the spin flop field $H_C$ (for MnF$_2$: $H_C \approx 9.5$ T). In the spin flop configuration the two magnetisation vectors are again degenerate with respect to the external field, meaning that their effective fields are the same. This should result in a single resonance line, not in two, as Kittel’s formula suggests. Nagamiya et al. [38] treated this problem, solved the LL equation for the magnetisations in spin flop configuration and find the antiferromagnetic resonance condition above the critical field as:

$$\omega_{\text{res}} = \pm \gamma \mu_0 \left[ H_0^2 - 2H_E H_A \right]^{1/2}, \quad H_0 > H_C, \quad \ldots \ldots (2.20)$$

which is depicted in Figure 2.5 as orange curve.

Hagiwara and co-workers showed [23], that all these resonance modes are indeed excited. They were able to measure the resonance frequencies of a MnF$_2$ crystal over wide regions, both in magnetic field and frequency, which is presented in Figure 2.7. The black dots show the data, the solid curve the fore-mentioned theory for the parallel configuration. Both, high and low resonance modes and the spin flop mode are observed. The theory describes the data well, except for the spin flop region. Here the data points deviate slightly from the theoretical curves and furthermore, the drop of the resonance frequency is not explained accurately by theory. As we will see
in Chapter 3.3 the alignment of external field and anisotropy is crucial for the AFMR dispersion, especially in the vicinity of \( H_C \). Indeed Kittel addressed this problem and extended his theory to arbitrary angles between \( H_0 \) and \( H_A \) \[31\], but its application is again restricted to small field strengths. Hence, no unified antiferromagnetic resonance theory exists to accurately describe the resonance frequencies of MnF\(_2\) over the whole magnetic field strength and especially within the spin flop region, with arbitrary alignment of \( H_0 \) with respect to \( H_A \).

### 2.3 Generation and Detection of Spin Currents

Now we turn from the discussion of localized spin moments to the influence of spin on the charge transport. As an electron carries not only charge, but also spin, it is possible to distinguish between conduction electrons in the spin up state (\( \uparrow \)) and electrons in the spin down state (\( \downarrow \)). These spin states possess individual carrier densities \( n_{\uparrow(\downarrow)} \). Their respective contribution to the total current density \( J_c = J_{\uparrow} + J_{\downarrow} \) is given by \( J_{\uparrow(\downarrow)} = -e n_{\uparrow(\downarrow)} v_{\uparrow(\downarrow)} \), where \( v_{\uparrow(\downarrow)} \) is the mean velocity of the electrons in the respective spin state. The motion of one spin species results in the transport of both charge and angular momentum. The transport of angular momentum is described by the spin current \( J_s \). The spin current of one conduction electron spin species \( J_{s,\uparrow(\downarrow)} \) is found by multiplying the charge current with the ratio of carried angular momentum to charge, \( J_{s,\uparrow(\downarrow)} = m_s \hbar / e J_{\uparrow(\downarrow)} \). For electrons \( m_s = \pm 1/2 \), which results in the total charge current \( J_c \) and total spin current \( J_s \) as:

\[
J_c = J_{\uparrow} + J_{\downarrow} \quad \quad \quad \quad \quad J_s = \frac{\hbar}{2e} (J_{\uparrow} - J_{\downarrow}) \quad \quad \quad \quad \quad (2.21)
\]
Figure 2.8: Illustration of spin dependent charge transport. (a) Equal spin-up and spin-down current densities result in a pure charge current. (b) Domination of one spin species results in both, charge and spin currents. (c) Opposing current directions of the spins cause a pure spin current. From [39].

Figure 2.8 illustrates the three possible current configurations. If both spin species have equal charge current contributions \( J_\uparrow = J_\downarrow \) (Fig. 2.8 (a)), no net angular momentum is transported and a pure charge current develops. This is the case in normal metals. If one spin configuration prevails (Fig. 2.8 (b)), i.e., its product of \( n \) and \( v \) exceeds the others, there is a transport of both charge and spin. This situation is called spin-polarized current and commonly found in ferromagnetic metals. A pure spin current is only established, if the current contributions of the two spin species are equally large, but oppositely directed (Fig. 2.8 (c)).

In the following we want to illuminate the different physical origins of \( J_c \) and \( J_s \). Charge currents are generated by a gradient of the electrochemical potential \( \mu = \mu_{\text{ch}} - e\Phi \), with chemical potential \( \mu_{\text{ch}} \) and the electric potential \( \Phi \). Extending this concept to the charge current contribution of the individual spin species, we may write:

\[
J_{\uparrow(\downarrow)} = \frac{\sigma}{e} \nabla \mu_{\uparrow(\downarrow)} = \frac{\sigma}{e} \left( \nabla \mu_{\text{ch},\uparrow(\downarrow)} - e \nabla \Phi \right),
\]

with spin-resolved chemical potential \( \mu_{\text{ch},\uparrow(\downarrow)} \) and obviously spin-independent electric potential \( \Phi \). The equation demonstrates the two sources of charge current, the diffusion and the drift current. The diffusion current is caused by a concentration difference and is proportional to \( \nabla \mu_{\text{ch}} \). The drift current is due to an electric field and proportional to \( \nabla \Phi = E \). Defining the spin accumulation \( \mu_s = \mu_\uparrow - \mu_\downarrow \) and using (2.22), (2.21) may then be written as:

\[
J_c = \frac{\sigma}{e} \nabla (\mu_\uparrow + \mu_\downarrow) \quad J_s = -\frac{\hbar \sigma}{2e^2} \nabla \mu_s
\]

Thus, the total charge current \( J_c \) originates from a gradient in the sum of the spin dependent electrochemical potentials \( \mu_\uparrow \) and \( \mu_\downarrow \), while a spin current \( J_s \) is caused by a gradient in the spin accumulation \( \mu_s \).
In the last decade, several methods for the generation of spin currents have been reported, among them the spin Hall effect \([40]\) and the spin Seebeck effect \([17]\). One of the most rigorously validated techniques for spin current generation is spin pumping \([41][19][20]\), which is presented in the following.

Sketching in Fig. 2.9, the typical spin pumping sample consists of a ferromagnet, covered on top by a metallic thin film with large spin orbit coupling. We will refer to this bilayer heterostructure as FM|N in the following. Ferromagnetic resonance (FMR) is excited in the FM by a microwave field \(h_{rf}\) and the magnetisation \(M(t)\) precesses with cone angle \(\Theta\) around its equilibrium position \(H_{eff}\), as discussed in Section 2.2.2. Compared with FMR in pure ferromagnetic samples, an additional damping mechanism \(D(t) \propto M(t) \times [M(t) \times H_{eff}]\) emerges, due to the presence of mobile spin carriers in the metallic layer. Electrons with spin direction \(D(t)\) are absorbed by the ferromagnet and due to conservation of angular momentum, lead to a relaxation of \(M(t)\) towards \(H_{eff}\). In the same way, electrons with spins opposite to \(D(t)\), are emitted towards the metallic layer. Due to conservation of charge, the
two current densities are equal in strength, but opposite in direction, leading to a
pure spin current $J_s$ crossing the FM|N interface. The spin direction of $J_s$ changes
with the direction of $D(t)$. As $D(t)$ is composed of a constant component along $H_{\text{eff}}$, indicated in red in Figure 2.9, and a purely time-varying component in the plane
perpendicular to $H_{\text{eff}}$, the spin-orientation of $J_s$ also incorporates both components.
In the following, we focus on the DC component of $J_s$ and denote the constant
component of the spin-orientation $\hat{s}$, with $\hat{s} \parallel H_{\text{eff}}$.

The strength of the spin current is determined by the ability of the FM|N interface
to transport spins. This ability is quantified by the spin mixing conductance $g_{\uparrow\downarrow}$,
which represents the number of transport channels per unit area for torque transport
[41]. $g_{\uparrow\downarrow}$ is a material-dependent interface parameter. The strength of the spin
pumping current is given by\footnote{Neglecting demagnetisation fields and under the assumption: $\text{Re}(g_{\uparrow\downarrow}) \gg \text{Im}(g_{\uparrow\downarrow})$. See \cite{39} for details.} [19]:

$$J_{s,\text{pump}} = \frac{1}{2\pi} \frac{\hbar}{2} \omega \text{Re}(g_{\uparrow\downarrow}) \sin^2 \Theta,$$  
(2.24)

where $\sin^2 \Theta$ is proportional to the area encircled by the magnetisation precession.
In resonance, $\Theta$ can be extracted from the microwave field strength $h_{\text{rf}}$ and the
resonance line width $\Delta H$ as $\Theta = h_{\text{rf}} / \Delta H$ \cite{14,39}. $h_{\text{rf}}$ is proportional to the root
of the microwave power $P_{\text{mw}}$, hence, using the small angle approximation: $J_{s,\text{pump}} \propto \sin^2 \Theta \propto h_{\text{rf}}^2 \propto P_{\text{mw}}$.

### 2.3.2 Spin Current Detection – The Spin Hall Effect and Its Inverse

It is not straightforward to measure spin currents directly—one can not buy a “spin
current ammeter”. Hence, an interaction mechanism to convert spin into charge
currents is required for detection of $J_s$. The conversion between the two kinds
of current is established via the spin orbit interaction (SOI). There are various
mechanism, all based on the SOI, which lead to an anisotropic scattering cross
section for the two spin species. In particular, extrinsic and intrinsic scattering
mechanisms are distinguished. The extrinsic mechanisms lead to a spin separation
by anisotropic scattering at impurities sites. Representatives are the skew \cite{13} and
side-jump \cite{14} scattering mechanisms. For the intrinsic mechanisms \cite{15}, the band
structure itself leads to an anisotropic velocity distribution, even in ideal crystals
\cite{16}.

All of them introduce an electric conductivity perpendicular to the applied cur-
cent, whose strength and direction depends on the orientation of the conduction
electron spin. Simply put, spin-up electrons are rather scattered to the “right” and
spin-down electrons to the “left”, generating a perpendicular spin current $J_s$. Sum-
maring the effects of intrinsic and extrinsic contributions into a single, material-
dependent charge-to-spin current conversion efficiency—the so called spin Hall angle $\alpha_{\text{SH}}$—, the conversion formulas between the two current types are written as [47]:

$$J_c^{\text{SH}} = \alpha_{\text{SH}} \left( \frac{2e}{h} \right) [J_s \times \hat{s}]$$

$$J_s^{\text{SH}} = \alpha_{\text{SH}} \left( \frac{h}{2e} \right) [J_c \times \hat{s}]$$

(2.25)

The superscripts introduce the names of these conversion effects, the spin Hall effect (SHE), converting a pure charge current to a perpendicular spin current and the inverse spin Hall effect (ISHE), converting a spin current to a charge current. The value of the dimensionless spin Hall angle, e.g. in platinum, differs significantly across the literature, with measurements ranging from $\alpha_{\text{SH,Pt}} \approx 4 \times 10^{-3}$ to $\alpha_{\text{SH,Pt}} \approx 8 \times 10^{-2}$ [48].

The Inverse Spin Hall Voltage Arising From Spin Pumping

In the spin pumping experiment sketched in Figure 2.9, the ISHE is used as detection mechanism. The spin current $J_s$ enters the normal metal and is converted to a perpendicular charge current $J_c$ by the inverse spin Hall effect. Due to the open circuit condition at the film boundary, the charges accumulate at the sides of the film, giving rise to a voltage $V_{\text{ISH}}$. The value of $V_{\text{ISH}}$ is given by [39]:

$$V_{\text{ISH}} = \frac{e}{\sigma_F t_F + \sigma_N t_N} \frac{\alpha_{\text{SH}} \lambda_{SD} \tanh \frac{2\pi}{2\lambda_{SD}} g_{\uparrow \downarrow}}{f_{\text{mw}} P L \sin^2 \Theta}$$

(2.26)

$\alpha_{\text{SH}}$ is the spin Hall angle, $t_{F/N}$ and $\sigma_{F/N}$ the thickness and conductivity of the ferromagnet (F) and normal metal (N), $f_{\text{mw}}$ the microwave frequency, $P$ a correction factor accounting for elliptical magnetisation precession [49], $L$ the distance between the contact probes of $V_{\text{ISH}}$ and $\Theta$ the cone angle of the precession. $\lambda_{SD}$ is the spin diffusion length in the metal, given by the spin flip time $\tau_{SF}$ and the diffusion constant $D$ as $\lambda_{SD} = \sqrt{D \tau_{SF}}$ [47].

Equation 2.26 has been rigorously tested for a variety of material systems in the work of Czeschka et al. [20, 39]. $V_{\text{ISH}}$ consistently describes the observed DC voltages for a variety of materials, including ferromagnetic metals, ferromagnetic semiconductors and ferromagnetic insulators.

A characteristic of $V_{\text{ISH}}$ is its sign inversion under reversal of the external magnetic field. As $H_0 \rightarrow -H_0$, the magnetisation and hence the spin polarisation of the spin current changes sign, too: $\hat{s} \rightarrow -\hat{s}$. The vector product of the ISHE, (2.25), thus leads to an opposite charge current direction, resulting in a sign inversion of $V_{\text{ISH}}$. 
2.4 The Spin Hall Magnetoresistance

With the spin Hall effect (SHE) and the inverse spin Hall effect (ISHE) in normal metals, spin currents can be converted into charge currents and vice versa. If both effects are combined, a spin current, arising from an initial charge current via the SHE may have an influence on its source, via a second conversion using the ISHE. In this section, a recently discovered new type of magnetoresistance [50], based on this phenomenon, is presented: the spin Hall magnetoresistance (SMR).

Figure 2.10 (a) sketches the spin transport in a metal (N) with finite spin Hall angle $\alpha_{SH}$, such as Pt. The metallic layer is assumed a thin film, stretching in the plane of $j$ and $t$. The incident charge current $J_{c,\text{inc}}$ leads to a perpendicular spin current $J_s^{\text{SHE}}$ along $-n$ due to the spin Hall effect\(^9\) (2.23). In the case of open circuit conditions, the spins accumulate at the left and right film sides, which gives rise to a spin concentration difference across the sample. The resulting gradient in spin accumulation $\nabla \mu_s$ causes a diffusion spin current $J_{s,\text{diff}}$ (2.23). Due to the inverse spin Hall effect, a part of $J_{s,\text{diff}}$ is reconverted to a charge current $J_c^{\text{ISHE}}$.

\(^9\)Note, that furthermore a spin current arises along $t$, which is omitted, as it does not contribute to the SMR effect.
which is directed in the same sense as \( \mathbf{J}_{\text{c,inc}} \). Hence, the net charge current for a given electric field strength is the sum of \( \mathbf{J}_{\text{c,ISHE}} \) and the part of \( \mathbf{J}_{\text{c,inc}} \) that was not converted to a perpendicular spin current. Charge currents in metals with SOI are inevitably composed of both contributions.

The open circuit condition for \( \mathbf{J}_{\text{s,SHE}} \) at the film sides may be affected by a ferromagnetic material (FM), attached to these boundaries, as illustrated in Figure 2.10 (b) in blue for the left interface of the Pt film. In this case, a part of \( \mathbf{J}_{\text{s,SHE}} \) can penetrate into the ferromagnet, where it is absorbed. We denote the spin current crossing the FM|N interface \( \mathbf{J}_{\text{s,STT}} \), as spin transfer torque (STT) is the driving mechanism. STT is treated in more detail below, for now it should only be noted that STT introduces additional spin flip scattering at the the FM|N interface. This reduces the spin accumulation of the interface, leading to a decrease of \( \nabla \mu_s \) and \( \mathbf{J}_{\text{s,diff}} \). The charge current \( \mathbf{J}_{\text{c,ISHE}} \) converted back from the spin diffusion current diminishes, which results in a smaller net charge current \( \mathbf{J}_c \) for a given electric field strength. This means, that the resistance increases, if the spin current \( \mathbf{J}_{\text{s,SHE}} \) is absorbed at the FM|N interface. Thus, the sample resistance is modified by a variation of the spin current boundary condition.

Interestingly, the use of an ferromagnet enables a continuous variation of the boundary condition. STT describes the reverse process to spin pumping, namely an absorption of spin current at a FM|N interface by an excitation and deflection of \( \mathbf{M} \). It was shown by Brataas et al. [41], that \( \mathbf{M} \) may only be excited if the spin polarisation of \( \mathbf{J}_{\text{s,STT}} \) is perpendicular to \( \mathbf{M} \). In the parallel configuration, an excitation is not possible and the spin current may not cross the FM|N interface. By rotating \( \mathbf{m} = \mathbf{M}/M \) from \( \mathbf{m} \parallel \mathbf{s} \) to \( \mathbf{m} \perp \mathbf{s} \), the STT mechanism at the boundary may therefore be activated, which opens the FM|N interface for \( \mathbf{J}_{\text{s,STT}} \), and thereby increases the longitudinal resistivity \( \rho_{\text{long}} \). Thus, the longitudinal sheet resistivity is larger if the STT is activated, occuring for \( \mathbf{m} \perp \mathbf{s} \) [51]:

\[
\rho_{\text{long, m \perp s}} > \rho_{\text{long, m \parallel s}}. \tag{2.27}
\]

For intermediate angles between \( \mathbf{m} \) and \( \mathbf{s} \) also a perpendicular “planar Hall-type” resistivity \( \rho_{\text{trans}} \) arises, as shown in the following. As the component of \( \mathbf{J}_{\text{s,SHE}} \) with spin orientation perpendicular to \( \mathbf{m} \) is absorbed within the FM, the component of \( \mathbf{J}_{\text{s,SHE}} \) with parallel spin orientation stays within N, leading to a spin accumulation \( \mu_s \) with spin polarisation parallel to \( \mathbf{m} \) at the interface. For a general orientation of \( \mathbf{M} \) within the film plane, this spin polarisation \( \mathbf{s} \parallel \mathbf{m} \) possesses components both along \( \mathbf{j} \) and \( \mathbf{t} \). A diffusion current sets in, wherein the part of \( \mathbf{J}_{\text{s,diff}} \) with spin orientation along \( \mathbf{t} \) results in the previously discussed charge current \( \mathbf{J}_{\text{c,ISHE}} \). The \( \mathbf{j} \) component of the spin polarisation, however, leads to a charge current along \( \mathbf{t} \), giving rise to a transverse in-plane resistivity \( \rho_{\text{trans}} \). A detailed analysis of the invoked vector products of the charge conversions and the STT gives an expression for the angular
dependence of $\mathbf{m}$ on the SMR resistivity \[53\]:

$$
\rho_{\text{long}} = \rho_0 - \Delta \rho m_i^2 
$$

(2.28)

$$
\rho_{\text{trans}} = \Delta \rho m_t m_j
$$

(2.29)

Here $m_j, m_t$ are the projections of $\mathbf{m}$ onto the incident current direction $\mathbf{j}$ and its transverse in plane component $\mathbf{t}$. $\rho_0$ is the magnetisation independent part of the $\rho_{\text{long}}$ and $\Delta \rho$ gives the strength of the SMR effect.

The SMR effect is best observed in isolating ferromagnet|metal heterostructures. With metallic ferromagnets, the SMR effect is superimposed by the anisotropic magneto resistance (AMR) \[54, 55\] that the charge current flowing in the ferromagnetic layer experiences. With Pt|YIG bilayers, SMR effects of $\Delta \rho/\rho_{\text{long}} \sim 10^{-3} - 10^{-4}$ are observable. The effect size depends thereby strongly on the Pt thickness $t_{\text{Pt}}$ with a maximum effect for $t_{\text{Pt}}$ in the 5 nm range \[56\], which is from theory twice the diffusion length $\lambda_{\text{SD}}$ \[50\].
Chapter 3
Antiferromagnetic Resonance of MnF$_2$

Spin current generation with spin pumping bases on the resonant excitation of exchange coupled magnetic systems. To gain a thorough understanding of the magnetic resonance in our model antiferromagnet MnF$_2$, we measure the antiferromagnetic resonance (AFMR) dispersion for magnetic field strengths in the vicinity of the spin flop field $8.5 \, \text{T} \lesssim H_0 \lesssim 10.5 \, \text{T}$ and for microwave frequencies up to 26.5 GHz.

In the following, the experimental setup is outlined first. Then, in Section 3.2, a typical AFMR measurement result is discussed, which reveals the necessity of a more elaborate AFMR theory. This is developed as a numerical AFMR simulation in Section 3.3, followed by a comparison of simulated and measured AFMR in Section 3.4. Finally, we present the temperature dependence of the AFMR in MnF$_2$ in Section 3.5.

3.1 Experimental Methods

This section discusses the main components of the AFMR measurement setup. First the magnet flow cryostat, supplying the required low temperatures and high magnetic field strengths is presented. Then the MnF$_2$ crystal mounting on the coplanar wave guide are discussed. The section ends with an introduction to the functional principle of vector network analyzer and a discussion of possible analysis methods to relate the measured $S$-parameters to physical quantities of the specimen under investigation.

3.1.1 Magnet Flow Cryostat

The antiferromagnetic resonance experiments were conducted in a commercial liquid Helium cryostat with a superconducting solenoid magnet specified to a maximum field strength of $\mu_0 H_0 = 17 \, \text{T}$. A schematic of the cryostat is depicted in Figure 3.1. The dewar contains liquid Helium-4, which cools the coils of the superconducting solenoid magnet below their critical temperature. Additionally, the He provides cooling power to control the sample temperature. The sample resides at the bottom of the dip stick, which is inserted into a variable temperature inset (VTI). The sample
space of the VTI is continuously pumped, thereby evaporating liquid Helium inside the capillary connecting the VTI with the helium bath. The evaporation takes place at mbar pressures, reducing the He vapor pressure which enables the access to temperatures below 4.2 K. The sample is situated right above the capillary to achieve maximum cooling power by the escaping He gas. In order to provide higher temperatures, a heater and a needle valve are located at the capillary, allowing to temper the gas and control the He gas flux strength. In this way, the VTI makes it possible to adjust the sample temperature continuously from $\sim 2$ K to 300 K.

### 3.1.2 Coplanar Wave Guide and Sample Mounting

The dip stick (cf. Figure 3.1) is equipped with stainless-steel\(^1\) microwave cables, at whose ends the coplanar wave guide (CPW), shown in Figure 3.2 (a), is attached via SMA to Mini-SMP adapters. The cross section of the CPW is illustrated in Figure 3.2 (b). A CPW consists of a center conductor, carrying the AC current and two ground planes, separated by an air gap. In general, a CPW has a complex electromagnetic field distribution \([57][58]\), which is here simplified to its main property: The magnetic microwave field $h_{rf}$ exhibits circular field lines around the center conductor. The utilized CPWs were designed by Martin Radlmeier, in the course of his diploma thesis \([59]\). The length of the center conductor is 12 mm and the total diameter of the CPW is 22 mm.

For the antiferromagnetic resonance experiments, three nominally identical MnF\(_2\) crystals were acquired\(^2\), subsequently denoted as $MnF_2\#1$, \#2 and \#3. The crystals

---

\(^1\)To reduce temperature insertion from the connector stage at room temperature.
\(^2\)Retailer: MaTecK GmbH, Juelich, Germany.
are of cuboid shape with a size of $3\,\text{mm} \times 1\,\text{mm} \times 0.5\,\text{mm}$ and the axis of uniaxial anisotropy is oriented perpendicular to the large face.

As depicted in Figures 3.2 (a) and (b), the crystal is placed on top of the center conductor such that the crystalline anisotropy axis $H_A$ is parallel with the external field $H_0$. Then it is fixed to this position using rubber cement. The misalignment of $H_A$ and $H_0$ in the experiment is denoted $\theta_H$.

To excite resonance, $h_{rf}$ must be perpendicular to the magnetisation vectors of the AFMR (cf. Sec. 2.2.1). For $H_0 < H_C$, the magnetisations align in parallel with $H_A$ and the resonance condition is fulfilled at the center of the sample, above the center conductor. In the spin flop configuration $H_0 > H_C$, the magnetisations orient perpendicular to $H_A$, and lie within the plane of the CPW. Then, the condition is fulfilled at the edges of the center conductor and above the gap between center conductor and ground plane. In summary, we expect the observation of antiferromagnetic resonance both below and above the spin flop field.

### 3.1.3 Vector Network Analyzer

The Vector Network Analyzer (VNA) serves in our AFMR measurement as microwave source and microwave detector. It is developed as a tool to characterize microwave frequency components and may thus measure electric quantities like microwave absorption, gain and phase relationships with high accuracy. In the follow-
Figure 3.3: Illustration of a Vector Network Analyzer (VNA) in the configuration of a two port reflection and transmission experiment of the device under test (DUT). The incident wave, generated by the microwave source, leaves the VNA at port 1 with voltage $V_{1,\text{out}}$, interacts with the DUT and creates both reflected ($V_{1,\text{in}}$) and transmitted ($V_{2,\text{in}}$) waves. By means of directional couplers, these are guided towards the detection stage and measured with respect to a reference of the incident wave ($V_r$). During the measurement, the source of port 2 is terminated with the characteristic impedance $Z_0$ to prevent reflected waves from this port from perturbing the measurement.

For further information, see [60].

For further information, see [60].
3.1 Experimental Methods

Microwave detection is difficult, as every connected voltmeter or current probe introduces unmatched impedances, which would alter the measurement results. The detection method of choice to minimize those problems is a tuned receiver. The detection microwave signal is mixed with the frequency of a local oscillator, whose frequency \( f_{\text{LO}} \) is slightly detuned from the source frequency \( f_{\text{mw}} \). The resulting, down-mixed intermediate frequency conserves the voltage and phase information and is more suitable for detection, being in the megahertz regime. It is further filtered by a bandpass filter, to improve the detection bandwidth and then fed into an analog-to-digital converter (ADC) and a digital-signal processing unit (DSP) \[61\].

The VNA measures scattering parameters or \( S \)-parameters \( S_{ij} \), which are defined as:

\[
S_{ij} = \frac{V_{i,\text{in}}}{V_{j,\text{out}}} \bigg|_{V_{k,\text{out}}=0, k \neq j} \tag{3.1}
\]

They are understood as follows: Port \( j \) is driven and emits an outgoing wave with voltage \( V_{j,\text{out}} \). At the DUT, the wave is scattered towards port \( i \), where the incoming wave possesses a voltage \( V_{i,\text{in}} \). The S-parameter is the ratio of these two voltages. During the measurement, all ports \( k \) except for the source port are terminated with a perfect load \( Z_0 \), as indicated in the top right of Figure 3.3. The termination with the characteristic impedance\(^4 \) \( Z_0 \) prevents the reflection of microwaves at this port, which would perturb the measurement.

The \( S \)-parameters are complex quantities and contain the phase information between the source and the measured wave. For a two port VNA, \( S_{11} \) and \( S_{22} \) are reflection coefficients, while \( S_{12} \) and \( S_{21} \) are the transmission coefficients.

3.1.3.1 Relating \( S \)-Parameter and Susceptibility

The physical quantity of interest in a magnetic resonance experiment is the complex dynamic susceptibility \( \chi = \chi' + i\chi'' \), containing the magnetic response to a given microwave excitation. \( \chi \) determines the position and the line width of the resonance, which are related to fundamental physical quantities like internal fields (Eq. 2.13) and the damping parameter \( \alpha \) (Eq. 2.14). The correct extraction of \( \chi \) from the measured \( S \)-parameter is, however, difficult and requires careful calibration and data analysis. In the following, two different types of resonance analysis methods are presented and their differences are discussed.

Analysis based on Microwave Power Absorption

The transmission parameter \( S_{21} \) relates the voltages of the microwave before and after its interaction with the magnetic sample. The transmitted power ratio is given

\(^4\)In electric circuits, the best microwave power transfer is achieved, if the components of the circuits have the same input impedance. Therefore, standard input impedances for microwave components have been defined—the characteristic impedance \( Z_0 \)— which is either 50\( \Omega \) or 75\( \Omega \).
by the squares of the microwave voltages, therefore by $|S_{21}|^2$. The absorbed power
$P_{\text{abs}}$, neglecting microwave reflection at the sample, is given as
\[ P_{\text{abs}} = (1 - |S_{21}|^2) P_{\text{mw}}, \quad (3.2) \]

with $P_{\text{mw}}$ as microwave source power. To extract the power absorbed by the magnetic sample, either a good calibration or a reference measurement is required. A simple method to exclude the absorption not stemming from the magnetic resonance, is the subtraction of a reference value $|S_{21}|^2_{\text{ref}}$ at a magnetic field strength for which no magnetic resonance is excited. The absorption caused only by the magnetic sample is thus written as:
\[ P_{\text{abs},\text{FMR}} = (|S_{21}|^2 - |S_{21}|^2_{\text{ref}}) P_{\text{mw}}. \quad (3.3) \]

On the other hand, the Poynting theorem states that the absorbed power of the magnetic system is given by the time derivative of the electromagnetic energy density $u$, integrated over the sample volume $V_s$.
\[ P_{\text{abs},\text{FMR}} = \text{Re} \int_{V_s} \partial_t u \, dV = \text{Re} \int_{V_s} \partial_t \frac{1}{4} (\mathbf{e} \cdot \mathbf{d}^* + \mathbf{b} \cdot \mathbf{h}^*) \, dV \quad (3.4) \]

$b$, $h$, $e$ and $d$ are the magnetic flux density, the magnetic and electric field and the electric displacement field. All fields are assumed to have a time dependency $\propto \exp(-i\omega t)$. Restricting to the magnetic contribution of the absorption and evaluating the time derivative $\partial_t \mathbf{b} \cdot \mathbf{h}^* = 2i\omega \mathbf{b} \cdot \mathbf{h}^*$ gives the microwave absorption of the magnetic sample as:
\[ P_{\text{abs},\text{FMR}} = \frac{1}{2} \text{Re} \left[ \int_{V_s} -i\omega \mathbf{b} \cdot \mathbf{h}^* \, dV \right] \quad (3.5) \]
\[ = \frac{1}{2} \text{Re} \left[ \int_{V_s} -i\omega \mu_0 \mu \mathbf{b} \cdot \mathbf{h}^* \, dV \right] \quad (3.6) \]
\[ = \frac{1}{2} \text{Re} \left[ \int_{V_s} -i\omega \mu_0 (1 + \chi) |\mathbf{h}|^2 \, dV \right] \quad (3.7) \]
\[ = \frac{1}{2} \int \omega \chi'' |\mathbf{h}|^2 \, dV \quad (3.8) \]

Here, $\mu = (1 + \chi)$ is the permeability and $\chi = \chi' + i\chi''$ the dynamic susceptibility. The absorbed microwave power increases with the volume of the magnetic sample and is directly proportional to the imaginary part of the magnetic susceptibility.

\[ ^5 \text{The tensor property of } \mu, \text{ expressed by the Polder permeability tensor } \hat{\mu}, \text{ is neglected in this derivation for simplicity.} \]
Combining this with the relationship between $P_{\text{abs,FMR}}$ and $S_{21}$ (3.3) gives:

$$\chi'' \propto |S_{21}|^2 - |S_{21}|_{\text{ref}}^2$$

This equation states, that the line shape of $|S_{21}|^2$ should follow the Lorentzian shape of $\chi''$, as given by (2.13). Therefore, the extracted resonance position and line width from $|S_{21}|^2$ should in principle correspond to the same quantities in $\chi$, which are related to the physical origins of the dispersion and the damping.

Bear in mind, though, that microwave reflection at the magnetic sample was neglected in the derivation of (3.9). When passing through the magnetic resonance, the microwave reflection is most likely affected to a certain degree. This modifies the Lorentzian form of the absorption $|S_{21}|^2$ and decreases the accuracy of the analysis based on the microwave power absorption [63].

**Analysis based on the Impedance Shift**

A more sophisticated analysis exploits the change of impedance of the CPW section loaded with the sample, when the sample is excited resonantly. In resonance, the permeability of the sample changes, which affects the wave impedance of the CPW in this section. This introduces additional reflections, absorption and phase shifts. Bilzer et al. [63] present a detailed analysis to extract $\chi$ from all four reflection and transmission $S$-parameters of an two port VNA. The analysis requires careful calibration of the feed lines and connectors, including the connector to the CPW at the respective measurement temperature. Such a careful calibration is not possible with the used CPW design and cryostat.

A different approach based on the impedance shift was reported by Kalarickal et al. [64]. Based on a transmission line model [65] and neglecting microwave reflection, they introduce a quantity $U$, which is related, but not strictly equal, to the actual complex microwave permeability $\mu$. This parameter is calculated solely from $S_{21}$ and a reference $S_{21,\text{ref}}$ at a magnetic field strength without magnetic resonance:

$$U = -\frac{i \ln [S_{21} / S_{21,\text{ref}}]}{\ln |S_{21,\text{ref}}|}$$

The calculated $U$ contains the complex $S_{21}$ values. As stated, the line shape of $U$ reflects the permeability of the sample, thus under ideal circumstances Im[$U$] contains the absorption profile, while Re[$U$] shows the dispersion.

The $U$ analysis is clearly less complex than the accurate analysis of Bilzer [63]. Nevertheless, the extracted resonance positions and line widths are compatible: The extracted resonance position of $U$ deviate from the full two port analysis less than 1% in [63]. A similar resonance position accuracy is found for the analysis based on the power absorption $|S_{21}|^2$. The extracted line widths of both analysis methods have a relative error of less than 10% compared with the full two port analysis. The
3.2 Experimental Results

A typical result of the antiferromagnetic resonance of MnF$_2$ at $T = 4$ K, based on the microwave absorption analysis is presented in Figure 3.4. Depicted in false color as a function of microwave frequency and magnetic field is the absorption $|S_{21}|^2$ of the MnF$_2$ crystal. Due to imperfect impedance matching of microwave cables, CPW and their interconnections, the measured transmission parameter $S_{21}$ depends
heavily on the frequency and in general drops towards higher frequencies. Therefore, as discussed in Sec. 3.1.3.1, a reference at 8.3 T is subtracted as $|S_{21}|^2 - |S_{21}|^2_{8.3 T}$. Due to the frequency limitations of the VNA we are restricted to frequencies below 26.5 GHz, which allows only the observation of the lower of the two AFMR resonance frequencies (cf. Fig. 2.5). The measurement was performed with a microwave source power of $P_{mw} = 5 \text{ dBm}$.

The resonance line enters the measurement window at 8.6 T as nearly straight line with negative slope. At the spin flop field, at $\sim 9.5$ T, the resonance reaches a minimal value and then either disappears to the top of the measurement window (cf. Fig. 3.5(a)), or, as is the case in this measurement, bends again at higher magnetic field strength and vanishes. Under field inversion, as shown in the inset of Figure 3.4, the dispersion of MnF$_2$ is unaltered.

Figure 3.5 shows a different measurement, with higher magnetic field resolution. Figure 3.5 (a) depicts the resonance absorption of MnF$_2$. Again the non-MnF$_2$-dependent absorption is subtracted by a reference at 8.75 T. Cuts around 24.5 GHz (indicated in red) are depicted in Graph 3.5 (b) for three adjacent frequencies. The two absorption dips are visible at $\sim 9.1$ T and $\sim 9.8$ T. The line shape resembles a Lorentzian for the cut at $f_{mw} = 24.504 \text{GHz}$ (orange), but exhibits dispersion shaped features at specific frequencies (blue cut at 24.500 GHz). These deviations are attributed to the omission of microwave reflection in the absorption based $S_{21}$ analysis, see Section 3.1.3.1. The zoom-ins—Figures 3.5 (c) and (d)—reveal that the resonances consist of several resonances modes, which has been observed by Kotthaus & Jaccarino [25], too, and is attributed to the excitation of spin wave modes. Unlike the uniform resonance mode, which is a coherent precession of all magnetic moments with no phase difference, the precession of neighbouring moments in the spin wave mode possesses a phase difference. The resonance frequencies of distinct spin wave modes are shifted with respect to the uniform mode by offsets proportional to $(n/d)^2$ [36], where $n$ is an integer and $d$, the sample thickness. As $d$ is comparatively large for bulk crystals, the frequency gap between distinct spin wave modes is rather small, as observed in Figure 3.5 (d). For a quantitative analysis of the spin wave spectrum of MnF$_2$, refer to [66].

When comparing Figure 3.5 (a) with the result of the Figure 3.4, the difference between the two observed AFMR dispersions is striking. Although the measurement setup is the same and the mounting process was made as reproducibly as possible, the position of the resonance lines differ significantly. While the resonance of Figure 3.4 reaches its minimal frequency value at $\sim 7.5$ GHz, the resonance of Figure 3.5 dips around 22.5 GHz.

The observed difference is due to the high sensitivity of AFMR to the alignment of the external field with respect to the axis of uniaxial anisotropy. Kittel’s resonance condition for arbitrary angles [31], states that a good alignment of external field with the axis of uniaxial anisotropy results in a reduction of the lower resonance.
Figure 3.5: AFMR measurement with high magnetic field resolution at $T = 4\,\text{K}$ with $P_{\text{mw}} = 0\,\text{dBm}$. The AFMR resonance (a) is calculated in the same way as Fig. 3.4 and uses the same color code. The horizontal red line marks the cut, shown in (b). Here, the absorption profiles of three adjacent frequency lines are depicted. The zoom-ins (c) and (d) reveal multiple resonance lines, attributed to the excitation of spin waves. While in (a), similar to Fig. 3.4, a reference line at 8.75 T is subtracted from the absorption magnitude $|S_{21}|^2$, (b)-(d) display the unprocessed magnitude.
modes frequency. Putting it the other way around: A misalignment of a few degree is enough to push the AFMR dispersion out of the available measurement window. As stated in Chapter 2.2.3, Kittel’s theory is only applicable in small fields. There is no analytical theory available to describe the antiferromagnetic resonance absorption in the spin flop region with arbitrary angles between external field and uniaxial anisotropy. However, we were able to develop a numerical simulation scheme to accurately determine the resonance dispersion for arbitrary alignment of the external field. The derivation and the results of this AFMR simulation are presented in the following section.

3.3 Antiferromagnetic Resonance Simulation

The simulation of antiferromagnetic resonance follows the concept of Smit & Suhl, presented in Chapter 2.2.2.1, and is now extended to two exchange coupled magnetisation vectors. First, the equations of motion for the two magnetisation vectors $M_i$ are converted to a system of linear equations for the spherical angles of $M_i$, describing the eigenmotion of the magnetisation vectors around their equilibrium position for a given shape of total free energy $F$. The steps to reach this system of equations are:

1. The LL equations are converted to polar coordinates.

2. The total free energy is assumed as $F = F_{\text{exchange}} + F_{\text{Zeeman}} + F_{\text{uniaxial}}$

3. The effective fields acting on the magnetisation vectors are inserted into the LL equation as a Taylor expansion of the total free energy derivatives in the vicinity of the equilibrium position. This results in a linear differential equation for $\delta \theta_i$ and $\delta \varphi_i$ and their time derivatives.

4. Assuming periodic solutions $\delta \theta_i$ and $\delta \varphi_i \propto \exp(i \omega t)$, the differential equation transforms to a linear equation system of the form $0 = P(\omega) \begin{pmatrix} \delta \theta_1 \\ \delta \varphi_1 \\ \delta \theta_2 \\ \delta \varphi_2 \end{pmatrix}$, where $P(\omega)$ is a matrix composed of the second derivatives of $F$ at the equilibrium orientations of $M_i$.

To determine the eigenfrequencies of the motion, which depend on the strength and orientation of the external field, the following steps are required at each magnetic field magnitude.

1. The equilibrium orientations of the magnetisation vectors are found by minimizing the total free energy $F_{\text{tot}}$. 


2. The free energy derivatives at the equilibrium orientations are calculated and inserted into $P(\omega)$, which gives a solvable linear system of equations.

3. $\det P(\omega) = 0$ determines the four complex eigenvalues $\omega$. The resonance position is given by $\omega_{\text{res}} = \text{Re}(\omega)$ and the line width by $\Delta \omega = 2 \cdot \text{Im}(\omega)$.

MATLAB is used as computation environment for the implementation of the AFMR simulation. MATLAB is able to combine the flexibility and speed of a numerical computation (crucial for the computation of the equilibrium orientation of $\mathbf{M}_i$ in different field configurations) with the accuracy of analytical expressions\(^6\) (for the free energy derivatives).

In the following, the steps to achieve the linear equation system and the expressions for $F_{\text{tot}}$ and $P(\omega)$, as implemented in the AFMR computation scheme, are presented.

### 3.3.1 Details of the AFMR simulation

The derivation follows the book of Vonsowskii on ferromagnetic resonance\(^3\), Chapter I§3 and I§10 and is adapted to the situation of two exchange coupled magnetisation vectors in antiferromagnes. Figure 3.6 depicts the field configuration of the antiferromagnet, with the orientation of the external field $\mathbf{H}_0$, the anisotropy field\(^7\) $\mathbf{H}_A$ and the molecular field $\lambda \mathbf{M}_i$. Additionally, the definition of the spherical angles $\varphi_i$ and $\theta_i$ is introduced, which follows the standard notation. Furthermore, we denote the azimuthal angle of $\mathbf{H}_0$, which is the misalignment of $\mathbf{H}_0$ with respect to $\mathbf{H}_A$, as $\theta_H$.

In the following, equations with subscripts $i$ are valid for both sublattice magnetisations with $j$ denoting the respective other magnetisations index. The magnetisation magnitude of both sublattices are equal and denoted $M = |\mathbf{M}_1| = |\mathbf{M}_2|$

#### LL equation in polar coordinates

In the first step, the LL equations (2.7) for the sublattice magnetisations are transformed to spherical coordinates. The LL equation for the individual magnetisations $\mathbf{M}_i$ reads as:

$$\frac{d\mathbf{M}_i}{dt} = -\gamma \left[ \mathbf{M}_i \times \mu_0 \mathbf{H}_{\text{eff},i} \right] - \alpha \frac{\gamma}{M} \left[ \mathbf{M}_i \times \left[ \mathbf{M}_i \times \mu_0 \mathbf{H}_{\text{eff},i} \right] \right]$$

(3.11)

The time derivative of $\mathbf{M}_i = M \mathbf{e}_{r,i}$ in polar coordinates is given by:

$$\dot{\mathbf{M}}_i = M \dot{\mathbf{e}}_{r,i} + M \dot{\theta}_i \mathbf{e}_{\theta,i} + M \dot{\varphi}_i \sin \vartheta_i \mathbf{e}_{\varphi,i}$$

(3.12)

\(^6\)In order to handle analytical expressions, MATLAB’s Symbolic Math Toolbox is required.

\(^7\)The effective field concept is only used for the illustration of the direction of the uniaxial anisotropy. The simulation uses the more accurate free energy expression.
Figure 3.6: Illustration of the antiferromagnetic sublattice magnetisations $M_1$ and $M_2$ in green and the important contributions to their respective effective fields: The external field $H_0$ in blue, the uniaxial anisotropy $H_A$ in red and the molecular fields $-\lambda M_i$. The definition of spherical angles $\varphi_i$ and $\theta_i$ follow the usual spherical coordinates notation, as depicted for $M_2$. $M_1, M_2$ lie within the plane defined by $H_0$ and $H_A$. The configuration depicts the magnetisation’s equilibrium positions in an external field just below $H_C$. $\theta_H$ denotes the misalignment of $H_0$ with respect to $H_A$.

The crossproducts of $M_i$ with $H_{\text{eff},i} = H_\phi \hat{e}_{\phi,i} + H_{\theta,i} \hat{e}_{\theta,i} + H_{\varphi,i} \hat{e}_{\varphi,i}$ evaluate to:

$$M_i \times H_{\text{eff},i} = 0 \hat{e}_{r,i} - MH_{\varphi,i} \hat{e}_{\theta,i} + MH_{\theta,i} \hat{e}_{\varphi,i}$$  \hspace{1cm} (3.13)

$$M_i \times [M_i \times H_{\text{eff},i}] = 0 \hat{e}_{r,i} - M^2 H_{\theta,i} \hat{e}_{\theta,i} - M^2 H_{\phi,i} \hat{e}_{\varphi,i}$$  \hspace{1cm} (3.14)

Insertion of (3.12) – (3.14) into (3.11) yields the LL equation of the sublattice magnetisations in polar coordinates. Aside from the trivial radial equation $\dot{M} = 0$, stating the conservation of magnetisation magnitude by the LL equation, the LL equations for the azimuthal and polar coordinate read:

$$\dot{\theta}_i = \gamma \mu_0 (H_{\varphi,i} + \alpha H_{\theta,i})$$

$$\dot{\varphi}_i \sin \theta_i = -\gamma \mu_0 (H_{\theta,i} - \alpha H_{\phi,i})$$  \hspace{1cm} (3.15)

Free energy and the effective field

The effective field is related to the free energy of the system by

$$\mu_0 H_{\text{eff}} = -\frac{dF_{\text{tot}}}{dM}.$$  \hspace{1cm} (3.16)

With the definition of the derivatives in spherical coordinates this yields for the spherical components of the effective field of sublattice $i$, $H_{\theta,i}$ and $H_{\varphi,i}$:

$$\mu_0 H_{\theta,i} = - \frac{F_{\theta,i}}{M}$$

$$\mu_0 H_{\varphi,i} = - \frac{F_{\varphi,i}}{M \sin \theta_{i,0}}.$$  \hspace{1cm} (3.16)

\(^8\)Note: Vonsowskii’s LL equation in polar coordinates (Eq. (10.1) in [32]) contains a sign error at the damping term. This leads to exponential enhancement, rather than damping of the motion. His results on resonance frequency and line width (Eqs. (10.4) and (10.5) in [32]) are, however, unaffected by this.
with $F_x$ as derivative of $F$ with respect to the variable $x$ and $\theta_{t,0}$ the $i$'th magnetisation’s equilibrium azimuthal angle.

Close to the equilibrium position, where per definition $F_{\theta_i|\theta_{eq}} = 0$, we can Taylor expand the free energy derivative in all four variables:

\[
F_{\theta_i} = F_{\theta_i|\theta_{eq}} + F_{\theta_i\delta \theta_j} + F_{\theta_i\delta \varphi_j} + F_{\theta_i\theta_j} \delta \theta_j + F_{\theta_i\varphi_j} \delta \varphi_j \tag{3.17}
\]

\[
F_{\varphi_i} = F_{\varphi_i|\theta_{eq}} + F_{\varphi_i\delta \theta_j} + F_{\varphi_i\delta \varphi_j} + F_{\varphi_i\theta_j} \delta \theta_j + F_{\varphi_i\varphi_j} \delta \varphi_j \tag{3.18}
\]

**The free energy terms**

The specific expression for the total free energy $F$ of a model antiferromagnet like MnF$_2$ has been discussed in Chapter 2.1 and consists of an exchange interaction contribution $F_{\text{exchange}}$, a Zeeman term $F_{\text{Zeeman}}$ and an uniaxial anisotropy term $F_{\text{uniaxial}}$:

\[
F_{\text{tot}} = \lambda \langle M_1 \cdot M_2 \rangle_{\text{exchange}} - \mu_0 H_0 \cdot (M_1 + M_2) + K_{\text{uni}} \left( (c \cdot m_1)^2 + (c \cdot m_2)^2 \right)_{\text{uniaxial}} \tag{3.19}
\]

Please note that the simulation can readily be adapted to other antiferromagnetic systems by adding further anisotropy terms, e.g. cubic anisotropies, to (3.19).

**Linear equation system of the motion coefficients**

Inserting (3.16)–(3.18) into the LL equation (3.15) for both magnetisations $i = 1, 2$, results in a system of linear differential equations for the motion of the spherical angles of the magnetisation vectors:

\[
\begin{pmatrix}
\delta \dot \theta_1 \\
\delta \dot \varphi_1 \\
\delta \dot \theta_2 \\
\delta \dot \varphi_2
\end{pmatrix} = N(F, M) \cdot \begin{pmatrix}
\delta \theta_1 \\
\delta \varphi_1 \\
\delta \theta_2 \\
\delta \varphi_2
\end{pmatrix}, \tag{3.20}
\]

with $N(F, M)$ containing the second derivatives of $F$ at the equilibrium position. An ansatz $\delta \theta_i = A_i \exp(i \omega t)$ and $\delta \varphi_i = B_i \exp(i \omega t)$ converts the differential equation to a linear equation system of the form:

\[
0 = P(F, M, \omega) \cdot \begin{pmatrix}
A_1 \\
B_1 \\
A_2 \\
B_2
\end{pmatrix}, \tag{3.21}
\]
3.3 Antiferromagnetic Resonance Simulation

<table>
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<td>47.7 kA/m</td>
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<td>51.5 T</td>
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<td>0.82 T</td>
<td>0.84 T</td>
</tr>
<tr>
<td>Gyromagnetic ratio</td>
<td>$\gamma$</td>
<td>0.94 $\times$ $\gamma_0$</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 3.1: Parameter set of the simulation, which best fits the measured AFMR dispersion of Chapter 3.4.1 and reported literature values. Refer to Table 2.1 for conversion formulas to $K_{uni}$ and $\lambda$.

where $P$ can be decomposed into $P = R + D$, with $R$ describing the precessional motion and $D$ containing the damping terms:

$$R = \begin{pmatrix}
F_{\theta_1 \varphi_1} + \frac{i \omega}{\gamma M \sin \theta_1,0} & F_{\varphi_1 \varphi_1} & F_{\varphi_1 \theta_2} & F_{\varphi_1 \varphi_2} \\
F_{\theta_1 \theta_1} & F_{\varphi_1 \varphi_1} - \frac{i \omega}{\gamma M \sin \theta_1,0} & F_{\theta_1 \theta_2} & F_{\theta_1 \varphi_2} \\
F_{\theta_1 \varphi_2} & F_{\varphi_1 \theta_2} & F_{\theta_2 \theta_2} + \frac{i \omega}{\gamma M \sin \theta_2,0} & F_{\varphi_2 \varphi_2} \\
F_{\theta_2 \theta_2} & F_{\varphi_1 \theta_2} & F_{\theta_2 \theta_2} & F_{\theta_2 \varphi_2}
\end{pmatrix}$$

$$D = \begin{pmatrix}
-\frac{\alpha}{\sin \theta_1,0} F_{\theta_1 \varphi_1} & -\frac{\alpha}{\sin \theta_1,0} F_{\varphi_1 \theta_1} & -\frac{\alpha}{\sin \theta_1,0} F_{\theta_1 \theta_2} & -\frac{\alpha}{\sin \theta_1,0} F_{\varphi_1 \varphi_2} \\
-\frac{\alpha}{\sin \theta_1,0} F_{\theta_1 \varphi_1} & -\frac{\alpha}{\sin \theta_1,0} F_{\varphi_1 \theta_1} & -\frac{\alpha}{\sin \theta_1,0} F_{\theta_1 \theta_2} & -\frac{\alpha}{\sin \theta_1,0} F_{\varphi_1 \varphi_2} \\
-\frac{\alpha}{\sin \theta_2,0} F_{\theta_1 \varphi_2} & -\frac{\alpha}{\sin \theta_2,0} F_{\varphi_1 \theta_2} & -\frac{\alpha}{\sin \theta_2,0} F_{\theta_2 \theta_2} & -\frac{\alpha}{\sin \theta_2,0} F_{\varphi_2 \varphi_2} \\
-\frac{\alpha}{\sin \theta_2,0} F_{\theta_1 \varphi_2} & -\frac{\alpha}{\sin \theta_2,0} F_{\varphi_1 \theta_2} & -\frac{\alpha}{\sin \theta_2,0} F_{\theta_2 \theta_2} & -\frac{\alpha}{\sin \theta_2,0} F_{\varphi_2 \varphi_2}
\end{pmatrix}$$

The solution of $\det P(\omega) = 0$ yields the four eigenfrequencies of motion $\omega_p$ with $p = 1 \ldots 4$. For each frequency, the null space of $P(\omega_p)$ yields the motion coefficients $A_i$ and $B_i$, thus fully describing the trajectories of the two magnetisation vectors.

3.3.2 Simulation Results

In this section, the results of our AFMR simulation are presented and compared to both the available theory on AFMR and literature data. This enables a general validation of the simulation results.

The parameters of the simulation—$K_{uni}$, $\lambda$ and $\gamma$—are adjusted to best describe the measured AFMR dispersion, as presented in Chapter 3.4.1. Throughout the course of this thesis, all simulation results use this single set of parameters, presented in Table 3.1. The thus obtained MnF$_2$ simulation parameters are in good agreement with MnF$_2$ literature values of Hagiwara [23], Kotthaus [25] and Johnson [67].
Figure 3.7: Comparison between measured magnetic moment (a) and the corresponding simulated magnetisation (b) of MnF$_2$. Panel (a) depicts the measured magnetic moment in the direction of the applied field. The solid curves represent Néel’s theory on the antiferromagnetic susceptibility with different angles $\theta_H$ between $H_A$ and $H_0$. (b) shows the corresponding result of the simulated magnetisation equilibrium angles. Due to the unknown MnF$_2$ crystal size of (a), the magnetisation axis of (b) cannot be converted to electromagnetic the units as in (a).

3.3.2.1 Magnetisation Curves and Spin Flop of MnF$_2$

In a first step, the equilibrium positions of the simulated magnetisation vectors are validated. To this end, the magnetisation curves derived from our simulation are compared to a MnF$_2$ magnetisation measurement by Jacobs et al. [22]. The solid dots of Figure 3.7 (a) depict the measured magnetisation as a function of external field, for different crystal orientations. The solid lines represent the corresponding theoretical magnetisation curves, derived by Néel for different alignments $\theta_H$ of $H_0$ with respect to $H_A$. Below the spin flop field, the magnetisation vectors are antiparallel resulting in no net magnetisation. In the spin flop configuration, the magnetisation vectors slightly bend towards $H_0$, giving rise to an abrupt finite value of $M$. With increasing misalignment $\theta_H$, this reorientation loses its abruptness. The theory is in accordance with the data for misalignments of $\theta_H = 2.5^\circ$ and $\theta_H = 7.5^\circ$, respectively.

Figure 3.7 (b) shows the corresponding net magnetisation in the direction of $H_0$ calculated from the equilibrium positions of the magnetisations using our simulation

9Kotthaus et al. report $M = 600$ Oe, Johnson et al. report $M \approx 590$ Gauss. We used Kotthaus’ value, as it is the more recent source. The value of $M$ does not have an influence on the AFMR resonance position or the line width.

10The theoretical description of the susceptibility in antiferromagnets was derived by Néel in 1936, as mentioned in his Nobel Lecture [68].
3.3 Antiferromagnetic Resonance Simulation

Figure 3.8: Simulated resonance frequencies of MnF₂ for different alignment values \( H_A \) and \( H_0 \) of \( H_A \) and \( H_0 \). (a) depicts the two resonance branches of the parallel configuration with small misalignments \( (0° - 9°) \) and the resonance with \( H_A \perp H_0 \) \((H_A \text{ and } H_0 = 90°)\). The gray rectangle in panel (a) indicates our experimentally available measurement window, which is depicted in (b). Herein, only the lower resonance line can be observed, even for perfect alignment. In both graphs, the thin dashed line visualizes \( H_C \). (a) is in excellent agreement with Hagiwara’s AFMR MnF₂ data (Fig. 2.7), proving that the simulation yields valid resonance frequencies in both parallel and perpendicular configuration.

To summarize, the equilibrium positions of the magnetisation vectors obtained from our theory are in good agreement with both the established theory and the measured magnetisation, published in [22].

3.3.2.2 Antiferromagnetic Resonance Frequencies

The main purpose of the simulation is the description of the resonance frequencies of MnF₂ with arbitrary external field strengths and various angles \( \theta_H \) between \( H_A \) and \( H_0 \). Figure 3.8 (a) shows the higher and lower resonance mode of MnF₂ for four different values of \( \theta_H \) \((0° - 9°)\) and for the perpendicular alignment of \( H_A \) and \( H_0 \) \((\theta_H = 90°)\), as obtained from our AFMR simulation.

For \( H < H_C \) the simulation resembles the results of Kittel’s theory (cf. Fig. 2.5). The zero field frequency of 251 GHz is similar to the value obtained by (2.19) using the simulation parameters: \( f_{\text{res,Kittel}|H_0=0} = \frac{1}{2\pi} \gamma \mu_0 H_C = 249 \text{ GHz} \). The difference could arise from the fact, that Kittel’s theory uses an effective field approach to model the uniaxial anisotropy, while the simulation uses the more accurate free en-
energy description $F_{\text{uniaxial}}$. For $H_0 < H_C$, the ascending and descending resonance branches are reproduced and, as expected for small $H_0$, relatively unaffected by a misalignment. At the spin flop, for perfect alignment $\theta_H = 0^\circ$, the higher resonance mode drops to a smaller, but finite resonance frequency value, while the lower frequency mode drops to 0 Hz.

Above the spin flop, only one resonance mode exists for perfect alignment, in accordance with the symmetry of the two magnetisation vectors in the spinflop configuration. This single line resembles Nagamiya’s result (Eq. 2.20) for $H_0 > H_C$. With increasing misalignment, the drop of the higher resonance line at $H_C$ is less sharp in field and decreases in frequency span. The symmetry of the two sublattice magnetisations for $H_0 > H_C$ is lifted and the lower frequency mode takes finite values. At $H_C$, the lower frequency mode exhibits a minimum, which shifts towards higher frequencies with increasing misalignment. This is depicted in detail in Figure 3.8 (b), which restricts the y-axis to the accessible frequencies of our VNA. Only the lower frequency line can be observed in our setup, even for perfect alignment. Alignments $\theta_H \lesssim 3^\circ$ are required to resolve the characteristic second bending of the resonance line at fields above $H_C$.

For a validation of the AFMR simulation results, it is fruitful to compare Figure 3.8 not only to the established theory, but also to AFMR data. Hagiwara’s “Complete Frequency chart of MnF$_2$” [23] is depicted in Figure 2.7. Indeed, our simulation accurately reproduces all measured features over a large magnetic field and microwave frequency range. Both the lower and higher resonance modes shown in Figure 3.8 are in qualitative$^{11}$ accordance with Hagiwara’s data over the whole external field range. The simulation is particularly powerful around $H_C$. The broadened drop of the higher frequency line observed in Hagiwara’s data—so far not explainable using Kittel’s theory—can be readily reproduced by the simulation with a misalignment of a few degrees.

In addition, the simulation proves to be valid in the configuration of perpendicular alignment of $\mathbf{H}_0$ and $\mathbf{H}_A$ ($\theta_H = 90^\circ$), too. The simulated resonance frequency of this configuration is shown as dashed black line in Figure 3.8 (a) and resembles the form of Hagiwara’s perpendicular dispersion, as depicted in Figure 2.7.

Taken together, our AFMR simulation thus clearly is valid and extents the state of the art in AFMR theory, as it enables a consistent description of the AFMR dispersion over the entire magnetic field range.
Figure 3.9: Illustration of the small angle precession of the two sublattice magnetisations as a function of applied magnetic field $\mu_0 H_0$. The main graph depicts the two resonance frequencies, for a misalignment of $H_A$ and $H_0$ of $1^\circ$. At the constant field cuts (a) to (d), the motion of $M_1$ and $M_2$ is presented in the top and bottom row for the respective resonance mode. The upper left sketch shows the interpretation of these motion graphs. The equilibrium angle of $M_i$ is positioned at the center of the graph. After excitation, the magnetisations describe a damped precession back towards equilibrium. The rotation direction and the phase relation between the two magnetisation vectors is illustrated by the black arrow inset. The coordinates $X$ and $Y$ are proportional to $\varphi$ and $\theta$, but have arbitrary units, as the deflection strength depends on the microwave field strength. The scale of $X$ and $Y$ is equal, depicting elliptic precessions in real space as such. The motion graphs of $M_1$ and $M_2$ at a specific field strength have equal scales, too, while deflection strengths between e.g. (a) and (b) and within top and bottom row may not be compared. The time span of all depicted magnetisation precessions are equal.
3.3.2.3 Motion of the Sublattice Magnetisations

The substitution of $\omega_{\text{res}}$ into Equation 3.21 yields the eigenvector of the matrix $P(\omega_{\text{res}})$ for the respective dispersion mode and field strength. The eigenvector includes the motion information of the two sublattice magnetisations $M_i$, which is a damped precession back to the equilibrium position. These trajectories, however, vary with applied external field $H_0$ and are substantially different in the two resonance modes. Figure 3.9 depicts the magnetisations’ motions at different magnetic field strengths for a misalignment of $\theta_H = 1^\circ$. A detailed explanation of Figure 3.9 is also given in its caption.

The constant field cut (a) in Fig. 3.9 at $\mu_0 H_0 = 2$ T confirms the intuitive picture proposed in Chapter 2.2.3, stating that the different resonance frequencies of the two modes are caused by respectively different “motion driving” magnetisations. In the high frequency branch, $M_1$ “drives” the motion. The effective field of $M_1$, composed of the anisotropy and the external field, is enhanced by $H_0$ as all fields are in parallel. This results in a higher resonance frequency. Consequently, as $M_1$ is excited at its resonance frequency, the deflection of $M_1$ should be greater than the deflection of $M_2$. This is indeed the case, as seen in the motion graphs (a) of the top row. The precession direction of both magnetisations is clockwise, with a phase difference of 180$^\circ$, caused by the strong molecular field, which forces an antiparallel alignment. In the lower frequency branch, $M_2$ leads the motion and in turn has the larger deflection. Here, the effective field is decreased by $H_0$, as $H_A$ and $H_E$ are antiparallel to $H_0$, which consequently reduces the resonance frequency. The motion direction is turned, as the effective field of $M_2$ is opposite to the effective field of $M_1$.

At $\mu_0 H_0 = 9$ T (Fig. 3.9(b)) the behaviour is essentially the same, only the resonance frequencies of the two modes differ more clearly. Therefore, the magnetisations of the higher frequency mode complete multiple revolutions in the given time span, while the magnetisations of the lower frequency mode barely complete one.

After the spin flop (constant field cuts (c) and (d) in Fig. 3.9), the isotropy between $X$ and $Y$ coordinate is lifted and the motion pattern becomes elliptic. A motion along the $X$ coordinate moves $M_i$ within the plane spanned by $H_0$ and $H_A$.

Unfortunately, we did not have access to the Hagiwara data, to fit the simulation to the complete frequency chart. It should be noted, that even though our simulation was fitted only to data below 26.5 GHz the predicted zero field split of our MnF$_2$ crystal of 249 GHz is in sound agreement with Hagiwara’s measured zero field split of 260 GHz.

In the upper left sketch, the effective field of $M_1$ is directed rearwards along the dashed line. Thus, if $M_1$ drives the motion, the motions of both magnetisations in this projection is counter clockwise. The effective field of $M_2$ is however oppositely directed, along the dashed line to the front. If $M_2$ is leading, the motion direction is thus reversed. Note: If $M_1$ leads the motion, $M_2$ seems to circle against the direction of its own effective field. However the effective field of $M_2$ also includes the strong molecular field, which rotates with $mathbf{M}_1$.\[11\]
while a motion along $Y$ drives the magnetisation out of this plane$^{13}$ (cf. Figure 3.6). Figure 3.9 (c) shows, that for the higher frequency mode a precessional motion is possible and this motion takes mainly place within the plane of $H_0$ and $H_A$. The low resonance mode moves perpendicular to this plane and its motion is heavily damped, making only one overshoot on its relaxation towards equilibrium.

The constant field cut in Figure 3.9 (d) shows the same behaviour except for one unexpected difference. The higher resonance mode’s $M_1$-movement in $Y$ direction inverts its sign, resulting in opposite rotation directions. Starting from (c), the $Y$ extent of $M_1$’s motion ellipse shrinks with increasing magnetic field. At a value of $\sim 12.4$ T, the ellipse is compressed to a one-dimensional oscillation along $X$. With still increasing field, the ellipse extents again in $Y$, with a phase addition of $\pi$. Effectively, the $Y$ component inverts its sign from (c) to (d). The result is, that the magnetisation no longer turn in the same direction with a phase difference of $\pi$, reducing their exchange interaction by an antiparallel alignment. Instead $M_1$ rotates clockwise and $M_2$ counterclockwise, which increases the angle between them at every half turn and increases the exchange energy. We could not find an intuitive explanation for this behaviour, which is clearly present in the simulation.

### 3.3.2.4 Resonance Line Widths

The imaginary part of the calculated eigenfrequencies $\omega$ results in a damping of the precessional motion $(\varphi_i, \theta_i \propto \exp(i\omega t))$ with time. If the motion is Fourier transformed to the frequency domain, the effect of damping is that the single absorption frequency of the undamped magnetic system is broadened to a Lorentzian shape. Now, the motion may be excited by a (Lorentzian shaped) range of frequencies and the full width half maximum (FWHM) of this Lorentzian is given$^{14}$ by $\Delta \omega = 2\Im(\omega)$. This relation is used by Vonsowskii$^{32}$ to derive the expression for the line width in the frequency domain (2.11).

Figure 3.10 depicts the calculated line widths for the two resonance modes of Figure 3.9. Up to the critical field, the line widths are proportional to the respective frequency of the resonance line, as expected from (2.14) for viscous damping. At the critical field, the line width of the higher mode drops and takes an almost constant value for $H_0 > H_C$. The line width of the lower frequency mode symmetrically increases at $H_C$. Both line widths are no longer directly proportional to their respective resonance frequencies, making (2.14) invalid above $H_C$. According to the previously discussed relationship between line width and the damping of the motion, the trend of the line width should be reflected in the motion graphs of Figure 3.9. Indeed, this is the case. When comparing the inner circles of motion, e.g. the final deflection after a certain time span, those graphs with greater damping correspond

$^{13}$In the notation of Figure 3.6, $X$ corresponds to $\theta$ and $Y$ to $\phi$

$^{14}$This may be derived by a simple Fourier transformation of $\exp(i\omega t)$, with complex $\omega$. 


to those resonance modes which exhibit greater line widths $\Delta \omega$ at this magnetic field strength and vice versa. This is the case for all motion graphs. The physical origin of smaller or enhanced damping in the AFMR is—up to this date—not intuitively explainable and may not be reduced to a simple expression like (2.14).

We are not aware of a consistent study of damping in MnF$_2$ over larger frequency and magnetic field ranges. It would be interesting to check the line width predictions from our simulation against such a MnF$_2$ line width study in the future. We would like to emphasize again that our simulation approach with the inclusion of viscous damping clearly goes beyond the current state of the art in AFMR theory.

### 3.4 Comparison of Simulation and Measurement

The comparison of simulation results with measured AFMR properties is the key validation of the simulation. In the following section, first the excellent agreement between the simulated and the measured MnF$_2$ dispersion is discussed for several measurements. Then, using the analysis based on the CPW impedance shift presented in Sec. 3.1.3.1, the measured MnF$_2$ dispersion and absorption is presented and corresponding constant field cuts are fitted to extract resonance position and line width. Finally, these resonance positions and line widths are compared with the simulation results.

#### 3.4.1 AFMR Dependence on the External Field Alignment

Figure 3.11 presents an insight into the power of the AFMR simulation: Different MnF$_2$ AFMR dispersions measured throughout the course of this thesis may be explained with the simulation, by adjusting only a single parameter $\theta_H$, the angle between external magnetic field $H_0$ and the axis of crystalline anisotropy $H_A$. 
3.4 Comparison of Simulation and Measurement

Figure 3.11: Different AFMR measurements and simulated resonance curves with adjustment of the angle $\theta_H$ between external field and uniaxial anisotropy. The depicted dispersions stem from two different samples, MnF2 #1 (a, d) and MnF2 #2 (b, c). The graphs use the same color code as the previously shown Figures 3.4 and 3.5. Again, a reference cross section without resonance taken from 8.5 T is subtracted from the absorption $|S_{21}|^2$ to exclude absorptions unrelated to MnF$_2$. The lower absorption of (c) is due to a smaller filling factor by a different sample mounting above the CPW (cf. Chapter 5.1.2). The sample temperature is $T = 4$ K, the microwave power is $P_{mw} = 5 \text{ dBm}$ in (a-c) and $P_{mw} = 0 \text{ dBm}$ for (d).
Figure 3.11 depicts four different AFMR measurements of two MnF$_2$ samples, each with new sample mounting and therefore possibly different crystal alignments. The measured AFMR dispersions vary substantially, with minimum resonance frequency values between 7.5 GHz and 23 GHz. The simulation is, however, capable of reproducing the different resonance dispersions, simply by appropriately adjusting $\theta_H$. The uncertainty in the exact $\theta_H$ value is only $\sim 0.1^\circ$, limited by the broad line width of MnF$_2$. As stated in Section 3.3.2, the parameters used for the simulation, given in Table 3.1, agree well with the MnF$_2$ properties reported by [23],[25] and [67].

The good $\theta_H$ resolution turns AFMR into a suitable tool for investigating the alignment of the external magnetic field with the anisotropy axis. The extracted value of $\theta_H$ is directly linked with the equilibrium positions of the magnetisations, which can thereby be predicted over the entire field range. From this, antiferromagnetic properties like the net magnetisation (and their $H_0$-dependence) can be extracted without the use of a high magnetic field SQUID magnetometer.

Furthermore, the $\theta_H$ of our measurements vary about several degrees, which is caused by the sample mounting process. A relatively small crystal is positioned onto a thin center conductor and then fixed using rubber cement, which may also exhibit non-linear distortions upon cooling down. These effects might well lead to misalignments of $5^\circ - 6^\circ$. In our setup, a viable way to obtain a better crystalline alignment would be to use an active positioning system and to iteratively measure the AFMR and readjust the alignment. Details on how such an positioning system could be implemented are outlined in the outlook, cf. Chapter 6.1.

3.4.2 Resonance Line Shape Analysis

The investigation of the resonance line shape is based on the impedance shift analysis of Kalarickal et al. [64], as presented in Chapter 3.1.3.1. This analysis introduces the complex quantity $U$, which is related to the complex permeability $\mu = \mu' + i\mu''$ of the specimen and yields more accurate line widths than a $|S_{21}|^2$ analysis. The feed lines and the CPW were not calibrated prior to the AFMR measurement, hence the phase of $S_{21}$ at each frequency point is arbitrary. This issue is resolved via the introduction of a reference plane at a magnetic field strength exhibiting no magnetic resonance. At this reference, the phase of $S_{21}$ is defined to take a specific value. For the presented data, the phase angle of $S_{21}$ of all data points is unwrapped, so that the microwave phase for all frequency points at $H = 8.5$ T is zero. In formula with phase angle symbol $\angle$, this reads:

$$\angle S_{21,\text{unwrapped}}(f, H) = \angle S_{21,\text{meas}}(f, H) - \angle S_{21,\text{meas}}(f, H = 8.5$ T)$$  \hspace{1cm} (3.24)

Thereafter, the value of $U$ is calculated according to (3.10).
Figure 3.12: The real (a) and imaginary (b) part of the dimensionless quantity $U_{\text{meas}}$, which is related to the dynamic permeability $\mu = \mu' + i\mu''$ of the sample ($T = 4\,\text{K}, P_{\text{mw}} = 5\,\text{dBm}$). The real part shows the dispersion signal, the imaginary part contains the absorption. The corresponding power absorption analysis of this measurement is presented in Figure 3.11(b).

Figure 3.12 presents the obtained values $U_{\text{meas}}$, with Re($U_{\text{meas}}$) displayed in panel (a) and Im($U_{\text{meas}}$) in panel (b). As expected for the susceptibility (cf. Fig. 2.4), the real part shows a dispersive line shape proportional to $(1 + \chi')$, with positive (red) values, if the microwave frequency $f$ is lower than the resonance frequency $f_{\text{res}}$, and negative (blue) values, if $f > f_{\text{res}}$. The dispersive line shape is observable in both, the constant frequency and constant magnetic field cuts. Im($U$) contains the expected Lorentzian absorption shape of the sample, proportional to $\chi''$. The absorption takes solely negative values at resonance, indicated by the blue color and is, again, observable in both cuts.

The cuts for constant microwave frequency exhibit less spurious signal than the constant magnetic field cross sections, as a sweep of the magnetic field is not affecting the electric transmission properties of the microwave cables and the CPW. Under magnetic field variation, only the impedance of the CPW section loaded with the magnetic specimen is affected. Hence, for the fitting of the resonance line shape, cuts at constant frequency are used.

3.4.2.1 Fitting of $U_{\text{meas}}$

In the following, it is assumed that the dynamic susceptibility of the antiferromagnet resembles the form of the ferromagnetic susceptibility (2.16). This is motivated as follows: Both, antiferromagnetic and ferromagnetic magnetisations are magnetisation vectors which precess in an effective field. In the antiferromagnetic case, the
only difference for one sublattice magnetisation vector is, that the effective field contains an additional term due to the molecular field. It seems unlikely, that this additional term significantly changes the functional shape of the magnetisation response to the microwave excitation.

Thus, in the following, we assume that the line shape of \( U \propto \mu = 1 + \chi \), should follow the complex susceptibility given by (2.13), with a dispersive real part and a Lorentzian imaginary part. In general this is the case as observed for \( U_{\text{meas}} \) of Figure 3.12. In detail, however, neglecting the reflection parameter in the evaluation of \( U \) introduces a mixing of the absorption and dispersion in the measured quantity \( U_{\text{meas}} \) [64]. This mixing leads to a complex phase shift \( \Phi \) of \( U_{\text{meas}} \propto (1 + \chi e^{i\Phi}) \). Taking this into account, we can define a complex fit function, as the sum of the (for our measurement window usually) two antiferromagnetic resonances along a constant frequency cut as:

\[
U_{\text{fit}}(H_0)\big|_{f=\text{const}} = C \left[ 1 + \chi_0 + \chi_a(H_0)e^{i\Phi_a} + \chi_b(H_0)e^{i\Phi_b} \right], \tag{3.25}
\]

where \( \chi \) is the dynamic susceptibility given by (2.16), \( C \) is a real scaling parameter, \( \chi_0 \) a complex offset and \( \Phi \) the phase shift adjustment. \( a \) and \( b \) represent the two resonance dips, above and below the spin flop field. Using a least squares algorithm, both, the imaginary and real part of \( U_{\text{fit}} \) are fitted simultaneously to the measured data \( U_{\text{meas}} \). Above 23.8 GHz only one resonance is observable, thus only one resonance is used in the fit function.

Figure 3.13 depicts the obtained fitted curves \( U_{\text{fit}} \) along with the measured values \( U_{\text{meas}} \) for a set of frequencies. Again, both real (Fig 3.13(a)) and imaginary (Figure 3.13(b)) parts are shown. \( U_{\text{fit}} \) nicely reproduces the line shape of \( U_{\text{meas}} \) for all depicted frequencies. Note also that the fit of the 14.5 GHz cut does not reproduce the spin wave modes evident at this field & frequency combination, since spin wave modes are not incorporated in the theory that \( U_{\text{fit}} \) is based on. The values of the phase shift adjustment \( \Phi \) of the fits are less than 15° for the presented frequencies.

In summary, it is found that the analysis of Kalarickal et al. [64] yields a reliable set of \( U_{\text{meas}} \), which from our raw data correctly separates the dispersive signal from the Lorentzian absorption. As corroborated on hindsight by the fit \( U_{\text{fit}} \), \( U_{\text{meas}} \) is adequately described using the complex susceptibility \( \chi \), defined in (2.16). We therefore take the resonance line width and position of MnF\(_2\) extracted from \( U_{\text{fit}} \) as a valid and robust database to compare our simulation results to.

### 3.4.3 Comparison Between Fitted Line Shape Properties and the Corresponding Simulation Results

In the following, the resonance field and the resonance line width of MnF\(_2\) are compared to the corresponding quantities calculated with our simulation approach.
3.4 Comparison of Simulation and Measurement

![Figure 3.13: Stack plots of the real and imaginary part of \( U_{\text{meas}} \) and the fitted resonance \( U_{\text{fit}} \) for a set of microwave frequencies. The data represents constant frequency cuts of the measurement presented in Figure 3.12. \( U_{\text{meas}} \) represents the experimental magnetic permeability \( \mu \) and is extracted from \( S_{21} \) according to (3.10). \( U_{\text{fit}} \) is obtained by fitting (3.25) to \( U_{\text{meas}} \).](image)

Therefore, we perform fits \( U_{\text{fit}} \) to the measured quantity \( U_{\text{meas}} \) for all frequencies shown in Figure 3.12 that exhibit a resonance signal (fits to \( \sim 680 \) frequency cuts). The thus obtained resonance position and line shape of \( U_{\text{fit}} \) are the experimentally determined properties of MnF\(_2\), as the previous section showed.

### 3.4.3.1 Resonance Position of MnF\(_2\)

The resonance fields of MnF\(_2\) obtained from \( U_{\text{fit}} \) are depicted in Figure 3.14 (a) as green symbols, along with the simulated AFMR dispersion (red line) and the experimental absorption data \( \text{Im}(U_{\text{meas}}) \) in false color code. The three sets of dispersions coincide very well. Small deviations of the fitted resonance position from the simulation occur close to the critical field \( H_C \approx 9.45 \)T and towards higher fields, as evident in the constant frequency cuts. At the critical field, the two distinct absorption Lorentzians merge into one broadened absorption, from which an accurate determination of the two resonance fields is no longer possible and which explains the deviation of simulation and fits in this region. At high magnetic fields, the resonance line width increases drastically (cf. Fig. 3.14 (b)), resulting in a reduced accuracy of the absorption center extracted from the fits.

Nevertheless, we would like to stress that the excellent agreement of the AFMR dispersions of \( U_{\text{meas}} \) and \( U_{\text{fit}} \) with the calculated AFMR dispersion corroborates our simulation approach once more.
Figure 3.14: The MnF$_2$ AFMR resonance position (a) and the line width (b), obtained from experiment (symbols) at $T = 4\,\text{K}$ and calculated from our simulation approach (red lines). Panel (a) depicts the absorption $\text{Im}(U_{\text{meas}})$ in false color code, as previously presented in Fig. 3.11 (b). The green crosses show the measured resonance position extracted from $U_{\text{fit}}$. The red line depicts the resonance position calculated from our simulation, assuming a misalignment of $\theta_H = 3.52^\circ$. Panel (b) shows the extracted MnF$_2$ resonance line width as a function of magnetic field strength along with the simulated magnetic line width using a damping parameter $\alpha = 10^{-3}$. 
3.4.3.2 Resonance Line Width of MnF$_2$

The line width as a function of external field is depicted in Figure 3.14 (b), with the experimentally obtained, fitted line widths as green symbols and the simulated line width as red curve. The simulated magnetic line width is extracted from the simulated frequency line width according to (2.14):

$$\Delta H_0 = \frac{d(H_{\text{res}})}{d\omega_{\text{res}}} \Delta \omega,$$

where the dispersion derivative $\frac{d(H_{\text{res}})}{d\omega_{\text{res}}}$ is calculated by numerical differentiation of the simulated resonance dispersion$^{15}$.

At small magnetic field strengths, the fitted resonance line width is small, with widths down to 10 mT. Around the critical field, the line width increases drastically, then returns to values of 50 mT just above the spin flop field and significantly increases again for $\mu_0 H_0 \gtrsim 10.2$ T.

For the simulation, it can be stated, that the behaviour of the magnetic field line width is more influenced by the derivative of the dispersion, than by the dependence of $\Delta \omega$ on external field (cf. Fig. 3.10). Within field regions of steep dispersion slope, e.g. $\mu_0 H_0 < 9.3$ T and $9.7$ T $< \mu_0 H_0 < 10$ T, the line width is small, while in field regions with flat dispersion, $\mu_0 H_0 \approx 9.5$ T and $\mu_0 H_0 > 10.5$ T, the line width increases drastically. The trend of increasing $\Delta \omega$ with increasing fields is superimposed to this behaviour, but plays a secondary role for the simulation.

The extracted MnF$_2$ resonance line width of the fits generally follows the trend of the simulation, but deviates in the vicinity of the critical field. The value of the derivative is crucial for the resonance line width, especially in this regime, where the derivative approaches zero. As noted in the previous section, the fitted and simulated resonance position differ slightly in this regime. This could lead to an underestimation of the derivative by the simulation, explaining the narrower line width of the simulation around $H_C$. Additionally, the two absorption dips superimpose and the spin wave modes become more pronounced in this region, which reduces the accuracy of the fitted line width values at $H_C$.

Nevertheless, the simulation reproduces the measured line width very well for $\alpha = 10^{-3}$. We would like to note that a damping parameter $\alpha = 10^{-3}$ used in the simulation is rather an upper limit for the damping parameter of the uniform mode. According to the previous discussion, reliable magnetic line widths may only be fitted for $\mu_0 H_0 \lesssim 8.7 \ll H_C$. Unfortunately, the available data in this region is scarce. Secondly, the inhomogeneous field distribution of the microwave magnetic field $\mathbf{h}_1$ of the CPW leads to an additional broadening of the resonance$^{70}$, up to a factor of 10.

Finally, the effect of spin wave mode excitation on the line width has to be considered. Small misalignments of crystal axis and external field will increase the

$^{15}$As numerical differentiation scheme, the dispersion was modelled by a piece wise steady polynomial, a spline of third degree. Compared to first order finite differences, the spline derivatives returns an improved approximation of the underlying derivative$^{69}$. 
line width of the uniform mode drastically, by the splitting of the uniform mode into several spin wave modes \[25\]. The high field resolution measurement of Figure 3.5 shows, that indeed spin wave modes are observed over a field region \(>50\,\text{mT}\), thereby broadening the line width. The smallest reported MnF\(_2\) line widths of the uniform mode (0.5mT, \[25\]) are about an order of magnitude lower than the smallest line widths of this study \(\sim 10\,\text{mT}\) at comparable microwave frequency\[^{16}\]. Unfortunately, it is not possible to compare our observed spin wave mode line widths to literature. The smallest reported width of a spin wave mode (10\(\mu\)T, \[25\]) is about a factor of 20 thinner than the magnetic field resolution \(\sim 200\mu\text{T}\) of our most detailed measurement\[^{17}\]. The narrowest spin wave mode we observed has a width of \(\sim 500\mu\text{T}\).

3.5 Temperature Dependence of the Antiferromagnetic Resonance

So far, all presented MnF\(_2\) resonance experiments were conducted at a sample temperature of 4 K. Now, the temperature dependence of the dispersion is investigated. MnF\(_2\) has a Néel temperature of \(T_N = 67\,\text{K}\) \[^{72}\]. Above \(T_N\), MnF\(_2\) is paramagnetic, exhibiting electron spin resonance ESR. ESR measurements performed at \(T = 80\,\text{K}\) (not shown) confirm the paramagnetic state, with a resonance dispersion in accordance with the linear ESR dispersion in accordance with the linear ESR dispersion of (2.4) using a \(g\)-factor of 2.

Below \(T_N\), AFMR is observed. The temperature dependence of the AFMR dispersion is shown in Figure 3.15. Starting with the 4 K measurement as a reference, a doubling of thermal energy to a temperature of 10 K does not affect the dispersion significantly. At 20 K, the minimum of the dispersion is pushed towards higher frequencies and higher magnetic fields. For 40 K, this trend continues and the resonance minimum drifts about 0.8 T to higher fields and \(\sim 5\,\text{GHz}\) along the frequency axis. Note that this drift seems even to quicken with higher temperature.

As known from the simulation, the minimum of the AFMR dispersion occurs at the spin flop field, which is related to the internal fields by \[^{2.2}\]: \(H_C = \sqrt{H_A (H_A + 2H_E)}\). Figure 3.15 shows that \(H_C\) increases with temperature, which would imply that either the crystalline anisotropy or the molecular field or both are enhanced by the additional thermal energy. However, a variation of the simulation’s effective field parameters does not allow to reproduce the measured AFMR dispersions for temperatures \(T \geq 10\,\text{K}\).

White \etal\[^{73}\], report an increase of \(H_C\) of MnF\(_2\) above \(\sim 10\,\text{K}\). They are able to explain the increase on the basis of spin wave theory with four- and six-magnon

\[^{16}\]However, \[^{25}\] report minimum line widths of 5mT, well within our observed range.

\[^{17}\]Due to our power supply of the magnet, we are restricted to a magnetic field resolution above 100\(\mu\text{T}\).
3.5 Temperature Dependence of the Antiferromagnetic Resonance

Figure 3.15: Temperature dependence of the antiferromagnetic resonance of MnF$_2$. The measurements were performed in order (a) to (d), varying the temperature of the sample and keeping the alignment $\theta_{H} = 2.02^\circ$ of uniaxial anisotropy axis and external field constant. The red auxiliary lines mark the 4 K position of the dispersion minimum. The alignment of (a) was evaluated in Figure 3.11 (a). The power of the incident microwave is $P_{\text{mw}} = 5$ dBm for all temperatures.
scattering processes. A treatment of AFMR to this theoretical degree is, however, beyond the scope of this work.

In this chapter we have investigated the antiferromagnetic resonance (AFMR) of MnF$_2$. Using a coplanar wave guide (CPW) in conjunction with a liquid Helium cryostat, we are able to measure the AFMR dispersion in broad-band at varying sample temperatures. This is—to our knowledge—the first report of CPW based AFMR experiments. For the discussion we focus on AFMR measurements at 4 K. The measured resonance dispersion is insufficiently described by the established AFMR theory. We therefore develop an AFMR simulation, which is able to calculate the resonance frequencies, the resonance line width and the motion of the sublattice magnetisations in arbitrary external field configurations. We find an excellent agreement of different measured MnF$_2$ AFMR dispersions and the simulated resonance frequencies by adjusting a single parameter: The misalignment of the external magnetic field and the crystalline anisotropy axis. The accordance of the measured and calculated resonance line width as a function of external field furthermore corroborates the power of our simulation approach. We can therefore conclude that the developed simulation advances the current state of the art in AFMR theory.
Chapter 4

Spin Pumping with MnF$_2$

Conventional spin pumping is performed in heterostructure samples consisting of a ferromagnetic material (FM) covered by a thin metallic (N) top layer. In ferromagnetic resonance, the magnetisation dynamics of the FM emits a spin current across the FM|N interface, which is converted to a charge current by the inverse spin Hall effect and thus detectable as DC voltage under open circuit conditions.

In this chapter, we present the results of spin pumping experiments with MnF$_2$|Pt bilayers, which is the first attempt of spin pumping with antiferromagnets to our knowledge.

4.1 Experimental setup

As mentioned in the previous chapters, MnF$_2$ is chosen as antiferromagnet, because its simple crystalline anisotropy turns MnF$_2$ into an excellent model antiferromagnet. The commonly used material for the metallic thin film is platinum (Pt), as its large spin Hall angle $\alpha_{\text{SH}}$\textsuperscript{[44]40} results in large inverse spin Hall voltages (cf. Sec. 2.3.2). Our spin pumping samples thus consist of a Pt–film evaporated onto the 0.5 mm × 3 mm side of the MnF$_2$ crystal, as depicted in Figure 4.1. Prior to the evaporation, the crystals are polished with diamond paste with a graining of 0.25 mm, giving a nominal surface roughness of 0.125 mm. From experience, the resulting surface roughness is significantly finer, but the exact value was not determined experimentally. Due to the considerable solubility of MnF$_2$ in water (1.06g/100mL,\textsuperscript{[75]})}, further grinding with water based polish was not executed. Still, we are able to get cohesive Pt films with a thickness of 7 nm. The Pt was deposited using electron beam evaporation with a shadow mask to provide a $\sim$ 210 $\mu$m × 3 mm sized strips. Two such Pt strips were prepared, on the crystals MnF$_2$#1 and MnF$_2$#2.

As seen in Fig. 4.1, the sample is placed on the CPW with its axis of crystalline anisotropy $\mathbf{H}_A$ perpendicular to the CPW plane. The Pt film to the side of the crystal has thus a surface normal perpendicular to $\mathbf{H}_0$. We introduce a coordinate system with $\mathbf{x}$ parallel to the Pt surface normal, $\mathbf{y}$ along the Pt-strip and $\mathbf{z}|\mathbf{H}_A$. As discussed in Chapter 3, AFMR is excited best, if the microwave field $\mathbf{h}_{\text{rf}}$ is
Figure 4.1: The setup of the MnF$_2$ spin pumping experiments. (a) depicts the end of the measurement dip stick, with the SMA to Mini-SMP adapter connecting the CPW loaded with the sample. (b) illustrates the measurement configuration on the CPW. In the MnF$_2$ crystal AFMR is generated by means of the microwave field $h_{rf}$. The generated voltage $V_{DC}$ across the Pt strip is measured via the gold wire contacts. The external field $H_0||z$ and the direction of the Pt film normal parallel to $x$ are chosen such, that above $H_C$, an induced and excited net magnetisation along $z$ would give rise to an electric field $E_{ISH}||y$, measurable as $V_{DC}$.

perpendicular to the magnetisations $M_i$. Since $M_i||z$ below the spin flop field, the resonance condition is best met right above the center conductor of the CPW and the MnF$_2$/Pt interface of the crystal is positioned exactly there. The strength of the microwave field decreases approximately exponentially with distance above the CPW\textsuperscript{1} \cite{76}. Consequently, the strength of the generated inverse spin hall voltage $V_{ISH}$ exhibits the same exponential decay with distance from the CPW. To avoid the short circuit of $V_{DC}$ over film areas where no or smaller $V_{ISH}$ is generated, the Pt strip is only evaporated close to the CPW onto the crystal. The exponential decay length of the microwave field magnitude is in the order of the width of the center conductor ($\sim 560 \mu m$) \cite{76}, hence the Pt strips width of $\sim 210 \mu m$ prevents such shorting of $V_{ISH}$.

The Pt strip is contacted with gold wires, which are attached to the platinum and the contact pads using silver glue. To avoid unintentional contacting of the CPW, an isolation layer of SU-8 photo resist \cite{77} is spin coated onto the CPW. The

\textsuperscript{1}The decay is not strictly exponential, but, as checked by simulation of the CPW microwave field, an exponential with decay length of the order of the center conductor width is a good approximation.
microwave transmission of the CPW remains unaffected by the additional isolation layer. Over the gold wires, via the contact pads and copper (Cu) wires, the Pt strip is connected to a Keithley 2182A Nanovoltmeter to measure $V_{\text{DC}}$. Analogous to the AFMR experiments, the CPW is connected to two ports of the VNA, by use of Mini-SMP to SMA adapter cables and the microwave cables of the dip stick.

The external field $H_0$ is ideally aligned parallelly with the axis of crystalline anisotropy $H_A$. Its unintentional, but experimentally unavoidable misalignment is, again, denoted $\theta_H$.

### 4.2 Results

The spin pumping measurements are executed as follows: For a fixed frequency, magnetic field sweeps are conducted at a sample temperature of 4 K. At each magnetic field point, $S_{21}$ is measured in continuous wave mode of the VNA and, simultaneously, the DC voltage across the Pt strip, $V_{\text{DC}}$, is recorded. We use the largest possible incident microwave power of our VNA at these frequencies, $P_{\text{mw}} = 11$ dBm.

Figure 4.2 shows the typical results of these measurements for two distinct microwave frequencies, 19.1 GHz in panel (a) and 16.9 GHz in panel (b). In orange, the microwave absorption $|S_{21}|^2$ is plotted against the magnitude of the magnetic field. In green, $V_{\text{DC}}$ is displayed. Again, the subtraction of a reference $|S_{21}|^2$ @ 9 T excludes the non-MnF$_2$ dependent microwave absorption contributions. In the same way a reference for $V_{\text{DC}}$ is subtracted. The measurements are taken not only for $H_0 > 0$ but also for $H_0 < 0$, referred to as negative fields. For the sake of clarity, the negative field data is offset by $\Delta_{\text{plot}}|S_{21}|^2 = 0.03$ and $\Delta_{\text{plot}}V_{\text{DC}} = 0.6 \mu\text{V}$.

Figure 4.2 (a) demonstrates that a DC voltage indeed arises over the Pt film, if and only if the antiferromagnet is excited at resonance. The positions of the absorption of $|S_{21}|^2$ and $V_{\text{DC}}$ coincide and the line shapes are similar. Under field reversal ($H_0 < 0$), the microwave absorption does not change significantly, as expected for AFMR (cf. Fig. 3.4). $V_{\text{DC}}$ is partly affected by the field inversion, as the line shape of the negative field resonance at 9.1 T does not match its positive field equivalent. The sign of $V_{\text{DC}}$ does not change under magnetic field reversal. Only the strength of the $V_{\text{DC}}$ dip decreases by $\sim 0.2 \mu\text{V}$. At 16.9 GHz, displayed in Fig 4.2 (b), the resonances shift their magnetic field positions, as expected from AFMR theory. Both the microwave absorption and the $V_{\text{DC}}$ signal in resonance increase in strength. The line shape of the two quantities generally coincide. Most of the spin wave modes of $|S_{21}|^2$ are also observed in the spectrum of $V_{\text{DC}}$. The field inversion, again, has no influence on the microwave absorption, however, $V_{\text{DC}}$ does not fully match its positive field correspondence, e.g. at $\sim 9.55 \text{T}$. Again, no sign change of $V_{\text{DC}}$ is found under external magnetic field inversion.
Figure 4.2: The microwave absorption $|S_{21}|^2$ in orange and generated voltage over the Pt strip $V_{DC}$ in green as a function of the magnitude of the external field, taken at two microwave frequencies (19.1 GHz (a) and 16.9 GHz (b)) with $T = 4K$ and $P_{mw} = 11$ dBm. The measurements are also conducted under field inversion (“neg. Field”), displayed with an offset for clarity of $\Delta |S_{21}|^2 = 0.03$ and $\Delta V_{DC} = 0.6 \mu V$, as indicated by the black arrow. As done in the previous Chapter, a reference at $\pm 9T$, is subtracted from $|S_{21}|^2$ and analogously from $V_{DC}$, too. Both (a) and (b) have equal axis scales.
Figure 4.3: The microwave absorption $|S_{21}|^2$ due to AFMR of MnF$_2$ (a,b) and the DC voltage $V_{DC}$ across the Pt strip (c,d), as a function of frequency and magnetic field, with both positive (a,c) and negative (b,d) magnetic fields. Analogous to Fig 4.2 a reference at ±9 T is subtracted from all four data sets to extract the MnF$_2$ dependent signal components. $P_{mw} = 11$ dBm and $T = 4$ K for both measurements.
Figure 4.3 shows the AFMR microwave absorption $|S_{21}|^2$ in gray color code and the DC voltage $V_{\text{DC}}$ in red-green-blue color code, for frequencies between 16 GHz and 19.5 GHz with $P_{\text{mw}} = 11$ dBm. The data is again obtained by magnetic field sweeps at fixed frequencies. Figure 4.3 (a) and (b) show the MnF$_2$ resonance dispersion for positive and negative fields, which coincide very well. Figure 4.3 (c) and (d) present $V_{\text{DC}}$, whose signal positions match the dispersions of the AFMR. Again, distinct modes can be tracked, both in $|S_{21}|^2$ and $V_{\text{DC}}$. Indicated by the blue color, only negative $V_{\text{DC}}$ values are measured and there is no sign reversal under field inversion. However, as noted for Fig. 4.2 (a), there is a small but consistent mismatch between the DC signals for positive and negative fields of about $\sim 0.2 \mu V$, which is faintly visible when comparing the colors of Figure 4.3 (c) and (d).

4.3 Discussion

We identify three effects that may generate a DC voltage across the Pt strip, if the magnetic specimen below is driven into magnetic resonance. The first effect is spin pumping, the conversion of spin current emitted by a resonantly magnetisation into a charge current. The second effect is a thermopower, caused by anisotropic resonant heating of the sample. The third possibility—microwave rectification—is a variation of the Pt resistance, due to anisotropic magnetoresistance, that in conjunction with an alternating current of the same frequency, generates a DC voltage. In the following, these three effects are presented and the corresponding $V_{\text{DC}}$ is discussed.

4.3.1 Spin Pumping

As presented in Chapter 2.3.1, a ferromagnetic magnetisation excited in resonance can relax towards equilibrium by the emission of a spin current $J_{\text{s pump}}$ into an adjacent normal metal (N). If the normal metal has a finite spin Hall angle $\alpha_{\text{SH}}$, $J_{\text{s pump}}$ is converted to a charge current, which is detectable as inverse spin hall voltage $V_{\text{ISH}}$.

In the derivation of $V_{\text{ISH}}$ (2.26), the magnetisation $\mathbf{M}$ is assumed to be perpendicular to the connection line of the two contacts that measure $V_{\text{ISH}}$. In antiferromagnets, this perpendicular configuration is not stable, as the magnetisations change their orientation across the spin flop transition. With the reorientation of the equilibrium orientation $\mathbf{M}_{\text{eq}}$, the polarisation of the spin current, $\hat{s}||\mathbf{M}_{\text{eq}}$ changes: $\hat{s} = \mathbf{M}_{\text{eq}}/M = \mathbf{m}_{\text{eq}}$. The electric field due to the inverse spin hall effect is $E_{\text{ISH}} \propto J_{\text{s}} \times \hat{s}$ (2.25). In the coordinate system of Fig. 4.1, the contacting of our sample is sensitive to the $y$ component of $E_{\text{ISH}}$, giving:

$$V_{\text{ISH}} = L \left(E_{\text{ISH}}\right)_y \propto (J_{\text{s}} \times \mathbf{m}_{\text{eq}})_y = J_{\text{s}} m_{\text{eq},y},$$

(4.1)
with Pt strip length \( L \), \( m_{\text{eq},z} \) the \( z \)-component of \( \mathbf{m} \) and the spin current \( J_s = J_s \hat{x} \) along the AFM|N interface normal in \( x \)-direction. With our contacting of the Pt film along \( y \), we are thus only sensitive to the \( z \) component of the polarisation of a spin current and will therefore measure only a fraction \( m_{\text{eq},z} \) of the maximal \( V_{\text{ISH}} \) if the magnetisation is not fully aligned with \( z \).

We treat the spin pumping in antiferromagnets as separate spin pumping from two sublattice magnetisations. The voltage generated by two magnetisation vectors then is the sum of their individual contribution \( V_{\text{ISH,AFM}} = V_{\text{ISH,M}_1} + V_{\text{ISH,M}_2} \). Their spin current strengths \( J_{s,i} \) pumped by the two sublattice magnetisations only differ in the precession cone angles \( \Theta_i \) and the equilibrium orientation \( m_{i,\text{eq}} \), and the expected antiferromagnetic spin pumping voltage is of the form:

\[
V_{\text{ISH,AFM}} \propto m_{1,\text{eq},z} \sin^2 \Theta_1 + m_{2,\text{eq},z} \sin^2 \Theta_2. \tag{4.2}
\]

As in conventional spin pumping with FM|Pt hybrids \([20]\), an inversion of the external magnetic field still leads to a sign change of \( V_{\text{ISH,AFM}} \): With \( H_0 \rightarrow -H_0 \), the magnetisation configuration is inverted, \( m_{i,\text{eq},z} \rightarrow -m_{i,\text{eq},z} \), while the precession cone angles \( \Theta_i \) are unaffected\(^2\), resulting in \( V_{\text{ISH,AFM}} \rightarrow -V_{\text{ISH,AFM}} \). Thus, spin pumping cannot be the origin of the \( V_{\text{DC}} \) observed in Figures 4.2 and 4.3, since \( V_{\text{DC}} \) does not change sign with field inversion. Spin pumping could, however, contribute to the \( \sim 0.2 \mu \text{V} \) difference of \( V_{\text{DC}} \) at resonance between positive and negative magnetic fields (cf. Fig. 4.3(a)). This also sets an upper boundary for the size of the spin pumping voltage in our setup to \( V_{\text{ISH}} < 100 \text{nV} – 200 \text{nV} \) at incident microwave power of \( 13 \text{mW} \).

Although antiferromagnetic spin pumping is not detected in our sample, we would still like to present a discussion of the expected spin pumping voltage \( V_{\text{ISH,AFM}} \) with antiferromagnets in Appendix A. Using our AFMR simulation we can calculate the equilibrium positions \( m_{i,\text{eq},z} \) and precession cone angles \( \Theta_i \) and may thus predict the sign of \( V_{\text{ISH,AFM}} \), which we expect to reverse close to \( H_C \).

### 4.3.2 Thermal Voltages

As the MnF\(_2\) sample is driven resonantly, the intrinsic damping transfers energy from the absorbed microwave, via the magnetic system into phonons. In other words, AFMR is heating the sample. The temperature raise depends on the incident microwave power and the thermal contact of the sample to its surrounding. If parts of the sample are exposed to stronger microwave radiation, temperature differences between the sample ends might occur. The two silver glue contact | Pt strip interfaces, located at these ends on different temperature levels, would then give rise to a thermopower \( V_{\text{Th}} \). The line shape of \( V_{\text{Th}} \) would follow the absorption

\(^2\)The precession cone angles depend on the strength of the microwave absorption, determined among others, by the perpendicularity of \( \mathbf{m}_i \) and \( \mathbf{h}_{\text{rf}} \). This is unaffected by \( \mathbf{m}_i \rightarrow -\mathbf{m}_i \).
shape of the microwave radiation. A field inversion would not alter the generated temperature gradient and therefore no sign change of $V_{Th}$ would occur, which turns $V_{Th}$ into a candidate to explain the measured $V_{DC}$. In the following, we estimate the magnitude of the Seebeck voltage $V_{Th}$ in our setup.

The junction between the platinum strip and the silver glue attaching the gold wire has a difference of Seebeck coefficients of $\alpha \simeq 5 \, \text{V/K}$ at 100 K. As $V_{DC} \approx 1 \, \mu\text{V}$ in resonance, the required temperature difference between the two strip ends for a Seebeck voltage of this magnitude would be $\Delta T = V_{DC}/\alpha \approx 200 \, \text{mK}$. Reported ferromagnetic heating raises the sample temperature about $4^\circ - 6^\circ \text{K}$ per Watt incident microwave radiation [79][80], achieved in thermally well isolated magnetic samples. In our measurement, the incident microwave radiation is $\sim 10 \, \text{mW}$, hence, assuming a similar conversion efficiency, the heating would cause a temperature increase of $\sim 50 \, \text{mK}$. This value would reflect the temperature difference across the sample only if just one end was exposed to microwave radiation. Due to the symmetric mounting of the crystal with respect to the CPW, however, and, compared with [79], the higher thermal conductivity of MnF$_2$[81], which diminishes arising temperature differences within the sample more efficiently, the temperature differences between the contact sites due to anisotropic resonant heating should be significantly smaller than 50 mK.

As an example, a temperature difference of $\sim 10\text{mK}$ would cause a thermo-voltage of $V_{Th} \approx 50 \, \text{nV}$, which can not account for the observed DC signal of $V_{DC} \approx 1 \, \mu\text{V}$. Therefore, we conclude that thermo-power is not the main source of the observed $V_{DC}$ in resonance. A partial contribution of $V_{Th}$ to the observed signal may however not be ruled out.

### 4.3.3 Microwave Rectification

A third effect that may generate a DC voltage across the Pt strip at magnetic resonance is called microwave rectification [82]. In this picture, the microwave electric field drives an AC current $I(t) \propto \cos(\omega t)$ in the metallic layer, whose voltage is given by $V(t) = RI(t)$. Its DC component is found by the time average $\langle \rangle$ of $V(t)$, which, under the assumption that $R$ exhibits no time dependence, is $\langle V(t) \rangle = 0$.

If the sample, however, does exhibit an anisotropic magneto resistance, $R$ depends on the orientation of the magnetisation $\mathbf{m} = \mathbf{M}/M$ and $R \rightarrow R(\mathbf{m})$. If $\mathbf{M}$ is excited at resonance, its orientation becomes time dependent $\mathbf{m} \rightarrow \mathbf{m}(t)$ with precession frequency $\omega$ and $R$ develops a time dependence. Splitting $R(\mathbf{m}(t))$ into a constant part $R_0$ and a purely time varying part $R_t(\mathbf{m}(t)) \propto \cos(\omega t + \eta)$, with a phase offset

---

3According to [82] the Seebeck coefficients at 100 K are: $\alpha_{Pt} = -4.1 \, \text{V/K}$, $\alpha_{Ag} = 0.73 \, \text{V/K}$ and $\alpha_{Au} = 0.82 \, \text{V/K}$. The thermo-voltage of the gold|silver junction is hence negligible and the silver|platinum junction has a difference of $\alpha \simeq 5 \, \text{V/K}$. 

\( \eta \) to the microwave drive, we find a DC voltage as:

\[
V_{\text{rect}} = \left\langle R(m(t))I(t) \right\rangle = \left\langle R_0 I(t) \right\rangle + \left\langle R(t) I(t) \right\rangle \propto \left\langle \cos(\omega t + \eta) \cos(\omega t) \right\rangle = \frac{1}{2} \cos(\eta)
\]

The voltage arises from a homodyne mixing of the AC current with a resistance that exhibits the same time dependence. It depends on the phase between \( I(t) \) and \( R(t) \), \( \eta \). However, the value of \( \eta \) is not straightforward to obtain and involves a detailed investigation of the magnetic resonance condition, the Polder susceptibility tensor and the angular dependence of the respective magnetoresistance.

Rectification voltages arising from the anisotropic magnetoresistance (AMR) and anomalous Hall effects (AHE) have been investigated by \[82\][39][83]. It is found that the resulting \( V_{\text{rect}} \) might follow an absorption line shape without sign reversal under magnetic field inversion, making it a candidate for \( V_{\text{DC}} \). The exact line shape of \( V_{\text{rect}} \) depends, however, strongly on the position of the voltage probes with respect to the electromagnetic field and the magnetisation direction, which hinders the direct adoption of the results in the literature. Furthermore, AMR requires a ferromagnetic conducting layer, which is not present in the MnF\(_2\)|Pt sample. In fact, no magneto resistance has so far been reported in a heterostructure of an isolating antiferromagnet with a metallic layer. Recently a new type of magneto resistive effect has been observed in Pt layers on an isolating ferrimagnet—the spin hall magnetoresistance (SMR) \[50\]. As detailed in Chapter 2.4, its angle dependence is similar to the AMR and may be suitable to account for the observed DC voltage. The rectification voltage with SMR in the configuration of Fig. 4.1 is derived in Appendix B and given by:

\[
V_{\text{rect}} = L \cdot E_y = -L\Delta \rho J_y(t) |m_y(t)|^2 \left[ 1 + 2L \Delta \rho J_z(t)m_y(t) m_z(t) \right], \tag{4.4}
\]

with \( J_y(t) \), \( J_z(t) \) the AC current density components, \( \Delta \rho \) the SMR effect strength \(2.27\), \( m_y(t) \) and \( m_z(t) \) the magnetisation unit vector components and \( L \) the Pt strip length.

The solution of (4.4) and a prediction of the line shape of \( V_{\text{rect}} \) and its transformation behaviour under field inversion is, however, difficult to obtain for the present antiferromagnetic sample for the following reasons:

First of all, the treatment of two sublattice magnetisation vectors in the context of SMR theory is unknown. On the one hand, one could argue, that the net magnetisation \( m_{\text{tot}}(t) = m_1(t) + m_2(t) \) should be used in (4.4). This concept is successful in the SMR treatment of the ferrimagnet yttrium iron garnet \[50\]. On the other hand, a treatment of the individual magnetisations is conceivable, too, as the effect SMR is based on, is the spin transfer torque to a real magnetisation vector. In this case, a connection of the resistances of the individual magnetisations would enter (4.4).
Secondly, the exact expression of the Polder susceptibility tensor $\hat{\chi}$ is required to relate the phase of the magnetisation precession to the phase of the microwave field $\mathbf{h}_{\text{rf}}(t) \propto \mathbf{J}(t)$. As no analytical solution of the AFMR Landau-Lifshitz equations with incorporated $\mathbf{h}_{\text{rf}}(t)$ exists, the components of the Polder susceptibility tensor are unknown. The use of the ferromagnetic susceptibility tensor for one magnetisation may, however, be a good approximation, as indicated by the agreement of fitted and measured $U$ value in Chapter 3.4.2.1. The motion of the second magnetisation could then be assumed 180° phase offset as obtained for the unexcited eigenmotion of the AFMR (cf. Fig 3.9).

In summary, microwave rectification effects could be the origin of the observed DC voltage $V_{\text{DC}}$, although up to now, no magneto resistive effects were known in an antiferromagnetic isolator|metallic layer heterostructures. As the following Chapter shows, the resistance of the Pt film indeed depends on the magnetisation orientations of MnF$_2$, which makes a contribution of $V_{\text{rect}}$ to $V_{\text{DC}}$ probable.

The type of magneto resistance of the MnF$_2$|Pt stack and its angle dependence with respect to the magnetisations remains unknown. A promising candidate is the SMR, which has been reported to alter the resistance of a ferrimagnetic isolator|metallic layer structure. Using the angle dependence of the SMR, no final expression and thus no prediction of $V_{\text{rect}}$ could be given, mainly due to the so far unknown treatment of SMR with two sublattice magnetisations. To clarify the origin of $V_{\text{DC}}$, a better understanding of the magneto resistance of a MnF$_2$|Pt bilayer structure is required.

In conclusion, spin pumping voltages in MnF$_2$|Pt heterostructures could so far not be detected. The observed voltage $V_{\text{DC}}$, appearing in antiferromagnetic resonance, does not exhibit the sign change under field reversal, which is characteristic for spin pumping. Although the definite origin of $V_{\text{DC}}$ could not be identified, there are several indications that microwave rectification effects, possibly induced by a spin Hall magnetoresistance of our MnF$_2$|Pt bilayers, could generate such a DC voltage.
Chapter 5

Magnetoresistance of MnF$_2$|Pt Bilayers

In this chapter, triggered by the results of the spin pumping experiments, we discuss the magnetoresistance of three MnF$_2$|Pt heterostructures. More precisely, the longitudinal (sheet) resistance of the Pt thin film is measured as function of external magnetic field strength. In the following, we introduce the sample preparation and the experimental setup. Then the measurement results are presented and in the last section discussed.

5.1 Magnetoresistance Sample and Experimental Setup

Two different types of MnF$_2$|Pt bilayer samples are utilized for the magnetoresistance (MR) measurements. First, the samples of the spin pumping experiments can be reused without modification. Then, specifically for the MR measurements, Hall bars are prepared on both a MnF$_2$ and a BaF$_2$ crystal. BaF$_2$ has a similar chemical structure to MnF$_2$, but exhibits no magnetic order and should thus be suitable to provide a reference resistance measurement. In the following the sample preparations and the measurement setup of the two sample types are presented.

5.1.1 Pt-Strip MnF$_2$ Samples

The spin pumping samples are reused for the MR studies without any modifications. The experimental setup could be adapted effortlessly, without removing the sample from the cryostat. As depicted in Fig. 5.1, a Keithley 2400 Sourcemeter providing a bias current $I_{\text{bias}}$ is connected in parallel to the Keithley 2182A Nanovoltmeter that measures $V_{\text{DC}}$ as a function of magnetic field strength. Thus, two point resistance measurements are performed.

The two Pt strip samples are denoted MnF2#1 and MnF2#2. Their sample resistances are in the kΩ regime at room temperature, while the resistance of the current leads is only $\sim 20\,\Omega$, well justifying the two point resistance measurement. The bias current is $\sim 10\,\mu\text{A}$, preventing a heating of the sample.

As depicted in Figure 4.1, the external field configuration is the same as in the spin pumping experiments, with the $H_0$ ideally in the plane of the Pt thin film and
Figure 5.1: Measurement setup for the two point resistance measurements of the Pt strip samples. The sample mount and magnetic field configuration is analogous to the spin pumping experiments of Figure 4.1. A current source provides $I_{\text{bias}}$ while the voltmeter in parallel measures the voltage drop $V_{\text{DC}}$ across the Pt strip. The CPW is used to probe $\theta_H$. During the resistance measurements, however, no microwave radiation is applied.

![Measurement setup](image)

<table>
<thead>
<tr>
<th></th>
<th>$t_{\text{Pt}}$</th>
<th>$l_{12}$</th>
<th>$w_{\text{HB}}$</th>
<th>$R_{12} @20^\circ \text{C}$</th>
<th>$\rho_{12} @20^\circ \text{C}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>MnF$_2$</td>
<td>Pt</td>
<td>$\sim 7$ nm</td>
<td>1050 $\mu$m</td>
<td>80 $\mu$m</td>
<td>1380 $\Omega$</td>
</tr>
<tr>
<td>BaF$_2$</td>
<td>Pt</td>
<td>$\sim 7$ nm</td>
<td>1055 $\mu$m</td>
<td>73 $\mu$m</td>
<td>1600 $\Omega$</td>
</tr>
</tbody>
</table>

Table 5.1: Properties of the Pt Hall bars on BaF$_2$ and MnF$_2$ #3. $l_{12}$ is the distance between contact pad 1 and 2 and $w_{\text{HB}}$ the width of the Hall bar. The resistivity is calculated as $\rho_{12} = R_{12}w_{\text{HB}}t_{\text{Pt}}/l_{12}$.

parallel to $H_A$. Again, the coordinate system is introduced with $z \parallel H_A$, $x$ parallel to the platinum surface normal and $y$ along the Pt strip.

5.1.2 MnF$_2$|Pt and BaF|Pt Hall Bar Samples

For the second type of resistance measurement, as depicted in Figure 5.2, Hall bars are patterned onto both the MnF$_2$ crystal #3 and a BaF$_2$ substrate. The commercial BaF$_2$ substrate is cut into squares with $\sim 3$mm edge length. The 3 mm $\times$ 0.5 mm side of MnF$_2$ #3, which is parallel to the anisotropy axis, is diamond polished, analogous to the preparations of crystals #1 and #2. Further grinding with water based polish is, again, not carried out. Then a positive photo resist is spin coated onto both crystals. Photo resist edge walls turned out to be an issue for the Hall bar patterning on MnF$_2$ #3, due to the small distance of 0.5mm between opposite edges. This is overcome, by shaping smooth transitions to the edges of the crystal using plastic modeling mass. With this, the photo resist could flow off during spinning and the resulting resist layer is flat enough to structure the Hall bars.

With both BaF$_2$ and MnF$_2$ crystals on the same vacuum sample mount, platinum with a thickness of $\sim 7$ nm is deposited using electron beam evaporation. Figures 5.2 (b) and (c) show microscope images of the two Hall bars after the lift off process. As presented in Table 5.1, the Hall bars agree well in dimensions and their longitudinal resistivities $\rho_{12}$ are comparable, too. Interestingly, $\rho_{12}$ of Pt on the highly polished
BaF$_2$ substrate slightly exceeds that of Pt on the nominal rougher, diamond polished MnF$_2$.$^1$

The crystals are then glued onto a plastic block using GE varnish (cf. Fig. 5.2 (a)). Alongside the crystals, two supporting contact pads are attached. From the contact pads, thin aluminium wires are bonded towards the Hall bars. Unfortunately, on both samples the bonding process towards the crystal was unsuccessful. Eventually, contacting had to be done “by hand” and the bonds were carefully attached to the Hall bars with silver glue. In the end, copper cables leading to an external breakout box are soldered onto the contact pads and the whole plastic block is mounted onto a CPW, as seen in Figure 5.2 (d). In this way, the dispersion of the AFMR could again be used to determine the crystal orientation $\theta_H$. Both Hall bars are connected via Cu leads to Keithley 2400 current sources (contact pads 1 & 4) and Keithley 2182 Nanovoltmeters (contact pads 2 & 3) to perform four-point longitudinal resistance measurements. The configuration of the external field $\mathbf{H}_0$ and

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$^1$Possible causes may be unequal Pt thicknesses, caused by different positions over the Pt evaporation plume and different interface scattering properties of the two materials.
5.2 Experimental Results

5.2.1 MnF$_2$|Pt and BaF$_2$|Pt Hall Bar Measurements

We first present the results of the MnF$_2$|Pt and BaF$_2$|Pt Hall bars resistance measurement at $T = 4$ K. Figure 5.3 depicts the obtained resistance of MnF$_2$|Pt in orange and BaF$_2$|Pt in purple as a function of external field. In small external fields (0 T → ±3 T), both Pt thin films exhibit a quadratic positive MR, in agreement with the literature [84]. With increasing field strength, the MR of MnF$_2$|Pt gradually becomes linear, while the MR of BaF$_2$|Pt further bends, assuming a logarithmic behaviour for fields larger than $\sim \pm 7$ T. On top of this MR, the Pt resistance on MnF$_2$ exhibits an additional feature at the spin flop field strength\footnote{Such a resistance change at $H_C$ could be caused by the enhanced magnetic induction originating from the induced net magnetisation $\mu_0 M_{\text{net}}$. From Fig. 3.7 we find induced magnetisation values of $\mu_0 M_{\text{net}} \approx 10$ mT. The observed Pt MR linearised over the whole field stretch is $\sim 0.2 \Omega/15$ T. $\mu_0 M_{\text{net}}$ thus can only induce an additional resistance increase of $\sim 0.1$ m$,\Omega$, which} $\mu_0 H_0 = 9.45$ T.
We conclude that the resistance of $\text{MnF}_2|\text{Pt}$ is composed of two different contributions as

$$R = R_{\text{Pt, intrinsic}} + R_{\text{Pt, MnF}_2}.$$  \hspace{1cm} (5.1)

$R_{\text{Pt, intrinsic}}$ is the intrinsic MR contribution of the Pt film, e.g. the magnetoresistive effect that a Pt film itself would exhibit under the influence of an in-plane magnetic field, applied transversely to the current direction. $R_{\text{Pt, MnF}_2}$ is the influence of the magnetic configuration of MnF$_2$ on the Pt resistance and is particularly prominent in the vicinity of $H_C$.

The measurement of the $\text{BaF}_2|\text{Pt}$ Hall bar was intended to provide a reference resistance measurement, which solely includes the intrinsic platinum resistance $R_{\text{Pt, intrinsic}}$. With this, it should have been possible to separate the antiferromagnetic resistance contribution $R_{\text{Pt, MnF}_2}$ from the MnF$_2|\text{Pt}$ measurement. However, as Figure 5.3 depicts, the resistance curves of the two samples differ significantly, even for field strengths far below $H_C$, where no influence of the MnF$_2$ is expected. Also, the overall line shapes do not coincide, which leads us to the conclusion that the intrinsic magnetoresistances on the two substrates are not comparable. Thus, with the given data set, it is not possible to separate the antiferromagnetic contribution $R_{\text{Pt, MnF}_2}$ from $R_{\text{Pt, intrinsic}}$ via resistance measurements of MnF$_2|\text{Pt}$ and BaF$_2|\text{Pt}$.

### 5.2.2 MnF$_2|\text{Pt}$ Magnetoresistance Measurements - Comparison of Measured Field Dependencies

In addition to the Hall bar sample $\text{MnF}#3$, magnetoresistance measurements were also conducted on the Pt strip samples $\text{MnF}#1$ and $\text{MnF}#2$ in the same external field configuration. With $\text{MnF}#2$, two measurement series were carried out. Within these series, measurements were taken at different rotation angles of the dip stick within the cryostat. Due to the imperfect alignment of the dip stick axis and the superconducting magnetic field, the misalignment of external field and crystalline anisotropy axis $\theta_H$ was thereby modified by $\sim \pm 0.2^\circ$. All MR measurements within a measurement series result in similar curve shapes and the only difference is an increase of the strength of the resistance change at $H_C$ with better external field alignment $\theta_H$.

Figure 5.4 presents representative examples of the two $\text{MnF}#2$ measurement series, along with the measurements of $\text{MnF}#3$ and $\text{MnF}#1$. The obtained results of $\text{MnF}#3$ and $\text{MnF}#2$ are robust over several magnetic sweeps $0 \text{T} \rightarrow 15 \text{T} \rightarrow -15 \text{T} \rightarrow 0 \text{T}$ and the presented MR curve is the average of five consecutive sweeps. The observed MR of the $\text{MnF}#1$ varies within subsequent sweeps, such that we is too small to explain the present resistance variation of $\sim 10 \text{ m}\Omega$, cf. Fig. 5.5. Furthermore, a resistance increase by $\mu_0M_{\text{net}}$ could not account for the observed different signs of the resistance variation at $H_C$ in Fig. 5.3.
Figure 5.4: Comparison of magnetoresistance measurements obtained for three different MnF$_2$|Pt samples. The resistance is normalized as $\frac{[R(H) - R(0)]}{R(0)}$. The presented data is averaged over five consecutive magnetic field sweeps, except for MnF#1, for which averaging is not viable, as consecutive sweeps deviate significantly. The alignment $\theta_H$ of the external field with respect to the MnF$_2$ anisotropy axis is obtained from AFMR measurements.

show individual sweeps. The MR values are normalised$^3$ as $\frac{[R(H_0) - R(0)]}{R(0)}$. The misalignment $\theta_H$ is extracted from the simulation of the respective AFMR dispersion.

The MR properties of the MnF#3 Hall bar sample (orange curve) have been discussed in the previous section, where two different resistance contribution $R_{\text{Pt, intrinsic}}$ and $R_{\text{Pt, MnF$_2$}}$ are identified. These two contributions are also observable in the resistance measurements of the MnF#2 (cyan, blue). Both MnF#2 measurements exhibit matching intrinsic resistance contributions $R_{\text{Pt, intrinsic}}$, which resemble the $R_{\text{Pt, intrinsic}}$ shape of MnF#3. The resistance contributions $R_{\text{Pt, MnF$_2$}}$ of the measurements MnF#2 and MnF#3, however, differ strikingly in both the shape and the sign of the resistance modifications at $H_C$. The resistance measurement with the best alignment of $H_0$ with respect to $H_A$—the measurement of the Pt strip sample MnF#1 (green)—furthermore adds to the inconsistent behaviour of $R_{\text{MnF$_2$}}$. Here, the measured resistance at $H_C$ changes in consecutive sweeps. In addition, the

$^3$MnF#1: $R(0 \, \text{T}) = 732 \, \Omega$, MnF#2 (blue): $R(0 \, \text{T}) = 1030 \, \Omega$, MnF#2 (cyan): $R(0 \, \text{T}) = 1037 \, \Omega$, MnF#3: $R(0 \, \text{T}) = 310 \, \Omega$
resistance curve is no longer smooth around $H_C$, but exhibits discontinuities and fluctuations. The modification of the resistance at $H_C$ seems to increase in strength with better alignment $\theta_H$, as observed for the four depicted measurements and for the two measurement series of $MnF\#2$.

### 5.2.3 Separation of the Antiferromagnetic Magnetoresistance Contribution

As shown in Section 5.2.1, it is not possible to extract the antiferromagnetic contribution $R_{Pt,MnF_2}$ using the measurement of the BaF$_2$|Pt Hall bar as a reference. We may, however, gain an approximation of the field dependence of $R_{Pt,MnF_2}$ by a linear Taylor expansion of $R$ from lower field values, as discussed in the following.

The orientation of the antiferromagnetic magnetisation vectors is relatively unaffected by a change of external field $H_0$ below $H_C$. Therefore, we now assume that $R_{Pt,MnF_2}$ is constant with the value $R_{Pt,MnF_2,0}$ for $H_0 < H_C$. Hence the slope of $R$ below $H_C$ is solely determined by the intrinsic contribution $R_{Pt,intrinsic}$. In the vicinity of $H_C$, $R_{Pt,intrinsic}(H_0)$ may thus be Taylor expanded to a first degree, using the slope determined from the measured resistance $R$:

$$R_{Pt,intrinsic}(H_0) \approx R(H_a) - R_{Pt,MnF_2,0} + \frac{\partial R}{\partial H_0} \bigg|_{H_a} (H_0 - H_a), \quad (5.2)$$

with Taylor expansion point $H_a \ll H_C$. For the constant resistance value $R_{Pt,intrinsic}$, we exploited the relation (5.1). Note that the value of $R_{Pt,intrinsic}$ is composed of the measured resistance $R(H_a)$ and the unknown MnF$_2$ contribution $R_{Pt,MnF_2,0}$ below $H_C$. This leaves an unknown offset $R_{Pt,MnF_2,0}$ to the approximated external field dependence of the intrinsic resistance contribution $R_{Pt,intrinsic}(H_0)$.

Figures 5.5 (a-c) depict the obtained, linearly approximated $R_{Pt,intrinsic}(H_0)$ (red curves) for the averaged MR measurements of Fig. 5.4. The presented data is again normalized as $[R(H) - R(0)]/R(0)$. The slope of all three $R_{Pt,intrinsic}(H_0)$ curves is obtained by fits to $R(H_0)$ between 5 T and 7 T. The magnetic field axis of the graphs are restricted to a range, where the linear approximation of $R_{Pt,intrinsic}(H_0)$ appears to be valid, although it may not be ruled out that deviations are occurring already within the presented region.

Using the approximated intrinsic contribution $R_{Pt,intrinsic}(H_0)$, $R_{Pt,MnF_2}(H_0)$ may be calculated as $R_{Pt,MnF_2}(H_0) = R(H_0) - R_{Pt,intrinsic}(H_0)$. The result is presented in Figure 5.5 (d-f). Note that due to the unknown constant offset of the Taylor approximated $R_{Pt,intrinsic}$, only the field variation of $R_{Pt,MnF_2}$ can be determined, and not its absolute value, which is indicated as $\Delta_H R_{Pt,MnF_2}$.

The obtained external field dependencies of $\Delta_H R_{Pt,MnF_2}$ are inconsistent. While the $MnF\#3$ measurement (Fig. 5.5 (d)) shows an increase of the resistance by a
Figure 5.5: Panels (a-c) show the magnetoresistance curves of three of the MnF$_2$|Pt measurements of Fig. 5.4 (squares) along with the linearly approximated intrinsic platinum magnetoresistance $R_{\text{Pt,intrinsic}}$ (red lines). Panels (d-f) depict the field dependent part of the MnF$_2$ attributed resistance contribution $R_{\text{Pt,MnF}_2}$, denoted $\Delta_H R_{\text{Pt,MnF}_2}$. $\Delta_H R_{\text{Pt,MnF}_2}$ is extracted by a subtraction of the approximated intrinsic contribution $R_{\text{Pt,intrinsic}}$ from the measured resistance $R$. All resistances are again normalized by dividing with $R(0)$. 

![Image of the figure showing magnetoresistance curves and field dependence of MnF$_2$ and Pt contributions](image-url)
Figure 5.6: Illustration of the SMR effect with MnF$_2$|Pt bilayers. The charge current $J_{c,y}$ flowing in the platinum generates a perpendicular spin current $J_{s}^\text{SHE}$ with spin polarisation $s$, due to the spin Hall effect. Depending on the orientation of the sublattice magnetisations $M_i$ with respect to $s$, $J_{s}^\text{SHE}$ is either absorbed or reflected at the MnF$_2$|Pt interface, which in turn affects the longitudinal resistance of $J_{c}$. The orientation of $M_i$ depends on the strength and direction of the external magnetic field $H_0$. The uniaxial easy axis anisotropy $H_A$ is again assumed parallel to $z$ (not depicted for clarity).

constant value at $\sim H_C$, one MnF#2 measurement (Fig. 5.5 (e)) exhibits a decrease. The second MnF#2 measurement (Fig. 5.5 (f)) again shows a resistance reduction, in this measurement, however, restricted only to the proximity of $H_C$.

5.3 Discussion

A possible origin of the antiferromagnetic resistance contribution is the spin Hall magnetoresistance (SMR). As discussed in Chapter 2.4, the SMR predicts that the resistance of a metallic thin film varies with the magnetisation orientation of an adjacent (isolating) ferromagnet, depending on the possibility to transfer spin momentum to the ferromagnet via the spin transfer torque (STT). In our configuration, depicted in Figure 5.6, the SMR longitudinal resistance of $J_{c}$ is sensitive to the $z$ component of the magnetisation vectors (2.28). Using the AFMR simulation, we can predict this magnetisation component for both sublattice magnetisations $M_i$, which enables us to test, if the observed resistance change at $H_C$ is explainable by the SMR theory.

Theoretical studies [85, 86] suggest that spin transfer torque of antiferromagnetic systems is governed by the transfer of angular momentum to the individual magnetisations, not to the net magnetisation. Thus, we treat the resistance contributions of the individual magnetisations separately and, based on Matthiessen’s rule [87], take the total resistance as the sum of the two individual sublattice resistance contributions. Extending (2.28) in this manner we propose a longitudinal
Magnetoresistance of MnF$_2$|Pt Bilayers

**Figure 5.7:** The antiferromagnetic resistance contribution $\Delta R_{\text{Pt, MnF}_2}$ of the MnF$_2$|Pt resistance and suggested SMR field dependencies (red lines). Additionally in (c), the negative $xy$ component of $m_{\text{tot}}$ is shown. For every graph, the SMR resistance is calculated using the AFMR simulation-obtained magnetisation components $m_{i,z}$.

antiferromagnetic SMR of the form:

$$R_{\text{SMR, } m_{\text{ind}}} = R_0 - \Delta R_{\text{ind}}m_{1,z}^2 - \Delta R_{\text{ind}}m_{2,z}^2$$  \hfill (5.3)

$R_0$ is a constant resistance contribution, $\Delta R_{\text{ind}}$ the SMR effect strength from one magnetic sublattice and $m_{1,z}$, $m_{2,z}$ are the $z$ components of the magnetisations’ unit vectors $m_i$ in the coordinate system of Fig. 5.6. Taking $\Delta R_{\text{ind}} > 0$ from SMR theory, this resistance dependence suggests that in the parallel alignment $m_i||H_0$ ($H_0 < H_C$, $m_{i,z} \approx 1$) the resistance is smaller than the resistance in the spin flop configuration ($H_0 > H_C$, $m_{i,z} \rightarrow 0$).

For comparison, we also introduce the SMR resistivity expected from spin transfer torque to the total magnetisation $m_{\text{tot}} = m_1 + m_2$,

$$R_{\text{SMR, } m_{\text{tot}}} = R_0 - \Delta R_{\text{tot}}m_{\text{tot},z}^2.$$  \hfill (5.4)

Since the total magnetisation is relevant for $R_{\text{SMR, } m_{\text{tot}}}$, (5.4) predicts a decrease of resistance above $H_C$, due to the induced net magnetisation in $z$ direction ($m_{\text{tot},z}^2 > 0$ for $H_0 > H_C$). Again $\Delta R_{\text{tot}}$ is assumed positive.

Figure 5.7 depicts the external field dependence of $R_{\text{SMR, } m_{\text{ind}}}$ and $R_{\text{SMR, } m_{\text{tot}}}$ in comparison with the measured antiferromagnetic resistance contribution $\Delta R_{\text{Pt, MnF}_2}$ of the three resistance measurements of Figure 5.5 (d-f). The value of $\Delta R_{\text{ind}}$ and $\Delta R_{\text{tot}}$ of each measurement were adjusted to best match the respective change of resistance at $H_C$. While the resistance of Figure 5.7 (a) is in accordance with the SMR curve expected from individual magnetisations, (5.3), Figure 5.7 (b) is rather described by $R_{\text{SMR, } m_{\text{tot}}}$ (Eq. 5.4). Finally the resistance of Figure 5.7 (c) is in disagreement with both SMR field dependencies. Therefore, we conclude that none
of the proposed SMR resistance dependencies is suitable to describe the observed magnetoresistance consistently.

Figure 5.7 (c) indicates that another contribution might also be of importance, the component of \( m_{\text{tot}} \) in the plane perpendicular to \( z \), denoted \( m_{\text{tot},xy} \). Please note that we have experimentally no control over the polar angle \( \varphi_H \) of the external magnetic field (cf. Fig. 5.6). As the AFMR dispersion is invariant under a variation of \( \varphi_H \), too, we can not determine, if the magnetisations align parallelly (\( ||y|| \)) or perpendicularly (\( ||x|| \)) to the platinum film in the spin flop configuration. Furthermore, this orientation is likely to change between different sample mounts, resulting in varying orientations for the presented measurements\(^5\). If—in contrast to SMR theory—the resistance of MnF\(_2\)|Pt depends on the \( x \) or \( y \) magnetisation components, different \( \varphi_H \) values in the different measurements could explain the observed \( R_{\text{Pt,MnF}_2} \).

A second possible explanation for the behaviour of \( R_{\text{Pt,MnF}_2} \) would be surface states. As the MnF\(_2\) crystals were covered \textit{ex situ} with Pt, dissimilar interface conditions could lead to the varying resistance dependencies. However, the measurements of MnF\#2 show different resistance line shapes with the same MnF\(_2\)|Pt interface, which makes an interface issue as the origin of the inconsistent \( R_{\text{Pt,MnF}_2} \) magnetic field dependence improbable. Yet, due to the small number of samples, it can not be totally excluded.

In summary, we observe a clear variation of the resistivity of a MnF\(_2\)|Pt bilayer caused by the reorientation of the antiferromagnetic sublattice magnetisations. To our knowledge, this is the first observation of a magnetisation-dependent sheet magnetoresistance in a heterostructure consisting of an electrically isolating antiferromagnet and a normal metal. The sign of the antiferromagnetic resistance variation and its form is qualitatively different in different measurement series. For a better understanding of the observed magnetoresistive effect, the experimental control of the sublattice magnetisation orientations with respect to the Pt layer is crucial. Such control could be gained by a rotation of the external field with respect to the sample, so called angle dependent magnetoresistance (ADMR) measurements. These are described in detail in the Outlook of this thesis, Chapter 6.3.1.

\(^4\)A net magnetisation in the \( x-y \) plane is induced in the vicinity of the spin flop field, when the reorientation of the magnetisations is occurring.

\(^5\)Note that within the two measurement series of MnF\#2, where the dip stick is rotated within the VTI, \( \varphi_H \) of Fig. 5.6 is not varying drastically. As the axis of rotation, i.e. the axis of the dip stick, is most likely better aligned with the external field than with the anisotropy axis, the rotation of the crystal occurs around \( \sim \mathbf{H}_0 \) and not \( \mathbf{z} \), leaving \( \varphi_H \) unchanged. This would explain, why the resistance curves are consistent within the measurement series, but not across different sample mountings.
Chapter 6

Summary and Outlook

In this thesis both the spin dynamics in the model antiferromagnet MnF$_2$ and the impact of the spin dynamics onto the electronic charges in an adjacent metallic platinum film have been studied. In the following we summarize the obtained results and discuss their implications on future experiments, with particular emphasis on experiments that could clarify the angular dependence of the observed, novel magnetoresistive effect in MnF$_2$|Pt hybrids.

6.1 Antiferromagnetic resonance of MnF$_2$

In Chapter 3 we presented broadband antiferromagnetic resonance (AFMR) measurements of three MnF$_2$ single crystal samples. Within the experimentally accessible frequency window of $f_{mw} \leq 26.5$ GHz, we could resolve the lower frequency AFMR mode, including spin wave excitations, in a field region of 8.7 T to 10.5 T. Since it was not possible to explain the observed AFMR dispersion using the established AFMR theory, we developed a simulation going beyond the current state of the art, based on the free energy ansatz of Smit & Suhl [43]. This simulation now allows to calculate the eigenfrequencies of the antiferromagnet without restrictions to the strength and the orientation of the applied magnetic field. We find excellent agreement between the simulated and experimental AFMR dispersions for all of our AFMR measurements, by adjusting only a single parameter: The misalignment between the external field and the axis of uniaxial anisotropy.

Moreover, we demonstrated that the resonance line shape exhibits both absorptive and dispersive parts. This enables a fit of the measurement data to the expected susceptibility of a (ferromagnetic) Polder tensor. The dependence of the extracted resonance line width on the external field strength is in good agreement with the simulated line width dependence, which furthermore corroborates the power of our AFMR simulation approach.

Turning towards conceivable improvements of the antiferromagnetic resonance experiment, we find that these are mostly on the experimental side. The primary
limitation is the restriction of microwave frequencies to 26.5 GHz compared with an AFMR excitation spectrum of 500 GHz (cf. Figs. 2.7 and 3.8). However, these frequency limitations are hard to push and would require great efforts, both financially (upgrade of the VNA) and technically (e.g. microwave transmission losses of the CPW and the cables).

Furthermore, an active control of the crystal orientation with respect to the external field would be highly desirable to manipulate the AFMR dispersion and the sharpness of the spin flop transition. The control of the alignment during operation in the cryostat could be achieved by two means. Firstly, the external field could be rotated to collinearity with the crystalline anisotropy axis, using a vector magnet. While this technique is very elegant, a vector magnet with the required field strengths (≥10 T) is not readily accessible to us. Secondly, the crystal orientation could be manipulated by means of a piezoelectric nanopositioning system. As this significantly increases the size of the sample mounting, it would no longer fit into our VTI. Still, this concept appears much more viable. In particular, a dilution refrigerator inset fitting into our 17 T magnet is available at the Walther–Meißner–Institute. This inset features significantly larger mounting space.

Using the dilution refrigerator with the nanopositioning system, it would be possible to study the line width of the uniform mode of MnF₂ at millikelvin temperatures. This is particularly attractive, as Kotthaus and Jaccarino [88] report a drastic reduction of line width with decreasing temperature, but were limited experimentally to temperatures above 1.5 K. A nanopositioning system is required for this study as every small misalignment will result in a severe broadening of the line width, due to the excitation of the spin wave spectrum [25].

### 6.2 Antiferromagnetic Spin Pumping

In Chapter 4, we investigated the high frequency interaction of the antiferromagnetic moments with the free electrons of a platinum thin film deposited on top of the MnF₂. We find a DC voltage in the μV range occurring only in antiferromagnetic resonance. The field dependence of the voltage follows that of the AFMR absorption, including the distinct modes of the spin wave spectrum. As the voltage sign is unaltered by a reversal of the external field direction, spin pumping is excluded as the main source of this voltage. An upper boundary for the size of the spin pumping voltage in our setup with incident microwave power of 13 mW can be set to $V_{ISH} < 200 \text{nV}$, as detailed in Section 4.3.1.

Turning towards other potential origins of the DC voltage, two further effects were considered: A potential Seebeck voltage due to anisotropic heating of the sample in resonance appears unlikely due to its small effect size. Instead, microwave rectification—the resonant mixing of a microwave induced AC current in the Pt film
6.3 Resistance of MnF$_2$|Pt heterostructures

with a resistivity that varies with the same frequency—is a plausible origin of the voltage. It requires an anisotropic magnetoresistance [82, 89], which is indeed expected to be present in our MnF$_2$|Pt samples, cf. Chapter 5.

The fact, that we did not detect spin pumping voltages does not mean that spin pumping is not present in antiferromagnets. To be able to observe antiferromagnetic spin pumping, a measurement configuration has to be identified, in which rectification effects are not occurring. In microwave cavity spin pumping experiments, rectification is suppressed if the sample is centered in the resonator, exactly in the node of the microwave electric field [90, 39]. Whether or not analogous improvements are possible for CPW-AFMR spin pumping experiments, depends on the exact angular dependence of the magnetoresistance causing the microwave rectification. We thus urge a detailed understanding of the observed MnF$_2$|Pt magnetoresistance to conclude if the spin pumping mechanism is also existing in antiferromagnets.

The discussion of antiferromagnetic spin pumping (Appendix A) might also affect the treatment of spin pumping in ferrimagnets (e.g. yttrium iron garnet), for which, in the established theory, the spin current is considered to be emitted by a single (net) magnetisation vector. Our model suggests that the two sublattice magnetisations both contribute, resulting in a more complex picture. Further investigations will be required to clarify if the sublattice structure of the ferrimagnet would modify the spin pumping formulae and if additional effects (e.g. the ferrimagnetic spin flop) are influencing the spin pumping voltage.

6.3 Resistance of MnF$_2$|Pt heterostructures

In Chapter 5, we presented the resistance measurements of three MnF$_2$|Pt bilayer samples. In all samples we find that the resistance exhibits an additional modification at the spin flop field on top of an intrinsic Pt magnetoresistance contribution. To our knowledge, this is the first report of such a variation of metallic resistance upon magnetisation reorientation in an adjacent isolating antiferromagnet.

Although the measurement configuration is kept as reproducible as possible between renewed sample mountings, the measured magnetoresistance variations at $H_C$ are qualitatively different: The resistance in the spin flop configuration is reduced for two measurements, increased for one, while for the best aligned sample, the resistance dependence at the critical field is different in consecutive magnetic field sweeps. Due to the contradictory results, an explanation of the resistance on the base of the current data is not possible. It should be noted though, that the recently discovered spin Hall magnetoresistance (SMR), which successfully describes the magnetoresistance of isolating ferrimagnet | platinum bilayers might be appli-
cable in the antiferromagnetic case as well.

Our magnetoresistance experiments in MnF$_2$|Pt leave many open questions. While the resistance modifications at $H_C$ are unambiguously present, their differing behaviours might indicate that another, so far unregarded factor influences the resistance. In our setup, we have no control over the magnetisation directions in the plane perpendicular to the anisotropy axis above $H_C$. This means, that the magnetisations in the spin flip configuration may align parallel with the Pt film or be oriented perpendicular to it, which could lead to different resistance values upon different sample mountings. To clarify the angular dependence of the antiferromagnetic resistance contribution, and to gain a better control over the magnetisation direction, we suggest to conduct angle dependent magnetoresistance (ADMR) measurements, as discussed in the following.

6.3.1 Angle Dependent Magnetoresistance Measurements

Resistance measurements as a function of external magnetic field orientation (ADMR measurements) are a powerful tool to investigate magnetoresistances. The external magnetic field is kept at constant field strength, while the orientation between sample and external field is changed stepwise and the corresponding resistance of the specimen is measured. For each magnetic field orientation, the (known) free energy surface of the sample determines the magnetisation orientation, such that one may thus acquire the resistance dependence on the magnetisation orientation.

Figure 6.1 presents possible ADMR measurements of our MnF$_2$|Pt samples. First notice—as depicted in Figure 6.1 (a) for a rotation in the $x−y$ plane—that a rotation for a field strength $H_0$ below the spin flop field $H_0 < H_C$ will not result in a variation of the orientation of $M_1$ and $M_2$. Therefore, no variation of the resistance caused by the antiferromagnet should occur. The same is expected for rotations in the $x−z$ and $y−z$ planes for $H_0 < H_C$, except that there is a resistance contribution from the conventional Hall effect of Pt.

The situation changes for external field strengths above $H_C$, where the magnetisation vectors align perpendicularly to the external field, within the plane of $H_0$ and $H_A$. Figure 6.1 (b) depicts this situation for a rotation of the external field in the $y−z$ plane. The magnetisations $M_1$ and $M_2$ follow the external field and may thus be aligned either along $z$ (first frame) or $y$ (second frame). No conventional Hall effect is expected for a field rotation within the film plane. Therefore, a rotation of the external field in the $y−z$ plane for $H_0 > H_C$ would allow the detection of $m_{i,y}$ and $m_{i,z}$ resistance dependencies. Analogously, a rotation in the $x−z$ plane (cf. Fig. 6.1 (c)) would yield information about $m_{i,x}$ and $m_{i,z}$ dependencies, superimposed by the conventional Hall effect. Finally, the rotation of external field in the $x−y$ plane, perpendicularly to the axis of crystalline anisotropy (cf. Fig. 6.1 (d)) should
6.3 Resistance of MnF$_2$|Pt heterostructures

Figure 6.1: Different rotation planes for ADMR measurements on MnF$_2$|Pt heterostructures. For external magnetic field strengths $H_0$ below the spin flop field $H_C$ (a), the magnetisations $M_1$ and $M_2$ align with the axis of uniaxial anisotropy $H_A$. For $H_0 > H_C$ (b-d) the orientation of $M_1$ and $M_2$ may be controlled by the external magnetic field $H_0$ in the depicted manner.
not affect the orientation of the magnetisation directions significantly, at least in our field range ($\mu_0 H_0 \leq 17 \text{T}$). Therefore, we would expect no major resistance variation by the magnetisations of MnF$_2$, but predominantly a conventional Hall contribution$^1$.

The combination of the three ADMR measurements with different rotation planes would allow the separation of conventional Hall effect and the resistance contribution connected to the magnetic structure of MnF$_2$. Thus ADMR experiments could clarify the origin and angular dependence of the observed MnF$_2$|Pt magnetoresistance.

To summarize this thesis, we could demonstrate that the spin dynamics of an electrically insulating antiferromagnet does influence the charge transport in an adjacent metallic film. The antiferromagnetic resonance simulation, developed in the framework of this thesis allows a quantitative description of the spin dynamics of exchange coupled sublattice magnetisations. The novel magnetoresistive effect opens up a possibility to interact with the sublattice magnetisations via conduction electrons. Further experiments are, required though, to reveal the microscopic mechanisms of this magnetoresistance.

We think that this thesis gives an impression of the added complexity, but also of the accompanied opportunities of a second sublattice magnetisation of an antiferromagnet. We therefore hope that it encourages further investigations of spintronic effects that manipulate two magnetisations vectors in one material.

$^1$While the presented discussion of the field rotation is correct, it is experimentally not achievable, as the rotation plane will always be slightly tilted with respect to the uniaxial anisotropy. Therefore, in Fig 6.1(b), when the external field crosses the $x-z$ plane, the external field vector will have an $x$ component, which will cause an orientation of the magnetisations along $+x$ and $-x$. This reorientation is only occurring in the vicinity of parallel $H_A$ and $H_0$. Therefore, it does not disprove the general reasoning of the ideal rotation planes. Also note that, while the given argumentation is of qualitative character, a quantitative prediction of the $x$, $y$ and $z$ components of the magnetisations for a given external field is possible using the simulation and its extracted MnF$_2$ properties of our samples.
Appendix A

Expected Antiferromagnetic Spin Pumping Voltage

Although the observed DC voltage of the spin pumping experiments may not be attributed to spin pumping itself (cf. Ch. 4.3.1), we still want to discuss some of the implications of the antiferromagnetic spin pumping equation 4.2, which are also of importance in a discussion of the observed difference of \( V_{DC} \) under field inversion, which is maybe caused by spin pumping.

In the qualitative discussion of spin pumping, one tends to underestimate the influence of the cone angles \( \Theta_i \) on \( V_{ISH} \). A typical assumption would be, that for fields \( H_0 < H_C \), the generated spin currents of the individual magnetisations would more or less compensate each other due to their antiparallel spin polarisation. Above \( H_C \), however, both magnetisation vectors have a component along \( +z \), the spin current of the two magnetisations would not cancel and thus a DC voltage may be detected. In the following, we show that such a simple argumentation is wrong and that the difference of the precession cone angles \( \Theta_i \) has a large influence on \( V_{ISH} \).

Chapter 3.3.2.3 shows, that the cone angles, or, which suggest is the more significant quantity for spin pumping, the areas \( A_i \) of the ellipses encircled by the magnetisation precessions, differ indeed for the unperturbed system, i.e. the eigenmotions of the magnetisation vectors. Introducing the precession area ratio \( \epsilon = A_2/A_1 \), we can rewrite the spin pumping voltage \( V_{ISH} (2.26) \) for the antiferromagnetic case in our configuration (cf. Ch. 4.3.1) as:

\[
V_{ISH,AFM} = V_{ISH,M_1} + V_{ISH,M_2}
\]

\[
= \frac{e \left[ \alpha_{SH} \lambda_{SD} \tanh \left( \frac{N}{2\lambda_{SD}} \right) \right]}{\sigma_F t_F + \sigma_N t_N} \left[ t_{g_{\uparrow \downarrow}} f_{MW} L \cdot \left( m_{1,eq,x} A_1 + m_{2,eq,x} A_2 \right) \right] \quad (A.1)
\]

\[
= \frac{e \left[ \alpha_{SH} \lambda_{SD} \tanh \left( \frac{N}{2\lambda_{SD}} \right) \right]}{\sigma_F t_F + \sigma_N t_N} \left[ \epsilon \cdot m_{2,eq,x} A_1 \right] \quad (A.2)
\]

\[
= \frac{e \left[ \alpha_{SH} \lambda_{SD} \tanh \left( \frac{N}{2\lambda_{SD}} \right) \right]}{\sigma_F t_F + \sigma_N t_N} \left[ \epsilon \cdot m_{2,eq,x} \right] A_1 \quad (A.3)
\]
Here, we replaced the expressions $P \sin^2 \Theta_i$ with the precession areas $A_i$. All the pre-factors are assumed to be equal for the spin currents generated from the two sublattice magnetisations. Only the equilibrium positions $m_{i,\text{eq},x}$ and the precession areas $A_i$ are expected to differ for the two sublattices. The factor to the right side of (A.3) determines the strength and sign of $V_{\text{ISH,AFM}}$ as a function of external field. Using our simulation, we may predict the field dependence of the different contributions, which is presented in Figure A.1 for two different alignment values $\theta_H$.

Figure A.1 (a) depicts the $z$ component $m_{i,\text{eq},z}$ of the equilibrium position for the two magnetisation vectors as a function of external field. $m_{i,\text{eq},z}$ determines the $z$ components of the spin polarisations of $J_{\text{s,pump}}^i$ that our sample contacting along $y$ is sensitive to (cf. 4.1). $m_{i,\text{eq},z}$ exhibits the expected spin flop behaviour, with antiparallel alignment along $\pm z$ below $H_C$ and a slight bending of both magnetisations towards $+z$ above $H_C$. The abruptness of the transition is reduced with increasing misalignment $\theta_H$.

Figure A.1 (b) presents the ratio of the precession areas $\epsilon = A_2/A_1$ for the unexcited motion, obtained from our simulation. As discussed in Chapter 3.3.2.3, the magnetisation oppositely directed to the external field $H_0||+z$ is stronger deflected for the lower AFMR mode. This is $M_1$ in our case, resulting in $\epsilon < 1$ for all external field strengths. Below $H_C$, the ratio is almost constant, while above $H_C$ it exhibits more variation. For $\theta_H = 0.5^\circ$, $A_2 \to 0$ for $\mu_0 H_0 \to 11.5 \text{T}$, because the magnetisation inverts its precession direction, as discussed in Ch. 3.3.2.3.

Figure A.1 (c) depicts the calculated factor $(m_{1,\text{eq},z} + \epsilon m_{2,\text{eq},z})$, as expected from our simulation. Please note, with reference to (A.3), that this factor is not the expected field dependence of $V_{\text{ISH,AFM}}$. For the field dependence of $V_{\text{ISH,AFM}}$, $(m_{1,\text{eq},z} + \epsilon m_{2,\text{eq},z})$ has to be multiplied with $A_1$. We may, however not determine the absolute value of the magnetisation deflection $A_i$ and can thus not predict the field dependence of $A_1$. However, the factor $(m_{1,\text{eq},z} + \epsilon m_{2,\text{eq},z})$ does determine the sign of $V_{\text{ISH,AFM}}$. We would therefore expect a sign change of the antiferromagnetic spin pumping voltage, occurring at $H_C$ for good alignment $\theta_H \to 0^\circ$ and at higher magnetic field strengths for increasing misalignment. Please also note that, if we assume $A_1$ to be constant, then the spin pumping voltage in the antiparallel configuration $H_0 < H_C$ is larger than the voltage in the spin flop configuration $H_0 > H_C$—a result which is in striking contrast to the result of the qualitative discussion. Furthermore note that the factor $|m_{1,\text{eq},z} + \epsilon m_{2,\text{eq},z}| < 1$, stating that the spin pumping voltage of MnF$_2$ is smaller than the voltage arising from a single ferromagnetic magnetisation with the same deflection.

As our AFMR simulation has proven valid, the obtained $\epsilon$ values are indisputable. The question is if the ratio of the precession areas is unaltered in the driven case, $1$ and should also be multiplied with the resonance frequency $f_{\text{MW}}$. 

---

1 And should also be multiplied with the resonance frequency $f_{\text{MW}}$. 
Figure A.1: The $z$ component of the equilibrium position of $\mathbf{m}_i = \mathbf{M}_i / M$ (a) and the ratio $\epsilon$ of the areas $A_i$ encircled by the precession of $\mathbf{M}_1$ and $\mathbf{M}_2$, as obtained from the simulation. These two values determine the field dependence of the factor $(m_{1,eq,z} + \epsilon m_{2,eq,z})$ (c), which enters the equation of the antiferromagnetic spin pumping voltage (A.3). All results are presented for two different degrees of external field alignment $\theta_H$. 
under microwave irradiation. We would assume so, as the driving microwave force in FMR experiments can be treated as a small perturbation, which would thus not affect the eigenmotion significantly\(^2\).

In conclusion, the importance of the precession area ratio \( \epsilon \) of the antiferromagnetic resonance for the expected spin pumping voltage \( V_{ISH,AFM} \) has been shown. For the lower frequency mode, we expect a sign change of \( V_{ISH,AFM} \) across the spin flop field. While the sign predictions are robust using the simulated \( m_{1eq,x} \), \( m_{2eq,z} \) and \( \epsilon \) values, the magnitude of \( V_{ISH,AFM} \) as a function of external field may not predicted at present, due to the unknown field dependence of \( A_1 \).

The presented discussion is specific to MnF\(_2\). Using an antiferromagnet with extremely small molecular field strength and thus low critical field, it might be possible to go to external field strengths \( H_0 \gg H_C \), where both sublattice magnetisations align mostly along \(+z\). In this case, the spin pumping voltage should be the sum of the two contributions and the factor \(|m_{1eq,x} + \epsilon m_{2eq,z}| > 1\). The simulation could clarify the required antiferromagnetic properties and external field strengths for this situation.

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\(^2\)If we could not transfer the results of the magnetisation eigenmotion to the case of microwave excitation, then also the predicted resonance frequency and line width would be unsuitable. However, these are in excellent agreement with the measurements.
Appendix B

Microwave Rectification With The Spin Hall Magnetoresistance

In the following, an expression for the electric field detected by the two contacts in the configuration of Fig. 4.1, due to microwave rectification with the spin hall magnetoresistance (SMR) is derived. The SMR with ferromagnetic insulators depends on the magnetisations orientation: \( \mathbf{m} = \mathbf{M} / M \). In the following, the rectification due to a single magnetisation is considered. Chapter 4.3.3 discusses the difficulties to adapt the result to antiferromagnets with two magnetisation vectors.

The Pt film on top of the magnetic sample is depicted in Figure B.1. The electric field \( \mathbf{E}_{rf}(t) \) of the CPW excites AC currents \( \mathbf{J}(t) \) within the thin film, which are determined by Ohm’s law:

\[
\mathbf{E}_{rf}(t) = \hat{\rho}(\mathbf{m}(t)) \mathbf{J}(t)
\]  

(B.1)

The contact measures the \( y \) component of \( \mathbf{E}_{rf}(t) \). Neglecting currents in \( x \) direction, the \( y \) component of (B.1) yields:

\[
E_y(t) = J_y(t)\rho_{\text{long}}(\mathbf{m}(t)) + J_z(t)\rho_{\text{trans}}(\mathbf{m}(t))
\]  

(B.2)

Figure B.1: Illustration of the Pt thin film of Fig. 4.1 with the AC current components \( J_y \) and \( J_z \) and the electric field of the CPW \( \mathbf{E}_{rf} \). The alternating currents lead in conjunction with an anisotropic magnetoresistance to a rectification voltage in magnetic resonance.
Inserting (2.28) and (2.29) into (B.2) gives:

\[
E_y(t) = J_y(t) \left[ \rho_0 - \Delta \rho \left( m_z(t) \right)^2 \right] + J_z(t) \left[ \Delta \rho m_y(t) m_z(t) \right]
\]  

(B.3)

\( m_y \) and \( m_z \) are the projections of \( \mathbf{m} \) onto the coordinate axes.

The time-average of (B.3) describes the DC electric field in the metal thin film caused by microwave rectification with the SMR effect.
Appendix C

Spin Hall Magnetoresistance Of Alternating Currents

Unrelated to the experiments with antiferromagnets, another type of experiments has been conducted in the course of this thesis: Measurements of the spin Hall magnetoresistance with alternating currents.

The recently discovered spin Hall magnetoresistance (SMR)\(^{50,53}\) has been discussed frequently throughout the course of this thesis (cf. Ch. 2.4). Occurring in ferromagnet | metallic thin film bilayers, it describes a resistance variation of the thin film, which depends on the magnetisation orientation of the (isolating) ferromagnet below. The origin of the resistance effect is the ability to modify the spin current boundary condition at the FM|N interface with the magnetisation of the ferromagnet. This in turn affects the charge transport via the inverse spin Hall effect. Previous studies focused on the influence of the spin Hall magnetoresistance on DC currents, which raises the question if AC currents are similarly affected. In the following, we present the results of our measurements, which, quite remarkably, not only confirm the SMR existence, but also show an increase of the SMR effect size with higher AC current frequencies.

The sample, depicted in Figure C.1, consists of an yttrium iron garnet (45 nm)\(^{50,53}\)|platinum (7 nm) bilayer, grown on a substrate of yttrium aluminum garnet. It is mounted in a sample box with microwave SMA connectors at the end of a dip stick and placed inside a rotatable electromagnet, which provides an in-plane magnetic field \(H_0\). The external field strength of 200 mT is larger than the internal anisotropy fields and the magnetisation \(M\) thus follows \(H_0\) directly. Therefore, \(\alpha\) is also the angle of the magnetisation vector \(M\) with the current direction \(J_C\). The experiments were conducted at room temperature.

Using a vector network analyzer, we apply an AC current \(J_C\) in parallel to \(j\) and measure the microwave transmission parameter \(S_{21}\) as a function of frequency \(f\) and angle \(\alpha\). \(S_{21}\) is then converted to an equivalent series impedance \(Z\) of the sample.
and its microwave cabling, using the relation \[ Z = Z_0 \frac{2(1 - S_{21})}{S_{21}}. \] \hspace{1cm} (C.1)

Please note that, due to the lack of microwave calibration, we restrict ourselves to magnitude statements and \( S_{21} \) and \( Z \) denote in the following the absolute value of their respective complex quantity.

Figure C.2 presents the thus obtained impedance as a function of applied microwave frequency up to 1 GHz. The impedance describes the effective impedance of the circuitry composed of the YIG|Pt sample, the sample box and the microwave cables. For low frequencies, the current is mostly passing through the platinum layer and yields an impedance value \( Z \) close to to the DC resistance of \( R = 35 \, \Omega \). For higher frequencies, the sample design is not ideal for microwave transmission and the measured impedance increases.

To measure the SMR effect angle dependent impedance measurements are performed. For each magnetic field orientation \( \alpha \), \( S_{21} \) is measured as a function of microwave frequency. As the impedance increases towards higher frequencies, the obtained data for each frequency cut has to be normalized for a comparison. As a first approach, for each frequency cut, the mean of the impedance over \( \alpha \), \( \langle Z \rangle_\alpha \), is subtracted from the measured impedance value \( Z \). The resulting impedance lift, denoted \( \Delta Z \), is depicted in Figure C.4 as function of \( \alpha \) and for frequencies up to 1 GHz. Additionally, the result of a DC SMR measurement performed with the same sample mounting is presented as red line at \( f = 0 \, \text{GHz} \).
Figure C.2: Impedance of the DUT composed of the YIG|Pt sample in the box and the microwave cabling, as a function of microwave frequency.

From SMR theory, the longitudinal resistance is determined by (2.28), which gives for the $\alpha$ dependence (cf. Fig. C.1):

$$\rho_{\text{long}} = \rho_0 - \Delta \rho m_i^2 = \rho_0 - \Delta \rho \sin^2 \alpha$$  \hspace{1cm} (C.2)

$\rho_0$ is a constant resistance offset and $\Delta \rho > 0$ the SMR effect strength.

First note that the DC resistance variation is in good agreement with the $-\sin^2 \alpha$ dependence, with minima at $\alpha = 90^\circ$ and $\alpha = 270^\circ$. This confirms the existence of an SMR effect in our YIG|Pt bilayer. The impedance for frequencies below 200 MHz does well resemble the observed DC SMR. For higher frequencies, however, the difference between the minimal and maximal values of $\Delta Z$ increases significantly. Furthermore, the $\alpha$-dependence is no longer proportional to $-\sin^2 \alpha$, but exhibits triangular peaks.

As the impedance $Z$ increases towards higher frequencies (cf. Fig. C.2), it seems naïvely logical that the same SMR effect would also result in a larger impedance lift $\Delta Z$. In this way, the increase of $\Delta Z$ towards higher frequencies would be explained by an increased impedance $Z$ of the sample. To exclude this effect, we also used a normalisation that cancels the effects of absolute impedance values by dividing through the mean absolute impedance value: $Z/\langle Z \rangle_{\alpha}$. This ratio is depicted in Figure C.4. Still, the impedance ratio $Z/\langle Z \rangle_{\alpha}$ increases towards higher frequencies and resembles the DC SMR for lower microwave frequencies.
Figure C.3: The impedance lift $\Delta Z$ over the magnetic field orientation $\alpha$, for frequencies to 1 GHz. For every frequency cut, the average impedance of this frequency $\langle Z \rangle_\alpha$ is subtracted from the measured impedance value $Z$.

Figure C.3 depicts again the impedance ratio $Z/\langle Z \rangle_\alpha$, now extended to a larger frequency range up to 8 GHz. The color code is equal to that of the 1 GHz measurement of Figure C.4. Again we find an increase of the SMR effect up to $\sim 1$ GHz. From 1 GHz till 6 GHz the SMR effect of the ratio $Z/\langle Z \rangle_\alpha$ stays constant. Most of the frequency cuts in this frequency range resemble the SMR suggested $-\sin^2 \alpha$ dependence. The triangular dependence observed in Figs. C.4 and C.3 is only measured in the vicinity of $\sim 1$ GHz. The significant discontinuities above $\sim 6$ GHz are so far not comprehended. These discontinuities do not change their frequency position when varying the external field strength, which excludes ferromagnetic resonance effects as their origin. It might be conceivable that the discontinuities arise from a microwave resonance within the box which is slightly sensitive to a magnetic field orientation.

The presented impedance analysis is far from complete, since no distinction is made between the impedance contributions of the sample box, the cabling and the...
actually interesting sample impedance. Nevertheless, given that an increase of both the absolute lift $\Delta Z$ and the relative impedance $Z/\langle Z\rangle_\alpha$ is observable towards higher frequencies, it seems difficult for us to argue that the increase of the SMR effect is not genuine to the YIG|Pt bilayer.

So far, we can only speculate about the origin of the SMR enhancement. The SMR effect bases on the spin accumulations at the sample boundaries. Therefore, an investigation of the scattering times of the mechanisms that build up (spin Hall effect) and reduce (spin transfer torque to the ferromagnet) the spin accumulation might be fruitful, but has not been realized yet.

Further experimental studies of the SMR effect in YIG|Pt bilayers with alternating currents are in process.
Figure C.5: The normalized impedance $Z/\langle Z \rangle_\alpha$ as a function of $\alpha$ and microwave frequency. This measurement extends the frequency range of Figure C.4 to 8 GHz. The false color code is the same as in Fig. C.4.
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