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# Optimized fabrication process for nanoscale Josephson junctions used in superconducting quantum circuits

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# Introduction

In the last decade, quantum information processing (QIP) has revealed itself to be a vivid and fruitful field of physics. Based on the analogy between the principal information unit (quantum bit or qubit) and a simple two-level atom, a deeper understanding of the quantum world has been obtained [1, 2]. From this knowledge, applications have been made possible, such as a quantum processor [3]. For this kind of system, multiple qubits with well-controlled properties are necessary. Single qubits have already been studied intensively regarding their readout [4], their coupling to single photons [5] and their tunable properties [6]. Moreover, quantum gates [7], a quantum bus [8] and an error correction scheme [9] are necessary for a quantum computer to use it for quantum simulations or communication.

One approach is the implementation of these systems in superconducting quantum circuits with built-in nonlinearities [10]. These circuits are macroscopic in size and can be described with a single macroscopic wavefunction. The main advantages of superconducting quantum circuits are large coupling strengths and potential for scalability due to the silicon-based lithographic fabrication technique [11]. For the sake of their specific eigenenergies, the circuits need to be operated at millikelvin temperatures in order to inhibit thermal excitations. Still, they can be intentionally excited and read out using microwave pulses [12–14]. Since cryogenic experiments are usually quite time consuming, it is convenient to judge the quantum properties of a qubit by means of room temperature measurements.

The construction of these superconducting circuits is a challenge in nanofabrication. State-of-the-art technology is compulsory for building working superconducting circuits. On the basis of thin-film fabrication, spin coating techniques, electron beam (e-beam) patterning and e-beam evaporation are used to accomplish the task.

At the WMI, a specific circuit is predominantly investigated: the superconducting flux qubit [15, 16], which consists of a superconducting loop interrupted by three Josephson junctions. A Josephson junction consists of two aluminum superconductors with a thin oxide barrier in between. This particular barrier can be established with an in-situ oxidation after the deposition of the first superconducting layer. The second superconducting layer then leads to a sandwich structure of the junction. Regarding the fabrication of such junctions, former studies were already done at the WMI [17–19]. Since it implies a large number of well-controlled Josephson junction, scalability of the number of qubits holds the promise of actually realizing a universal quantum computer [20], but also involves the

need to fulfill stricter fabrication requirements. Hence, with a rising number of qubits, we face the need for the fabrication of many Josephson junctions with well-controlled quantum properties.

It is the goal of this work to provide for fabrication routines which guarantee reproducibility of Josephson junctions. In this context, it is important to emphasize that microand nanofabrication have to be optimized in each laboratory, taking into account the specific configuration of the involved complex machinery there. Recently, at the WMI, the spin coating procedure and the e-beam lithography were optimized regarding their precision and reliability [21]. Following up on this work, we here analyze the e-beam resist development and the oxidation of the Josephson junction barriers. While these two processes were in principle established at the WMI in previous works, they still suffer from a lack of systematical analysis and optimization. In this work, we contemplate on the physics behind these fabrication steps and analyze the influence of single factors experimentally.

In Ch. 1, we consider the theoretical background of superconductivity and the Josephson junction. We start with a short historical outline of the discovery of and explanation for superconductivity. Then, we derive the Josephson equations which describe the quantum electrical properties of Josephson junctions in detail. The RCSJ model is also part of this introduction to Josephson junction physics. Finally, we briefly review the direct current superconducting quantum interference device (DC SQUID).

In Ch. 2, we demonstrate and investigate the fabrication process from the sample preparation with optical lithography until the completion of the sample. We also take into account the use of different materials for auxiliary on-chip feed lines. Moreover, we provide an insight into the e-beam lithography and the e-beam resist development. After having analyzed the dose, temperature and time influences on the development, we introduce an improved development method. Data from the evaporation and oxidation process is collected using a piezoelectric quartz crystal sensor. We present data logs from this process step and interpret them, in order to gain knowledge for further fabrication processes.

In Ch. 3, we concentrate on the analysis of DC SQUIDs. Investigations on SQUIDs with a DC readout have the advantage of determining the critical current which modulates in the presence of a magnetic field. Both criteria, the maximum critical current and the modulation, give account of the quality of the Josephson junction itself.

Finally, we conclude and summarize our studies and give an outlook onto future plans regarding the fabrication of superconducting nanostructures with built-in Josephson junctions.

# Chapter 1

# Theory

In this chapter, we lay the theoretical foundations for our work. At first, we investigate the phenomenon of superconductivity briefly. A short historical background will be given from the discovery of superconductivity until the quantum mechanical explanation of this effect.

As an essential element in our fabricated samples, we have a closer look on so called Josephson junctions. We derive the Josephson equations and point out characteristic energies and current-voltage characteristics.

The next section covers the modeling of such junctions by an equivalent electrical circuit. It obeys a differential equation of second order. Therefore, we present a classical mechanics analog and introduce the Stewart-McCumber parameter in order to describe the damping of the circuit.

Finally, the application of Josephson junctions in SQUIDs will be discussed. In such a superconducting circuit, the junctions serve as nonlinear inductances. Here, it is also possible to compare the system with a classical analog of an interferometer. We particularly focus on DC SQUIDs and analyze the maximum supercurrent depending on the external magnetic field. We conclude with the voltage state of a DC SQUID. In the following, we rely strongly on Ref. [22] and [23].

# 1.1 Superconductivity

Superconductivity describes the phenomenon of zero resistance and, more importantly, the expulsion of magnetic fields out of the material when the temperature falls below a certain critical temperature  $T_c$ . The vanishing resistance was discovered by Heike Kamerlingh Onnes in 1911 on mercury [24]. For bulk aluminum, one finds  $T_c = 2.1$  K. Walther Meißner and Robert Ochsenfeld made the discovery of the latter effect and claimed that a superconductor behaves like a perfect diamagnet [25]. It took only two years from the publication of the Meißner-Ochsenfeld-effect until the brothers Fritz and Heinz London put down a phenomenological theory for superconductivity in 1935 [26], followed by V. L. Ginzburg and L. D. Landau in 1950 [27]. These theories explain the perfect diamagnetism of a superconductor with a macroscopic model.

Decades later, the BCS-theory, named after J. Bardeen, L. N. Cooper and J. R. Schrieffer, was able to explain the phenomenon of superconductivity quantum mechanically [28]. The groundbreaking observation is that superconductivity is a macroscopic quantum effect. That is, a parameter of a macroscopic sample, such as the magnetic flux, has a distinct quantization. Usually, these effects only occur on the microscopic scale, say atoms or molecules. In the case of superconductivity, the electrons close to the Fermi energy level form phase correlated Cooper pairs which behave according to the Bose-Einstein statistics. Therefore, at low temperatures the ground state can be occupied by many Cooper pairs at once. Since the size of Cooper pairs (10 nm to 1  $\mu$ m) is much larger than their distance in between, the phase is locked. The macroscopic wave function of all the Cooper pairs is as follows [22]:

$$\Psi(\mathbf{r},t) = \Psi_0(\mathbf{r},t) \cdot e^{i\theta(\mathbf{r},t)} = \sqrt{n_s(\mathbf{r},t)} \cdot e^{i\theta(\mathbf{r},t)}$$
(1.1)

with  $n_{\rm s}(\mathbf{r},t) = |\Psi|^2$  being the local macroscopic density and  $\theta(\mathbf{r},t)$  the locked macroscopic phase of Cooper pairs.

In order to break a Cooper pair, an energy of  $2\Delta$  is required.  $\Delta$  refers to the superconducting energy gap. Hence, below this energy, no excitations of the condensate are possible.

The supercurrent  $J_s$  of the Cooper pairs follows the following equation:

$$J_{\rm s} = \frac{q^* \hbar n_{\rm s}(\mathbf{r}, t)}{m^*} \left\{ \nabla \theta(\mathbf{r}, t) - \frac{2\pi}{\Phi_0} \mathbf{A}(\mathbf{r}, t) \right\} \equiv \frac{q^* \hbar n_{\rm s}(\mathbf{r}, t)}{m^*} \gamma(\mathbf{r}, t) \,. \tag{1.2}$$

In this equation  $q^* = 2e$  and  $m^* = 2m_e$  are the specific charge and mass of Cooper pairs. The vector potential **A** is defined as  $\mathbf{B} = \nabla \times \mathbf{A}$  with **B** being the magnetic field. This superconducting current density is proportional to the gauge invariant phase gradient  $\gamma(\mathbf{r}, t)$ . That is, a phase difference causes superconducting current.

### **1.2 Josephson Junctions**

A Josephson junction consists of two superconductors coupled weakly together (cf. Fig. 1.1a). They can be separated by a thin normal conducting or an isolating barrier of a few nm. In our case, we deal with superconductor-isolator-superconductor (SIS) junctions. Brian D. Josephson was the first to predict the behavior of such a system. He formulated two equations, called the Josephson equations [29].

### 1.2.1 Josephson equations

The first Josephson equation describes the current-phase relation of the system. In this equation  $\varphi = \theta_2 - \theta_1$  denotes the phase difference between the two superconductors  $S_1$  and  $S_2$ .



Figure 1.1: (a) Schematic drawing of Josephson junction: On the left and the right the junction consists of a superconductor (green), each with a individual phase  $\theta_1$  or  $\theta_2$ , respectively. In the center is a tunneling barrier with thickness d (orange), which consists of an isolator material. (b) In the graph, the distribution of the normalized Cooper pair density  $n_s(x)$ , the phase gradient  $\gamma(x)$  and the integral of the phase gradient  $\int \gamma(x) dx$  over the barrier is depicted. This integral equals the phase difference  $\varphi = \theta_2 - \theta_1$ .

$$I_{\rm s} = I_{\rm c} \cdot \sin(\varphi) \tag{1.3}$$

The second Josephson equation describes a relationship between the phase difference and the voltage drop over the barrier:

$$\frac{\partial \varphi}{\partial t} = \frac{2\pi}{\Phi_0} V \tag{1.4}$$

In the following, a short explanation of these equations will be given. By using the macroscopic wavefunction, the tunneling of the Cooper pairs through the thin barrier of thickness d can be described correctly. It is much more likely than for normal conducting electrons due to the macroscopic wave function. We solve the Schrödinger equation for the macroscopic wave function of each area and apply the wave matching method. In the end, we get the following current density through the barrier:

$$J_{\rm s} = \frac{e\hbar\kappa}{m_{\rm e}} \frac{\sqrt{n_1 n_2}}{\sinh(2\kappa d)} \sin(\varphi) \equiv J_{\rm c} \sin(\varphi) \tag{1.5}$$

where  $J_{\rm c}$  is the critical current density and  $\kappa$  is a characteristic decay constant

$$\kappa = \sqrt{\frac{4m_{\rm e}(V-E)}{\hbar}}.$$
(1.6)

For  $\kappa d \gg 1$ , the supercurrent decays exponentially with the thickness of the barrier due

to  $\sinh(2\kappa d) \approx \frac{1}{2}\exp(2\kappa d)$ . Finally, the critical current density is formulated as

$$J_{\rm c} = \frac{e\hbar\kappa}{m_{\rm e}} 2\sqrt{n_1 n_2} \exp(-2\kappa d).$$
(1.7)

Starting from Eq. (1.5), we yield the first Josephson equation (1.3) by integrating over the area of the junction. It becomes obvious that the first Josephson equation describes the observation of a tunneling current of superconducting Cooper pairs through the barrier.

The second Josephson equation tells us that a voltage drop over the barrier can be measured due to a time-varying phase difference. A time-constant phase difference, as it is for  $|I| < I_c$ , results in zero voltage, since the supercurrent accounts for the entire applied current. The phase difference  $\varphi$  is then adjusted as described in the first Josephson equation. The maximum current  $I_c$  is also called the critical current. It is set by the thickness and the area of the barrier and hence, the coupling strength between the two superconductors. The derivation of the second Josephson equation can be found in Ref. [22].

### 1.2.2 Characteristic energies

There are two types of energies to be considered when investigating a Josephson junction. The first type is the capacitive energy, which depends on voltage differences in the system. The second type is the coupling energy, which comes from currents in the junction.

A planar Josephson junction can approximately be viewed as a capacitor with a homogeneous area of A and a thickness of d. The barrier material is considered with the dielectric constant  $\epsilon$ . As a result we get a capacitive energy  $E_{\rm C}$ :

$$E_{\rm C} = \frac{1}{2}CV^2 = \frac{1}{2}\frac{\epsilon\epsilon_0 A}{d}V^2 \tag{1.8}$$

The second energy of interest is the coupling energy. It can be seen as the binding energy of two particles due to their overlapping wave functions. If we increase the bias current with time, we change the phase according to the first Josephson equation (1.3). This phase change creates a voltage drop over the junction in conformity with the second Josephson equation (1.4). Therefore, we can define the coupling energy  $E_{\rm J}$ :

$$E_{\rm J} = \int_0^{t_0} I_{\rm s} V dt = \frac{\Phi_0 I_{\rm c}}{2\pi} (1 - \cos(\varphi)) \equiv E_{J0} (1 - \cos(\varphi)) \,. \tag{1.9}$$

From the coupling energy we are able to derive a nonlinear inductance. The coupling energy comes from the motion of the Cooper pairs, that is the changes in current with time. If we take the time derivative of the first Josephson equation and substitute  $\partial \varphi / \partial t$  with the second Josephson equation, we get the junction inductance.

$$\frac{dI}{dt} = I_{\rm c}\cos(\varphi)\frac{2\pi}{\Phi_0}V = \frac{1}{L}V \tag{1.10}$$

$$L = \frac{\Phi_0}{2\pi I_c \cos(\varphi)} \equiv L_c \frac{1}{\cos(\varphi)}$$
(1.11)

The total energy of the system is the sum of the capacitive energy and the coupling energy.

### 1.2.3 Current-voltage characteristics

Due to the superconducting character of the electrodes on both sides of the junction we need to consider the gap energy  $\Delta$  which is specific to the superconductor material. Regarding a Josephson junction, it manifests itself in a gap voltage  $V_{\rm g}$  which is defined as

$$V_{\rm g} = \frac{2\Delta}{e} \,. \tag{1.12}$$

If a junction is biased with a higher voltage than the gap voltage, the junction becomes normal conducting with a specific resistance  $R_n$ . That is, Cooper pairs are broken up and account for a normal conducting current. For aluminum, this gap voltage lies at  $V_{\rm g, Al} = 360 \,\mu\text{V}$ . Between  $-V_{\rm g}$  and  $+V_{\rm g}$  one can observe a supercurrent when the circuit is driven by a constant current source (cf. Fig. 1.2).



Figure 1.2: Schematic *I-V* characteristic of a Josephson junction with applied current: For  $|I| > I_c$  we see the resistive branches. At  $I_c$  the junction jumps into the pure superconducting state by overcoming the gap voltage  $V_g$ . This graph is valid only for  $T \ll T_c$ . The hysteresis is explained in Sec. 1.3.

Regarding the current, one can distinguish the superconducting and the normal conducting part. If  $|I| < I_c$ , Cooper pairs can tunnel without any resistance through the barrier. For  $|I| > I_c$  the first Josephson equation is no more valid and the junction behaves like an ohmic element with  $R_n$ . The Ambegaokar-Baratoff relation includes the critical current  $I_c$ , the normal resistance  $R_n$  and the energy gap  $\Delta$  or the gap voltage  $V_g$ , respectively, when using Eq. (1.12) [30].

$$I_{\rm c}R_{\rm n} = \frac{\pi\Delta(T)}{2e} \tanh\left(\frac{\Delta(T)}{2k_BT}\right) = \frac{\pi V_{\rm g}(T)}{4} \tanh\left(\frac{V_{\rm g}(T)e}{4k_BT}\right)$$
(1.13)

It serves for proving the quality of a Josephson junction. The product of  $I_cR_n$  is also called characteristic voltage  $V_c$ . For very low temperatures, we can approximate

$$\tanh\left(\frac{V_{\rm g}(T)e}{4k_BT}\right) \approx 1$$

### 1.3 RCSJ model

In order to describe the I-V characteristic of a Josephson junction in detail, we apply the resistively and capacitively shunted junction (RCSJ) model. This was introduced by Stewart and McCumber in 1968. The key aspect of this model is to represent the Josephson junction by an equivalent electrical circuit containing a nonlinear inductance  $L_J$ , a normal resistance  $R_n$  and a junction capacity C. Noise sources are left out intentionally in this discussion in order to maintain simplicity. These elements are connected in parallel and biased by a voltage V (cf. Fig. 1.3a).



Figure 1.3: (a) Scheme of an equivalent circuit of a Josephson junction according to the RCSJ model.(b) Plot of washboard potential: With the help of the RCSJ model the Josephson junction can be understood as a moving particle in a tilted washboard potential. The higher the bias current I the steeper is the tilt of the washboard.

In accordance with Kirchhoff's law, the total current I splits up into three different currents. First, there is the supercurrent  $I_{\rm s} = I_{\rm c} \sin(\varphi)$  over the nonlinear inductance. Second, some current also flows over the normal resistance  $I_{\rm n} = V/R_{\rm n}$ . And third, the capacitance causes a displacement current  $I_{\rm d} = C \, dV/dt$ .

$$I = I_{\rm s} + I_{\rm n} + I_{\rm d} = I_{\rm c}\sin(\varphi) + \frac{V}{R_{\rm n}} + C\frac{dV}{dt}$$
(1.14)

If we now take the second Josephson equation (1.4) and substitute V, we obtain

$$I = I_{\rm c}\sin(\varphi) + \frac{1}{R_{\rm n}}\frac{\Phi_0}{2\pi}\frac{d\varphi}{dt} + C\frac{\Phi_0}{2\pi}\frac{d^2\varphi}{dt^2}$$
(1.15)

which is a nonlinear differential equation. The nonlinearities come from the nonlinear behavior of the Josephson junctions stated in Eq. (1.11). After rewriting and using the Josephson coupling energy  $E_{J0} = \hbar I_c/2e$ , the situation described by this differential equation can be compared with a particle moving in a potential:

$$\left(\frac{\hbar}{2e}\right)^2 C \frac{d^2\varphi}{dt^2} + \left(\frac{\hbar}{2e}\right)^2 \frac{1}{R_{\rm n}} \frac{d\varphi}{dt} + \frac{d}{d\varphi} \left[E_{J0} \cdot \left(1 - \cos(\varphi) - \frac{I}{I_{\rm c}}\varphi\right)\right] = 0 \tag{1.16}$$

The factor in front of  $d^2\varphi/dt^2$  refers to the mass of the particle and the factor in front of  $d\varphi/dt$  resembles the friction or damping. Considering the potential landscape, we get a tilted washboard potential as depicted in Fig. 1.3b, where the bias current I defines the tilt. For  $I < I_c$  the potential is tilted in a way, that the particle can run all the way down, which means that there is a superconducting current inside the junction.

By introducing the Stewart-McCumber parameter

$$\beta_{\rm C} \equiv \frac{2e}{\hbar} I_{\rm c} R_{\rm n}^2 C \tag{1.17}$$

and the normalized time  $\tau \equiv t/(2eI_cR_n/\hbar)$  the differential equation (1.16) turns into

$$\beta_{\rm C} \frac{d^2 \varphi}{d\tau^2} + \frac{d\varphi}{d\tau} + \sin(\varphi) - \frac{I}{I_{\rm c}} = 0. \qquad (1.18)$$

### 1.3.1 Strong and weak damping

On the basis of the Stewart-McCumber parameter  $\beta_{\rm C}$ , we can demonstrate the underdamped ( $\beta_{\rm C} \gg 1$ ) and overdamped ( $\beta_{\rm C} \ll 1$ ) case of a Josephson junction. In both cases a supercurrent can be observed. This can be explained in the picture of the particle in the tilted washboard potential. If we ramp a current and tilt the washboard thereby, the particle will start to move with a characteristic frequency, the plasma frequency  $\omega_{\rm p} = \sqrt{2\pi I_{\rm c}/(\Phi_0 C)}$ . A supercurrent can be observed according to the second Josephson equation (1.4), as there is no voltage due to no phase change.

In the case of underdamping, the particle has a large mass and is subjected only to small damping. Once in motion, the particle will not stop immediately due to its large kinetic energy, but only when the potential is brought back to the horizontal state. Hence, hysteresis effects can be observed (cf. Fig. 1.4a).

In contrast, an overdamped particle's mass is small and damping is large. This implies

that the particle can be stopped very quickly when tilting the potential back or forth. For a Josephson junction, this means that no hysteresis is measurable when sweeping the current (cf. Fig. 1.4b).

From this hysteresis, we are able to calculate the Stewart-McCumber parameter and determine how strong the damping of our circuit is.

$$\beta_{\rm C} = \frac{16}{\pi^2} \left(\frac{I_{\rm c}}{I_r}\right)^2 \tag{1.19}$$

with  $I_c$  being the critical current of the junction and  $I_r$  the current, which needs to be applied to restore the junction from the resistive to the superconducting state. This is smaller than the critical current due to the hysteresis.

Real Josephson junctions are found in between those two opposite cases, but this intermediate case is much more complex to be covered and can be looked up in [22].



**Figure 1.4:** *I-V* characteristic of a Josephson junction with regard of damping and hysteresis: Hysteresis occurs only for an underdamped Josephson junction. No hysteresis is observable in the overdamped case.

### 1.3.2 Exceeding the critical current

When we apply a higher current than the critical current of the Josephson junction, we can measure a voltage drop at the junction. This is due to a normal current, which appears beside the supercurrent. The voltage is subjected to the second Josephson equation (1.4) and therefore oscillating. In the case of strong damping ( $\beta_{\rm C} \ll 1$ ), we get a time-averaged voltage of

$$\langle V(t) \rangle = I_{\rm c} R_{\rm n} \sqrt{\left(\frac{I}{I_{\rm c}}\right)^2 - 1} \,.$$
 (1.20)

This equation describes a hyperbola which is plotted in Fig. 1.4b.

### 1.4 DC-SQUIDs

When connecting two Josephson junctions in parallel in a superconducting loop, we obtain a superconducting quantum interference device (SQUID). The magnetic flux penetrating the ring can be measured up to one flux quantum due to interference effects. For our purpose we choose the DC SQUID and therefore concentrate on the theoretical background of these devices. In the DC SQUID a direct current is fed into the circuit.

The basic idea behind a DC SQUID is the interference of the wavefunctions' phase. For reasons of simplicity, we treat both junctions as identical and small compared to the size of the loop. Hence, the critical current  $I_c$  is the same for both junctions and we do not need to consider effects of magnetic flux threading the junctions.



Figure 1.5: Schematic drawing of a DC SQUID: The applied current splits into both branches of the DC SQUID. Due to the magnetic field, a phase difference  $\varphi_{\rm T}$  or  $\varphi_{\rm B}$  is obtained at the top or the bottom junction, respectively. The dashed line represents the integration path for calculating the total phase difference.

According to Kirchhoff's law, the total supercurrent  $I_s$  is given by the sum of the currents in both branches of the circuit.

$$I_{\rm s} = I_{\rm c} \sin(\varphi_{\rm T}) + I_{\rm c} \sin(\varphi_{\rm B}) = 2I_{\rm c} \cos\left(\frac{\varphi_{\rm T} - \varphi_{\rm B}}{2}\right) \sin\left(\frac{\varphi_{\rm T} + \varphi_{\rm B}}{2}\right)$$
(1.21)

For calculating the phase difference terms, we use (1.2) and  $\varphi = \int \gamma(\mathbf{r}, t) \cdot d\mathbf{l}$  and integrate the phase gradient over the DC SQUID loop. Due to the choice of an integration path deep inside the superconductor, where  $J_s = 0$ , we obtain a phase difference which is only dependent on the total flux  $\Phi$  penetrating the DC SQUID loop.

$$\varphi_{\rm T} - \varphi_{\rm B} = \frac{2\pi\Phi}{\Phi_0} \tag{1.22}$$

It becomes evident that the phase differences for each junction in sum are subjected

to the fluxoid quantization of a superconducting ring. Finally, we can insert this result into (1.21) and obtain

$$I_{\rm s} = 2I_{\rm c} \cos\left(\pi \frac{\Phi}{\Phi_0}\right) \sin\left(\varphi_{\rm B} + \pi \frac{\Phi}{\Phi_0}\right) \,. \tag{1.23}$$

From this, we can infer that the maximum supercurrent  $I_{\rm s}^{\rm max}$  is dependent on the flux threading the loop (cf. Fig. 1.6a).

$$I_{\rm s}^{\rm max}(\Phi) = 2I_{\rm c} \left| \cos\left(\pi \frac{\Phi}{\Phi_0}\right) \right| \tag{1.24}$$



Figure 1.6: (a) Plot of the interference pattern of a DC SQUID: The circuit's maximum supercurrent  $I_s^{\max}$  in dependency of the flux  $\Phi$  threading the DC SQUID loop. Different cases with regard of the screening parameter  $\beta_{\rm L}$  are shown. (b) Schematic interference pattern of double slit experiment.

This pattern is similar to the interference pattern of a double slit experiment where two beams of coherent light interfere (cf. Fig. 1.6b). A length difference in the beam paths creates a difference in the phase and causes the waves to overlap constructively or destructively, respectively. In our case, a single Josephson junction can be referred to as a single slit with a Fraunhofer diffraction pattern [22]. When combining two junctions, one receives the same result as in optics due to a distinct phase shift. However, this phase shift is tuned by applying an external magnetic field, whereas in optics it is set by a variation in the beam paths.

### 1.4.1 Screening currents due to self inductance

In this discussion so far, we neglected the self inductance of the DC SQUID loop, which also accounts for a certain flux. Therefore, the ideal maximum supercurrent  $I_{\rm s}^{\rm max}$  cannot be measured to its full extent even at zero external flux. The total flux  $\Phi$  is to be taken as the sum of the external flux  $\Phi_{\rm ext}$  and the flux created by the loop with inductance L:  $\Phi = \Phi_{\rm ext} + LI_{\rm loop}$ . A parameter to describe the self inductance is the so called screening parameter  $\beta_{\rm L}$ .

$$\beta_{\rm L} \equiv \frac{2LI_{\rm c}}{\Phi_0} \tag{1.25}$$

In the case of negligible screening, that is  $\beta_{\rm L} \ll 1$ , we can neglect the flux created by the loop itself and set  $\Phi \approx \Phi_{\rm ext}$ . As a result, this directly enters Eq. (1.24).

For  $\beta_{\rm L} \gg 1$  however, the flux from the screening current affects the DC SQUID severely. It can be shown [23] that the maximum supercurrent decreases with increasing  $\beta_{\rm L}$  almost proportionally to  $1/\beta_{\rm L}$ .

$$\frac{\Delta I_{\rm s}^{\rm max}(\Phi)}{2I_{\rm c}} \approx \frac{1}{\beta_{\rm L}} \tag{1.26}$$

where  $\Delta I_{\rm s}^{\rm max}(\Phi)$  is the difference between the maximal and the minimal measurable current with regard to  $\Phi$ . Nevertheless, a full modulation of  $I_{\rm s}^{\rm max}$  in dependency of  $\Phi$  can be reached for  $\beta_{\rm L} \leq \pi/2$ . Furthermore, at integer multiples of  $\Phi_0$  the screening does not affect the DC SQUID's characteristic in terms of the positions of the maxima and minima.

### 1.4.2 Voltage state of a DC SQUID

Until now, the DC SQUID has only been treated in the zero voltage state, that is when  $I < I_{\rm s}^{\rm max}$ . If the DC SQUID is operated with a current larger than  $I_{\rm s}^{\rm max}$ , the supercurrent is not able to provide for the full current, but besides a normal current and a displacement current can be observed. Then, a voltage starts to develop at the Josephson junctions. For further discussion, we take the displacement current as negligible due to a very small capacitance, which is valid for  $\beta_{\rm C} \ll 1$  (overdamped junction). Moreover, the screening current should be insignificantly low ( $\beta_{\rm L} \ll 1$ ).

Similar to the case with only one Josephson junction, the RCSJ model gives us the following formula for the average voltage according to Eq. (1.20):

$$\langle V(t)\rangle = I_{\rm c}R_{\rm n}\sqrt{\left(\frac{I}{2I_{\rm c}}\right)^2 - \left(\frac{I_{\rm s}^{\rm max}(\Phi)}{2I_{\rm c}}\right)^2} = I_{\rm c}R_{\rm n}\sqrt{\left(\frac{I}{2I_{\rm c}}\right)^2 - \left[\cos\left(\pi\frac{\Phi}{\Phi_0}\right)\right]^2} \qquad (1.27)$$

In Fig. 1.7, we find this relation plotted. From this we learn that if there is a voltage applied, the modulation of  $I(\Phi)$  does not reach zero anymore but appears rounded at its minima. Still, the maxima and minima remain at the same positions. They can be found at  $n\Phi_0$  and  $(n + \frac{1}{2})\Phi_0$ .

In the voltage state  $(I > I_c)$ , the normal resistance  $R_n$  is a characteristic parameter. Effectively, it is proportional to the number of Cooper pairs broken up, which contribute to a normal current. Cooper pairs separate into single electrons when the voltage is higher than the superconducting gap voltage  $V_g$ . Hence, the normal resistance, the critical



Figure 1.7: Plot of voltage state of DC SQUID in dependency of the magnetic flux and the applied current: For zero voltage the DC SQUID's modulation of I in dependency of the external flux can go down to zero. At non-zero voltages the modulation becomes rounded and is shifted to higher current values. However, the maxima and minima remain at  $n \cdot \Phi_0$  or  $(n + 1/2) \cdot \Phi_0$ , respectively. This plot is based on Eq. (1.27)

current and the gap voltage have to fulfill a certain relation. It is the Ambegaokar-Baratoff relation (1.13) which serves as a benchmark for a working DC SQUID.

# Chapter 2

# **Fabrication process**

The fabrication of superconducting nanostructures relies strongly on thin-film technology and nanopatterning. This section deals with the fabrication of DC SQUIDs, which are theoretically described in Ch. 1. We aim at achieving a high yield and reproducible parameters with the equipment available at the WMI. The standard WMI process, which at the beginning of this work suffered from low yield and limited predictability of the junction parameters, is composed of two important stages. First, auxiliary structures larger than  $2 \,\mu\text{m}$  are deposited onto a thermally oxidized silicon wafer. Second, e-beam lithography is used to write the actual DC SQUID circuit with elements in the sub-micron regime.

In the following passage, we cover the preparation of the sample with feed lines and contact pads. Basically, we distinguish between circular wafers with a diameter of 1 in and rectangular wafers in the size of  $10 \times 6 \text{ mm}^2$ . As the material for the structures which surround the DC SQUID is of importance, a closer look on connection difficulties and their solution is given.

Afterwards, we come to show how the DC SQUID circuit is patterned onto the wafer by e-beam lithography. In the course of this thesis, we prioritize the correct development of e-beam exposed patterns on the e-beam resist. Moreover, we present a new development method which features adaptability to varying ambient conditions.

As a third major point, shadow mask evaporation of aluminum onto the wafer is considered with a particular focus on the oxidation step. For shadow evaporation, a double layer resist is necessary which possesses bridge structures with a sufficient undercut underneath. The material is then evaporated from two different angles onto the substrate and therefore causes overlaps. Between the two evaporation steps, the deposited material gets oxidized in-situ. This results in an oxide barrier in between two superconducting layers. In the course of this work, our evaporation system will be described in detail and the surface roughness of the evaporated layer is discussed in dependency of the evaporation rate. A main aspect is the discovery that the usual layer thickness measurement with a piezoelectric quartz crystal during evaporation can also be used to determine oxidation parameters.

### 2.1 Sample preparation

This section covers the preparation of the sample until it can be used for e-beam lithography. Most of the steps are done in the clean room environment of the WMI. An overview of the preparation process is given in Fig. 2.1.

A polished 1 in or  $10 \times 6 \text{ mm}^2$  silicon wafer with 50 nm thermally oxidized SiO<sub>2</sub> on top serves as substrate in the optical lithography process (cf. Fig. 2.1). In both cases, it has to be cleaned thoroughly with acetone and isopropanol (IPA) in an ultrasonic bath, otherwise dirt particles will cause considerable problems during spin coating.

The  $10 \times 6 \text{ mm}^2$  pieces are used for qubit experiments with resonator or transmission line coupling, respectively. These circuits are made of niobium and are fabricated with a positive process. To this end, a niobium film with a thickness of 80 nm to 100 nm is sputtered onto the substrate, followed by a reactive ion etching (RIE) step.

In contrast, the DC SQUID fabrication is done on 1 in wafers because they offer much more space for multiple circuits of the same design. This is necessary for the investigations on reproducibility and yield discussed in Sec. 3.1.3. This kind of wafer is treated with a lift-off process, where the etching step is omissible. Nevertheless, some of our samples are etched beforehand to provide for the same surface structure.

The process continues for both types of wafers in a similar way. The etched or nonetched samples are spin coated with optical resist (cf. Fig. 2.1b). Specific spin coating parameters can be found in App. A.2. The resist needs to be baked on a hot-plate to evaporate residual solvents and to harden the resist. After baking, the resist features a final thickness of approximately  $1.4 \,\mu\text{m}$ . For the lift-off process, a short flood exposure with UV-light and another baking step is done in order to crosslink the uppermost layer of the resist. In this way, the uppermost resist becomes less sensitive to exposure.

An optical mask patterned with chromium is applied to the coated wafer in a mask aligner (cf. Fig. 2.1c). We shine UV-light through the mask onto the wafer for a certain amount of time until the resist's clearing dose is obtained. In the case of the 1 in wafers, we use two different kinds of masks (cf. Fig. 2.2), whereas the  $10 \times 6 \text{ mm}^2$  wafers have resonator designs with arbitrary length and contain several auxiliary circuits (cf. Fig. 2.3a).

After exposure, the sample is developed in AZ Developer. The  $10 \times 6 \text{ mm}^2$  wafer is etched in the RIE after development to obtain the required structures. For the lift-off process with the 1 in sample, the development is followed by the sputtering step. Since the bottom layer is more sensitive to UV-light than the cross-linked top layer, a reverse T-profile is created during development. This type of structure is referred to as an undercut and helps to attain smooth edges of the sputtered feed lines without any high spikes, which could cause problems when depositing another metal layer on top.

In our case, different metals can be used, which are discussed in Sec. 2.2. The deposition of the feed line material is done in a tabletop sputtering system with an argon atmosphere. Details on this process step are summarized in App. A.4. Usually, a 30 nm to 40 nm thin



Figure 2.1: Sample preparation process overview: Photographs of (a) UHV magnetron sputtering system for metals, (b) spin coater Delta 20 BM from BLE with programmable coating programs, (c) mask aligner MJB-3 from Süss, (d) tabletop magnetron sputtering system MED 020 Coating System from BAL-TEC, and (e) reactive ion etcher Plasmalab 80 Plus from Oxford Instruments.



Figure 2.2: Optical mask designs used in this work. The colored pads provide for 4-point-measurements, when the DC SQUID is placed between the contacts. (a) 72 DC SQUIDs on one wafer, real 4-point-measurement possible. (b) 48 DC SQUIDs on one wafer, easy to connect with bonds due to advantageous position of bond pads.

layer is sufficient for the feed lines. In the end, a lift-off step removes the spare metal and the wafer is now ready for e-beam lithography as it is shown in Fig. 2.3.

# 2.2 Connecting measurement lines to the sample

Regarding the connections of the DC SQUIDs, one finds two critical transitions: The first is from the sample holder to the wafer, the second one is from the on-chip feed lines to the actual DC SQUID structure. Both transitions demand a closer look at. For the first transition, we use aluminum bonds which are applied with an ultrasonic bonder (cf. Fig. 2.4a). This is a standard task also used industrially in electronics and therefore offers high reliability. From a needle, a thin aluminum thread of 30 µm in diameter is anchored by ultrasonic welding onto a copper pad of the sample holder. The same welding is done



Figure 2.3: Picture of fabricated silicon wafer with feed lines: (a) Niobium transmission lines on  $10 \times 6 \text{ mm}^2$  wafer. (b) Platinum feed lines and contact pads on 1 in wafer.

at the bonding pad on the wafer (cf. Fig. 2.4b). For a reliable connection, we recommend at least two bonds per connection.

#### **On-chip feed line materials**

Typically, the connection between bond and copper pad is stable as long as the copper is cleaned from copper oxide beforehand with the help of formic acid. Problems occur at the bond-to-wafer transition depending on the used material. For this reason, we investigate three different materials: Gold, platinum and niobium. The first two can be sputtered onto the wafer in a small tabletop sputtering system, whereas the latter one needs to be processed in an UHV sputter cluster.



1 cm

Figure 2.4: (a) Photograph of ultrasonic bonder with needle and thin Al wire. (b) Photograph of a sample connected to sample holder with aluminum bonds.

In our studies, we investigate the three metals in detail for their use as feed lines on silicon wafers and are able to set up the following requirements. A good feed line material has to meet the requirement of being very adhesive on the silicon surface. Otherwise, a working connection can turn out to be impossible due to cracks or parts peeled away somewhere in the feed lines. Moreover, it is required that the evaporated aluminum lines and bonds stick to the feed line metal. Since some metals grow an oxide on their surface when exposed to ambient conditions, the oxide might cause severe problems due to its nonconductivity. Thirdly, the material should be processed very easily and not prohibit the aluminum lift-off step after deposition. In the following Tab. 2.1, the qualitative result of our investigation is summarized:

| Material | Surface    | Adhesion/Durability | Connection                       |
|----------|------------|---------------------|----------------------------------|
| Gold     | No oxide   | Very low            | Problematic (purple plague)      |
| Platinum | No oxide   | Middle              | Fine                             |
| Niobium  | NbOx layer | High                | Problematic (NbOx-Al connection) |

Table 2.1: Comparison of different materials for on-chip feed lines.

### Gold

When using gold feed lines, it becomes evident that they get stripped off by the use of the ultrasonic bath during lift-off, which makes it virtually unusable for our purposes. Moreover, gold feed lines get scratched off very quickly. Reasons for this can be found in the mechanical softness of gold and the poor adhesion to the silicon dioxide surface. Despite the use of chromium as an adhesive layer underneath the gold surface, extremely high caution and diligence need to be taken during fabrication. The adhesion layer can be made thicker – probably several tens of nanometers – in order to increase the gold durability, but thicker feed lines also tend to produce torn edges between feed line and DC SQUID.

Moreover, on-chip bond pads made of gold in combination with aluminum bonds show a phenomenon called purple plague after ultrasonic welding [31]. The welding creates an AuAl<sub>2</sub> alloy of gold and aluminum with purple color and very poor conductivity. This intermetallic formation is also responsible for mechanical failures regarding bond wire and pad. That is, the aluminum bonds tend to fall off from the gold pads, because the bonds are only connected to the brittle alloy instead of the gold surface. At the transition between the gold feed line and the aluminum circuit, this kind of problem does not occur, since no ultrasonic welding is applied there.

### Niobium

Niobium proves itself to be very adhesive on the wafer and offers a good durability. However, our aluminum Josephson junctions sometimes show current-voltage characteristics which are reminiscent of single-electron tunneling.

In Fig. 2.5, such an I-V characteristic is shown. (For details on how to perform I-V curve measurements at 500 mK, the reader is referred to Sec. 3.2.1). It does not resemble



Figure 2.5: I-V characteristic (left) and optical micrograph of the sample (right) of an aluminum DC SQUID with niobium contact pads measured at 500 mK. The niobium-to-aluminum transition area is 270 µm<sup>2</sup> for each line. The niobium is sputter deposited and has a thickness of approximately 40 nm. The aluminum is evaporation deposited and has a thickness of 90 nm.

the tunneling characteristic of a superconducting aluminum Josephson junction. In addition to the qualitatively different shape, the characteristic voltage range is two orders of magnitude larger than the gap voltage of superconducting aluminum,  $V_{\rm g, Al} = 360 \,\mu\text{V}$ . We assume that these tunnel contacts are between the aluminum DC SQUID circuit and the niobium feed lines. As is known, niobium forms an oxide when exposed to ambient conditions. Since in the WMI standard process, the surface of the on-chip niobium is not cleaned in UHV before evaporation, the aluminum may be evaporated onto the niobium oxide which then serves as an isolating tunnel barrier [18]. If the oxide layer is thick enough, the tunneling of Cooper pairs is suppressed and only single-electron tunneling remains. Indeed, we never observe *I-V* characteristics such as the one shown in Fig. 2.5 when the whole circuit is made of aluminum without any Nb-Al transition [32].

One can solve this problem by removing the niobium oxide with an ion gun mounted inside the evaporation chamber. This is verified experimentally. To this end, a simple aluminum line between two ion gun cleaned niobium contact pads is established by evaporation. The thickness of the niobium is 80 nm and that of the aluminum 100 nm. In Fig. 2.6, we see the I-V characteristic of this specific sample and observe a supercurrent with a critical current of 3.1 mA, which is a reasonable value for our geometry. For the cleaning of the wafer inside the evaporation chamber, the ion gun is operated with an acceleration voltage of 2.4 kV and pointed directly at the wafer for 60 s from a distance of approximately 0.3 m. If we do not apply the ion gun cleaning procedure, the I-V characteristic similar to the one shown in Fig. 2.5.

### Platinum

In the end, platinum seems to be the perfect feed line material, since there are neither problems due to connecting with aluminum bonds nor is it scratched off too easily. Never-



**Figure 2.6:** *I-V* characteristic of a thin aluminum strip (without shadow-evaporated Josephson junction) using niobium contact pads and an ion-gun cleaning step: The asymmetry of the graph comes from the overload of the voltage amplifier at both ends of the measured range. Hence, the voltage gap of the niobium cannot be determined. However, we are able to observe the voltage gap of aluminum which is at 0.36 mV as expected. Inset: Optical micrograph of the sample.

theless, sometimes severe lift-off problems occur, which are not yet analyzed in detail. The metal is evaporated on top of the e-beam resist and only reaches the wafer underneath where the resist has openings. During lift-off, spare metal is removed by removing the resist layer. Usually, this is done in an ultrasonic bath. With platinum feed lines and bond pads the lift-off in the subsequent aluminum process does not work as reliably as for gold or niobium feed lines. By heating the ultrasonic bath and increasing the vibration power, however, we achieve satisfying results.

For future sample fabrication, we suggest to use platinum as a bottom layer and gold as a top layer. This combines the advantages of platinum, being sufficiently adhesive, and the advantageous lift-off properties of gold. The gold layer may even serve as a sacrificial layer when it comes off during lift-off. Moreover, niobium might be suitable for our process when it is cleaned thoroughly with an ion gun right before evaporation. However, in the course of this work, it is possible to connect a sufficient number of DC SQUIDs well enough with all three materials. The resulting measurement results are discussed in Ch. 3.

### 2.3 Electron beam lithography

Electron beam lithography proves itself to be an indispensable tool for the accurate patterning of arbitrary structures in the nm-size regime. First, we spend some general thoughts on e-beam lithography. Afterwards, we introduce the e-beam system at the WMI (cf. Fig. 2.7a) and describe the usage in our special case of fabricating DC SQUIDs. Then, we explain how the development of the exposed e-beam resist works and which aspects can be improved under the conditions of temperature and developing time.

### 2.3.1 Nanostructuring

For e-beam lithography, instead of photons, electrons are accelerated and account for exposure of the respective resist. This technique surpasses the limits of usual photo lithography by a factor of approximately two hundred with respect to the resolution. In contrast to e-beam lithography, optical lithography is limited by the wavelength of the incident light and diffraction. For our photo lithography system, we find the resolution limit to be approximately  $1 \,\mu\text{m}$  to  $2 \,\mu\text{m}$ .

With high resolution e-beam resists, the smallest structures possible with e-beam lithography are in the order of 10 nm. The de Broglie wavelength of electrons accelerated with 30 kV is approximately 7 pm. Hence, the resolution is not limited by the electron wavelength, but by the electron distribution inside the resist and secondary electrons from the reaction with the resist polymers. From Monte-Carlo simulations, we know that the beam forms a characteristic cone due to scattering effects (cf. Fig. 2.7b) [21].



Figure 2.7: (a) Photograph of the e-beam lithography system at the WMI: Phillips XL 30 SFEG with Raith writing extension attached. (b) Monte-Carlo simulation of e-beam penetrating the double layer resist: 70 nm of PMMA 950K (blue), 650 nm of PMMA-MA 33% (green), 100 nm of SiO<sub>2</sub> (orange) and 1 µm of Si (yellow). The acceleration voltage is set to 30 kV in this case. We use "Casino" for our Monte Carlo simulations [21, 33].

The e-beam lithography process needs to be carried out in vacuum in order to prevent the electrons from scattering with air molecules and hence maintaining a long mean free path. Moreover, serial patterning accounts for a low sample output. On the one hand, this implies that large structures demand a long writing time. On the other hand, arbitrary patterns can be written and no special mask needs to be manufactured beforehand.

#### Electron beam lithographer

The e-beam lithography system at the WMI consists of a commercial Phillips XL 30 SFEG scanning electron microscope (SEM) equipped with a writing extension from Raith (cf. Fig. 2.7a). The main hardware component of the latter is a sample holder with a motorized stage which can be fine positioned using piezo actuators. For controlling the positioning in X-Y-direction in a sub-µm range, a laser interferometer working at 633 nm is used. Further electronics, such as a pattern generator and a beam blanker, allow the beam to be positioned according to a design file.

During operation, the e-beam is generated with a Schottky field emission gun (zirconium filament) at the upper end of the SEM column and runs through an array of electronical lenses to focus the beam. Usually, we accelerate the electrons with an acceleration voltage of 30 kV. At the bottom end of the column, deflection coils are able to move the beam over the sample. Here, the Elphy Plus software from Raith translates the previously designed patterns into beam movements. Furthermore, the software is capable of setting the correct dose by letting the beam remain on one spot for a certain amount of time.



Figure 2.8: Standard layout for DC SQUIDs fabricated in this work. Black: Feed line contacts made with optical lithography. Green and blue: aluminum feed lines. Orange and red: DC SQUID pattern. The color encodes the EBL exposition dose. Blow-up: Pattern for a shadowevaporated Josephson junction.

#### Focusing the e-beam

In order to create well-defined and accurate patterns in the resist, the focus of the e-beam is important. With a focused writing spot, only the required parts of the resist will be exposed and areas nearby remain unexposed. In principle, two problems can occur which cause discrepancies between the designed pattern and the real structure. First, the beam focal point can be out of the focus plane, which results in an isotropic blurring. Second, the beam suffers under astimagtism, which makes the beam elliptical in one direction. Both problems can be solved with the following method.

We use 25 nm small gold nanoparticles to determine the focus of the beam [21]. They are added onto the hardened resist before placing the sample into the lithographer. These particles have the advantage of being tiny and naturally having a spherical shape. By focusing on them, one can correct the astigmatism easily because only with a correct adjustment, the SEM image displays them as round particles. If the objects are clearly visible as circular structures, we can conclude that our focus aberration is much less than 25 nm.



Figure 2.9: SEM micrograph of 25 nm Au particles with correct focus settings.

Furthermore, in a previous work [21], the influences of the deflection magnet coils close to the sample stage are studied. Magnetic stray fields cause the beam to widen before reaching the focal plane and therefore result in a blurred image. This can now be resolved with a demagnetization script, which is implemented into our system [21].

#### Correct alignment of the wafer

It is not only necessary that the written structures are accurate, but it is also compulsory that the DC SQUID circuit is correctly aligned to the surrounding feed lines at the right position as depicted in Fig. 2.8. Tolerances for this alignment can be increased by larger contact pads, but still remain in the order of  $5 \,\mu\text{m}$ . Therefore, we need to obtain a precise alignment of the wafer. The Elphy software needs the information where the auxiliary feed lines lie and where to start with the patterning. For this, an alignment procedure needs to be undertaken.

With special alignment marks in the feed line design on the wafer, a coordinate system can be established. At least three distinct points on the wafer with sufficient distance to each other are necessary to achieve an acceptable alignment. Into this wafer coordinate system, writefields need to be placed properly because the beam deflection area is limited to a certain size. For our purposes, we choose a writefield size of  $120 \times 120 \,\mu\text{m}^2$ . A semi-automated writefield alignment script is used for rectifying the writefield onto the shifted wafer coordinate system as introduced by F. Sterr [21].

#### Electron beam and dose considerations

The electrons in the beam are accelerated with  $30 \,\mathrm{kV}$  and pass an aperture with an diameter of 1 µm. The acceleration voltage defines the kinetic energy of the electrons and the aperture the broadening and the total beam current. Small apertures allow the e-beam to be controlled in a more focused way, since lens errors occur mainly at the outer borders of the lenses. For our e-beam resists, a minimal acceleration voltage of  $12 \,\mathrm{kV}$  is necessary. We use  $30 \,\mathrm{kV}$  in order to obtain a good focus because faster electrons suffer less from scattering inside the resist. Regarding both the minimum aperture size and the maximum acceleration voltage, we are limited by the specifications of the system.

In our case, we pattern our DC SQUID structures into a double layer resist system in order to yield an undercut. Applied by spin coating, the bottom layer (PMMA-MA 33%) is approximately 670 nm thick, whereas the top layer (PMMA 950K) is 70 nm thin and serves as a shadow mask during evaporation (cf. Sec. 2.4.1). The resist consists of polymer chains, which are broken by the e-beam. A different sensitivity of the two kinds of resists to electron-beam dose can be observed. The bottom resist is much more sensitive due to an increased amount of monomers.

Figure 2.10 clearly shows that a certain minimal dose is necessary to generate any undercut at all during development in IPA. In this case it is around  $600 \,\mu\text{C/cm}^2$ . Especially in the beginning of the undercut formation (around  $600 \,\mu\text{C/cm}^2$  to  $750 \,\mu\text{C/cm}^2$ ), some parts develop faster than others.

For different doses, the top layer does not suffer from degradation during development as it is visible in the graph. This allows for a large choice of suitable doses in the range of  $800 \,\mu\text{C/cm}^2$  or higher. For our structures, we use doses of around  $1200 \,\mu\text{C/cm}^2$  to  $1800 \,\mu\text{C/cm}^2$ . The reason for this large variance is that the secondary electron dose has a much stronger impact on larger structures, such as the feed lines. To account for this effect, we apply a proximity correction and vary the dose with respect to the structure size as depicted in Fig. 2.8.

In comparison to the WMI standard process before this work, our dose is higher by a factor of 2 to 3. This is possible because the resist is already broken at a certain irradiation and will not change significantly anymore until a critical dose is reached where the polymers start to crosslink again. Within this dose window, one may choose an arbitrary dose for the e-beam patterning.

Finally, one can obtain the same undercut with a smaller dose but with a longer development time. The only prerequisite is that the dose has to be sufficiently large to



Figure 2.10: Plot of undercut formation for different doses: The developed structure is measured at several positions as indicated in the inset. From the measured length the structure's width is subtracted and then divided in half to get the undercut on one side. A slight asymmetry can be observed between left and right. The top part has a thinner line and therefore a smaller undercut due to less stray dose. Developing parameters are in this case 3 min 45 s in 28 °C IPA. For lower doses, the undercut size varies stronger than for higher doses.

break the polymers. For our resist system, this is around  $700 \,\mu\text{C/cm}^2$  to  $800 \,\mu\text{C/cm}^2$ .

### Finite accuracy due to beam diameter and resist properties

Regarding the accuracy of our fabrication method, we state that the top layer openings in the resist of the fabricated samples are larger than those drawn in the design patterns. The openings are approximately 15 nm larger at each edge than the set size from the design file. This effect appears systematically. It can not be related solely to the beam diameter, as this is around 3 nm to 5 nm wide. Simulations show that this broadening appears mainly due to the scattering properties of the electrons inside the resist [21].

However, as our system is now calibrated very well, we present in Fig. 2.16 the achievable resolution, which is close to the limit of the resist resolution [34].

### Homogeneous development

Although measures are taken to ensure homogeneous development, a slight asymmetry in undercut size between different parts of the wafer remains (cf. Fig. 2.10). Between left and right, the undercut varies approximately  $0.46 \,\mu\text{m}$  at most. This variation refers to  $25.3 \,\%$  with regard to the average undercut, but is still within a tolerable range, since the top layer remains unaffected.

In the past, the WMI standard process required the wafer to be dry-blown with nitrogen manually. Our experience shows that the blow-dry step with nitrogen causes the structures to develop very fast and inhomogeneously on different parts of the wafer. We expect the evaporation of IPA to cause this fast development. For this reason, it is important to stop the IPA development first by removing IPA residuals with distilled water before the blow-dry step with nitrogen.

We deliver a more detailed explanation of the chemistry of resist development in the following section.

### 2.3.2 Development of the resist

After the required parts of the resist on the wafer are exposed to the electron beam, we proceed to the development of the resist. The following section gives an insight into the chemistry of our resist and the reaction to electron bombardment. Afterwards, we investigate the influence of two major factors during development: time and temperature. In the last part, we present a new method which gives us control over these parameters.

### Resist chemistry and solubility

PMMA, the resist used in our process, is the same as acrylic glass regarding its chemical structure. For the use as a resist, it is solved in a solvent, such as ethyl lactate. The abbreviation stands for Poly-Methyl-Meth-Acrylate  $(C_5O_2H_8)_n$  and describes the individual contents. The polymers have a chain length of more than a thousand monomer units. In our process, we use two kinds of PMMA. The difference between the resists PMMA-MA 33% and PMMA 950K is the content of methacrylate monomers. The attributes of the two resists are summarized in Tab. 2.2.

|                           | PMMA-MA 33 %         | PMMA 950K     |
|---------------------------|----------------------|---------------|
| Trade name                | AR-P 617.08          | AR-P 679.02   |
| Solvent                   | 1-methoxy-2-propanol | ethyl lactate |
| Solid content (%)         | 8                    | 2             |
| Molecular weight (kg/mol) | 110-120              | 950           |
| Density $(g/cm^3)$        | 0.976                | 0.967         |

Table 2.2: Properties of e-beam resists

It has been demonstrated that PMMA fragments which are scissoned by an e-beam, react differently to methyl isobutyl ketone (MIBK,  $C_6H_{12}O$ ) and isopropanol (IPA,  $C_3H_8O$ ) [35]. The dose of the e-beam causes a certain distribution of fragment lengths. Higher doses scission long polymers into shorter fragments. However, for very high doses a phenomenon called crosslinking occurs. Scissioned polymer fragments start to band together again due to the high energy input of the e-beam.

Regarding the developing chemicals, the diffusivity of the fragments is 60% to 160% higher in MIBK as compared to IPA. Moreover, longer fragments interact stronger with MIBK. For IPA development, shorter fragments are required which can be achieved with a more sensitive resist. Alternatively, the short fragments can be obtained with an e-beam dose 4.3 times higher than for MIBK development.

All in all, the dissolution kinetics relies on the fragments' molecular weight, the solvent properties and the mobility of the fragments which decreases with increasing molecular weight. It is evident, that shorter fragments lead to a faster development.

An ideal solubility curve is sketched in Fig. 2.11. When a critical dose is reached, the polymers are scissoned into fragments sensitive to the developer and solubility increases suddenly. Contrarily, long polymer chains remain at unexposed parts and are insensitive to development.



Figure 2.11: Resist solubility curve for ideal resist: High contrast between exposed and unexposed areas.

The top layer, which is made of PMMA 950K with long and heavy polymers, is more robust against the developer. The PMMA-MA 33% bottom layer consists of a lot of light-weight fragments. First, we use the strong MIBK developer to dissolve exposed parts of the top layer. Of course, it also starts to develop the bottom layer. In the next step, we use IPA to treat the exposed bottom layer. The top layer with its long fragments remains stable because IPA only reacts with shorter fragments. That is why it is possible to form an undercut with a shadow mask on top, which is necessary for shadow evaporation as already pointed out in the beginning of this chapter.

#### Influence of time and temperature on the development

Two factors influence the development mainly: temperature and time, since it is a chemical process. At higher developer temperature, the chemical reactions are faster. As a rule of thumb, a 10 °C increase doubles or quadruplicates the speed of the process. A more precise value can be obtained by looking at the  $Q_{10}$  temperature coefficient, also called the van't Hoff rule

$$Q_{10} = \frac{R_2}{R_1}^{10/(T_2 - T_1)} \tag{2.1}$$

where T is the temperature and R the rate. It describes the rate of change in a chemical system by increasing the temperature by 10 °C. It is a special case of the Arrhenius law, which relates an exponential dependency of the rate to the temperature.

For investigations on these parameters, we prepare several DC SQUID structure patterns of the same kind written with e-beam lithography at different doses. A development series is then carried out at two distinct temperatures of 18 °C and 28 °C. During development, we take out the sample after a certain period of time and observe it under an optical microscope. In particular, we are interested in the undercut formation for different doses, times and temperatures. Moreover, the dissolution of the top layer resist is of great concern. The undercut is measured in the same way as described in Fig. 2.10.

In Fig. 2.12, the undercut formation and the stability of the top layer mask is plotted for two different temperatures. This series is recorded for a sample with an e-beam dose of  $1140 \,\mu\text{C/cm}^2$ . According to the van't Hoff rule, the undercut develops much faster at higher temperature. It is a nonlinear process at 28 °C, whereas at 18 °C it behaves linearly for this time window. We can obtain a development rate for the undercut by fitting it linearly and taking the derivative. At 18 °C the undercut forms with a rate of about  $R_{18} = 3.6 \,\mathrm{nm \, s^{-1}}$ . For the further development we expect it to saturate at a certain value as it is the case at 28 °C. Here, the rate starts with  $R_{28}^{\mathrm{ini}} = 17.4 \,\mathrm{nm \, s^{-1}}$  in the beginning. It then decreases to  $R_{28}^{\mathrm{t_1}} = 6.4 \,\mathrm{nm \, s^{-1}}$  with  $t_1 = 127.79 \,\mathrm{s}$  being the time constant.

We can argue on the exponential behavior of the undercut development as follows. The dose deposited by the e-beam decays exponentially with respect to the lateral distance. Thus, the monomer concentration in the resist also decreases exponentially. During the undercut development, parts with a high monomer concentration are developed faster. When the development continues, the rate decreases according to the monomer concentration. Summing up, this shows us that the diminished rate during development in the case of 28 °C can be explained by the exponential decay of the irradiated dose. However, the linear behavior at 18 °C cannot be explained in this way.

An additional suspicion is that the decrease of rate is related to the concentration of the developer and the geometric properties of our sample. Since the top layer is more or less insensitive to IPA, the reaction is locally limited to the openings of the top layer, where pure IPA is in direct contact with the bottom layer. At these spots, the exposed bottom resist gets dissolved very fast at high temperatures. By dissolving, the concentration of pure IPA diminishes due to solved fragments. When it comes to developing the undercut, a cavity is formed beneath the top layer resist. Without intermixing, the concentration of solved fragments accumulates steadily in the cavity and fresh IPA can only be supplied very slowly through the small opening. We expect this to be the reason for a reduction of the rate. At room temperature, this effect is insignificant, since the initial rate is already quite slow and fresh IPA can diffuse fast enough into the undercut cavities leading to a constant concentration of IPA. Eventually, this keeps the rate at the same level.

Taking into account that this development series is recorded at a dose of  $1140 \,\mu C/cm^2$ ,


Figure 2.12: Undercut formation plotted against time: At a development temperature of 28 °C, the undercut u forms much faster according to an exponential decay  $u = A_1 \cdot e^{-x/t_1} + u_0$ . Also, the top layer mask starts to dissolve after approximately 3 min. At lower temperatures, the development takes place linearly without dissolving the top layer mask. This series is recorded for a dose of 1140 µC/cm<sup>2</sup>.

we need to state that for the 18 °C case this is already close to the saturation dose (cf. Fig. 2.13). For the 28 °C case it is far from the saturation dose. This aspect of the amount of dose could have an influence on the chemistry of the development. Due to a lack of data, we do not investigate this in detail.

For our case,  $Q_{10}$  is approximately 4.8. It describes that a 10 °C rise in temperature increases the reaction rate by a factor of almost five. This is still in the usual range of a rate increase in chemical reactions. For even lower temperatures, we estimate the reaction rate to be even smaller and the reaction to be less time sensitive in terms of start and end effects. Regarding the top layer mask, it is recommended to perform the development at lower temperatures since the top layer also starts to dissolve at 28 °C.

#### Effect of the applied dose at different development temperatures

Considering the investigation of undercut formation at different temperatures as a function of the applied dose, the results can be seen in Fig. 2.13. It is intuitive that at higher temperatures, less dose is required to develop an undercut. Also, a larger undercut is achievable with the same amount of developing time. At 18 °C we fit the data with a dose response function

$$u = A_1 + \frac{A_2 - A_1}{1 + 10^{(x_0 - x) \cdot h}}.$$
(2.2)

It is a sigmoid curve which is used to describe systems with a slow progression at the beginning, an acceleration in the middle and a saturation in the end. Here,  $A_1$  and  $A_2$  are the two asymptotic values at the beginning and the end of the curve,  $x_0$  is the x-value of the center and h is the slope at the center. The calculated parameters are summarized in Tab. 2.3.

For higher temperatures the undercut formation follows a biphasic dose response function:

$$u = A_1 + (A_2 - A_1) \cdot \left[\frac{p}{1 + 10^{(x_{01} - x) \cdot h_1}} + \frac{1 - p}{1 + 10^{(x_{02} - x) \cdot h_2}}\right]$$
(2.3)

Such a function applies to systems which can be disassembled into two sections with two different asymptotic values and slopes. The transition between the first and second section is around a dose of  $950 \,\mu\text{C/cm}^2$ . The parameter p gives the proportioning of both sections. Other parameters are similar to the simple dose response function. Due to the fact that in our case the second section is only partly existent, the error values of the fit's second part are high (cf. Tab. 2.3).

|                 | Dose response $(18 ^{\circ}\text{C})$       | Biphasic dose response $(28 ^{\circ}\text{C})$ |  |  |  |
|-----------------|---|--|--|--|--|
| $A_1$           | 0 (set)                                     | 0 (set)  |  |  |  |
| $A_2$           | $0.56 \pm 0.09$                             | $4.07 \pm 61.00$                               |  |  |  |
| $x_0 / x_{01}$  | $964.60 \pm 41.02$                          | $772.21 \pm 23.50$                             |  |  |  |
| x <sub>02</sub> | _   | $1286.42 \pm 2663.36$                          |  |  |  |
| $h / h_1$       | $4.74 \cdot 10^{-3} \pm 1.16 \cdot 10^{-3}$ | $7.61 \cdot 10^{-3} \pm 1.71 \cdot 10^{-3}$    |  |  |  |
| $h_2$           | -   | $4.96 \cdot 10^{-3} \pm 18.37 \cdot 10^{-3}$   |  |  |  |
| <i>p</i>        | _   | 0.29   |  |  |  |

Table 2.3: Fitting parameters of dose response function

A shallow inflection point forms at a dose of  $1020 \,\mu\text{C/cm}^2$ . This can be a hint for polymers which crosslink to clusters at exactly this amount of energy. Crosslinked fragments are harder to dissolve and therefore hamper the development of the resist. There is a general trend to much more monomer scissioning at higher doses.

Now, we are able to extend our previous discussion about solubility (cf. Sec. 2.3.2) with this additional knowledge about the temperature dependence. The ideal solubility curve, shown in Fig. 2.11, is only valid for temperatures much lower than room temperature. For higher temperatures, the critical dose gets shifted to lower values and the curve is smeared out (cf. Fig. 2.14). The reason why we are not able to resolve the ideal curve is also due to the finite dose deposition area of the e-beam which creates a distribution of polymer fragments (cf. Sec. 2.3.2). Thus, the solubility which is sensitive to the fragment length, becomes smeared.



Figure 2.13: Undercut formation plotted against dose: For higher temperatures, the required clearing dose is much lower. In this case,  $650 \,\mu\text{C/cm}^2$  are sufficient at 28 °C development temperature, whereas at 18 °C at least 900  $\mu\text{C/cm}^2$  are necessary. The top layer mask stays the same for the whole range of doses tested in this series. This is recorded for a development time of  $130 \,\text{s.}$ 



Figure 2.14: Resist solubility curve for high and low temperature: This graph is valid for a finite development time.

In the end, we choose a temperature of 4 °C, since the contrast between the exposed and unexposed areas comes closer to the ideal case. Another problem, which can be solved with a lower temperature, is the perpetuation of the top layer mask. At 28 °C the top layer mask already started to dissolve, when the undercut formation is not yet completed. As a third advantage of lower temperatures, we may state that the process becomes less time critical, since development time increases. As a consequence, reproducible results can be obtained for timing accuracies of a few seconds. Furthermore, the process saturates with time for useful undercuts.

However, for even lower temperatures  $(T < 0 \,^{\circ}\text{C})$ , we estimate the critical dose to be very high. In our dose series, higher dose values are left out, since the crosslinking effect becomes stronger for higher dose.

On this occasion, we implement a cooling technique with the use of a peltier cooler (cf. Fig. 2.15). It cools down a beaker glass filled with IPA fast and can maintain a stable temperature with a deviation of  $\pm 0.1$  °C.



**Figure 2.15:** Photograph of the peltier cooler UETR-MOST-16A from Uwe Electronic: It is modified with additional Styrofoam for thermal isolation. A temperature sensor measures the temperature of the developer and feeds the measured value back to the controller unit, which then adapts the cooling power to reach the set temperature.

#### Mask accuracy at different temperatures

We compare the accuracy of the aluminum feed lines evaporated onto the wafer when the resist system is developed at different temperatures. On the SEM image, a very distinct position for comparison is the area close to the Josephson junction. According to the design parameters, the line should be exactly 506 nm wide. As it can be seen in Fig. 2.16, the most accurate structures can be obtained at the lowest temperature, however, the changes below 25°C are minor. Nevertheless, this result tells us how the top layer mask becomes developed at different temperatures, since it is the openings in the mask, which define the size of the evaporated structures underneath.

As already pointed out in Sec. 2.3.2, the top layer mask starts to dissolve at around  $28 \,^{\circ}\text{C}$ . Going to even higher temperatures (50  $^{\circ}\text{C}$ ), the openings in the top layer widen by

a factor of 1.4. Our best results are achieved at a temperature of 4 °C. Here, the resulting feed lines are 536 nm wide. The offset of 15 nm on each side can be explained with the finite diameter of the electron beam, which also causes nearby polymers to scission and become sensitive to the developer. Moreover, we estimate that with a development at 4 °C we are very close to the maximally achievable accuracy. This estimated optimal value is limited only by the electron beam diameter and the resist properties.



Figure 2.16: Measured width of an aluminum line plotted versus the development temperature: The design line width is 506 nm. We choose the development time for each structure in a way that the undercut is sufficient. Inset: SEM micrograph of a typical structure.

As a short side note we remark that according to our experience, the resist becomes insensitive to IPA after the development with MIBK and a longer storage time (several weeks). An explanation can be found in the exposure to UV-light from the sun during storage. Additionally, long storage at room temperature leads to the recombination of the monomers.

## 2.3.3 Iterative development method

In the course of this work, a new method for developing the e-beam patterned resist is introduced. Former methods lack reproducibility due to varying environmental conditions, such as humidity, temperature and consistency of the developer. Moreover, there has been no possibility to check the developed structures before evaporating aluminum on top. Now, we have gained control over several important factors, contrived a pre-check routine and maximized the reproducibility rate. Furthermore, this new method can be adapted to various development situations.



Figure 2.17: Sketch of iterative development method: First the exposed sample is put into the commercial developer AR 600-56 for 60 s, which is a mixture of MIBK and IPA in a ratio of 1:3. After the sample is blown dry with a nitrogen gun, it is placed for 30 s into a beaker with cold IPA on a peltier cooler at approximately 4 °C. Now, we use water to stop the development in order to investigate the sample under an optical microscope. These last two steps can be done repeatedly until the sample shows an acceptable undercut.



Figure 2.18: Controlled undercut and pin sharp top-resist: (a) Micrograph obtained with optical microscope: The dark violet lines represent the openings in the top layer, whereas the light blue parts show the undercut in the bottom layer. Particularly, the regions around the junctions (marked in green) are of special interest, since there the undercut is necessary for shadow evaporation. In this case the undercut is sufficient. (b) Micrograph obtained with SEM: The edges of the junction fabricated with this new method look very rectangular due to a sufficient undercut. For better visibility, the edges of the undercut are marked in orange.

The key step in our new development procedure is a check for sufficient undercut between each step, which makes the process more independent from environmental influences. If, for example, the concentration of the developer has changed, it can be compensated with a longer or shorter development time. Moreover, temperature is brought to a lower level and kept constant with the help of a peltier cooler as already described in Sec. 2.3.2. Iteratively, the sample is put into IPA, which serves as a developer for the bottom resist, for around 60 s, followed by a check under the optical microscope. During this examination it is crucial to stop the development reliably. Therefore, an efficient development stopping method allows to control the chemical reaction whenever it is necessary. To this end, the wafer is put into deionized water for several seconds and tossed a little. Afterwards, we blow-dry it with pure nitrogen manually and observe it under the microscope. Depending on the result of the inspection, the development step with IPA is repeated until the undercut is clearly visible (cf. Fig. 2.18a). The total development time remains stable for our structures and is around 2 min to 3 min. However, if the design pattern changes and thus the secondary electron exposure, the necessary development time also changes.

## 2.4 Evaporation and Oxidation

Evaporation is an established technique in order to obtain nm-thin films of a metal. Typically in UHV, the metal is brought to its liquid phase by an e-beam with high current. Thus, parts of the metal evaporate out of the liner and settle uniformly on the sample as a very thin layer.

The following section deals with the properties of the evaporation system used at the WMI. Then, we investigate the surface roughness of the evaporated films. As a third point, the thickness measurement with the use of a piezoelectric quartz crystal is of particular interest. Finally, this chapter is completed with a method to estimate the thickness of the oxide barrier of the Josephson junctions.

## 2.4.1 Evaporation system and shadow evaporation

The evaporation system at the WMI is home-made and suits the needs of shadow evaporation [36]. Before we describe the system in detail, we first need to explain the method of shadow evaporation.

Inside an UHV chamber, a silicon wafer with a double layer resist, which is patterned by e-beam lithography, is mounted (cf. Fig. 2.19). The top layer resist serves as a mask and in the bottom layer one finds an undercut. In the middle is a suspended bridge which creates a shadow on the silicon surface. Now, aluminum is evaporated onto the wafer under an angle of 17° and with a thickness of 40 nm. This first layer is then oxidized at a constant pressure by letting in oxygen into the UHV chamber. After a certain oxidation time, the second aluminum layer is evaporated onto the sample under an angle of  $-17^{\circ}$ in order to create an overlap between the oxidized bottom layer and the new top layer. Finally, one obtains an Al/AlO<sub>x</sub>/Al sandwich with a well-defined area which then serves as a Josephson junction.

For this technique, several measures need to be taken to guarantee process stability.



Figure 2.19: Scheme of shadow evaporation: The process can be divided into three steps. In the first step aluminum is evaporated onto the sample in an angle of  $17^{\circ}$ . It is followed by an in-situ oxidation at constant oxygen pressure. Then another layer of aluminum is evaporated under an angle of  $-17^{\circ}$ .

First, the purity level of aluminum used in our system is at 99.999%. It comes in  $\emptyset 6.35 \times 6.35 \,\mathrm{mm^3}$  slugs and is pre-melted in the liner. During pre-melting, the e-beam is manually swept across the whole liner to create an homogeneous melt. As a result, the actual evaporation for our samples takes place very uniformly with a constant evaporation rate.

A motorized manipulator at the sample mount enables us to tilt the sample to the correct angle for shadow evaporation. Moreover, the sample is placed at a sufficient distance of 0.65 m away from the hot liner so that it does not suffer from the extreme temperature differences present during aluminum evaporation.

During oxidation, the oxygen flow is kept stable with the help of a mass flow controller. The adjustable valve in front of the turbo pump closes to 50% and the turbo pump helps to maintain a pressure equilibrium. Since the pressures are in the range of  $10^{-4}$  mbar, the turbo pump needs to be properly dimensioned regarding its pump power. Pressure sensors for the UHV and the HV are installed inside the chamber to monitor pressures during evaporation and oxidation. In Tab. 2.4, the process parameters are summarized.

All sensors and devices are brought together in a LabView program which allows for comfortable and automatic control of the whole system. The software features several algorithms to maintain fabrication safety and reproducibility. For example, the degradation of the quartz crystal is measured and a maximum pressure for the beginning of the evaporation can be set. Before the shutter opens and aluminum reaches the sample, rate fluctuations coming from the melting step have to be in an acceptable range. Additionally, the filling level of the liners is recorded and the program displays a warning for empty crucibles. Finally, the software gathers real-time values of the important process

| Material                 | 99.999 % Aluminum                                    |  |  |  |
|--------------------------|--|--|--|--|
| Evaporation pre-pressure | $< 10^{-8}  {\rm mbar}$                              |  |  |  |
| Evaporation rate         | $12\mathrm{\AAs^{-1}}$                               |  |  |  |
| Evaporation angle        | $\pm 17^{\circ}$                                     |  |  |  |
| Evaporated thickness     | $40\mathrm{nm}$ and $50\mathrm{nm}$                  |  |  |  |
| Filament current         | 20 A   |  |  |  |
| Oxidation time           | 900 s to 2300 s                                      |  |  |  |
| Oxidation pressure       | $2.0 \cdot 10^{-4}$ mbar to $3.7 \cdot 10^{-4}$ mbar |  |  |  |

Table 2.4: Parameters for evaporation and oxidation

parameters, such as evaporation rate, pressure, oxygen flow and temperature. These data logs can be evaluated after the process. For future modifications of the LabView program, an adaptive compensation of critical process parameter changes due to environmental influences is desirable.



Figure 2.20: (a) Schematic diagram of the evaporation system with all components necessary for shadow evaporation. (b) Photograph of evaporation system. The rack on the right hand side contains all required devices for process control. (c) Photograph of liner mounted in a water cooled copper block. For heating the aluminum an e-beam comes out from the opening at the bottom of the picture (2), describes a circular trajectory and hits the center of the liner (1) [36].

## 2.4.2 Surface roughness of an evaporated layer

The surface roughness is an important parameter for fabricating well-defined Josephson junctions. In Sec. 1.2, we point out that the thickness of the junction barrier defines the

quantum mechanical properties of the Josephson junction, such as the critical current density or the current-voltage characteristics. Hence, a homogeneous surface with an homogeneous barrier is desirable. The surface roughness is mainly determined by the substrate surface but also by the bottom aluminum layer. The latter one can easily be tuned by modifying the evaporation parameters.

A standard parameter for surface characterizations is the root-mean-squared roughness, which is defined as

$$R_{\rm RMS} = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (z_i - \bar{z})^2}$$
(2.4)

with  $z_i - \bar{z}$  being the vertical distance from the mean value. It is possible to view  $R_{\text{RMS}}$  as the standard deviation of the height. Usually, it is a Gaussian shaped graph. We record our surface data by the use of atomic force microscopy (AFM).

#### AFM technique and setup

For our surface roughness investigations, we use the Asylum MFP 3D AFM (cf. Fig. 2.21a) with a tetrahedral silicon tip (approximately 30° opening angle, cf. Fig. 2.21b) on a diving board silicon cantilever. In tapping mode, the AFM scans over the sample surface and records the surface potential with an oscillating tip resonance frequency of 70 kHz [37]. However, the lateral resolution of an AFM micrograph suffers from drifts and tilts. Thus, we concentrate on the surface roughness.



Figure 2.21: AFM setup: (a) Depicted is the AFM head which sits on top of a microscopy stage and a vibration damping platform (not shown). The sample is put on a glass plate under the illuminated area of the head. (b) SEM micrograph of AFM tip. [37]

Usually, during AFM measurements, the sample is tilted, resulting in an height distribution with a tilted linear offset. This leads to a wrong surface roughness calculation because the RMS roughness is the standard deviation of a Gaussian, which is, in this case, fitted to an incorrect distribution. For this reason, it is crucial to flatten the data before calculating the RMS roughness. To this end, we take the flat wafer as the zero baseline and then subtract the tilt offset.

#### Evaporation rate dependent surface roughness

A test series with different evaporation rates is carried out in order to examine the surface roughness. For this reason, we evaporate aluminum onto  $10 \times 6 \text{ mm}^2$  sized silicon wafers. Except cleaning with acetone and isopropanol no other treatment is applied to the wafers. We use three different evaporation rates at 0.6, 0.9 and  $1.2 \text{ nm s}^{-1}$ . Figure 2.22 shows the RMS roughness of these samples. Grains in the size of  $100 \times 100 \times 3 \text{ nm}^3$  come from surface diffusion of atoms, nucleation and the coalescence of metal clusters [38]. A clear trend can be observed: The higher the evaporation rate, the smoother the evaporated material. This is contrary to other studies on this topic [39], where the roughness and the grain size increase with increasing evaporation rate.



Figure 2.22: Surface roughness due to different evaporation rates. Insets: AFM pictures from which the roughness is deduced. Aluminum grains in the size of approximately  $100 \times 100 \times 3 \text{ nm}^3$  are clearly visible.

We claim the individual setup of each evaporation system to be the cause for such an effect. Parameters which need to be considered are the distance between the liner and sample and the cooling of the liner. This can be different from facility to facility, leading to a divergence in the trend of surface roughness depending on the evaporation rate.

For our system, we think that the slightly higher temperature of the substrate during evaporation leads to smoother surfaces at higher evaporation rates. The aluminum inside the crucible needs to be heated more for a higher evaporation rate. Then, the radiated heat from the crucible is also stronger, leading to a increased heat load on the substrate. Taking everything into account, for our system a rate of  $1.2 \,\mathrm{nm \, s^{-1}}$  is preferable in order to obtain a smooth surface.

## 2.4.3 Thickness measurement

The thickness of the aluminum lines of the DC SQUID is very important, because they influence the inductivity and the screening parameter  $\beta_L$  (cf. Sec. 1.4). Hence, it is necessary to measure and control the growth of the aluminum layers. A common method makes use of piezoelectric quartz crystals. First, we explain the measurement principle and the setup for our system. In the second part, the thickness measurement is validated with an AFM cross-check.

#### Piezoelectric quartz crystal and measurement theory

Artificially grown quartz crystals have a broad variety of usage. For example they can be used as a microbalance or for the measurement of thin films [40, 41]. In general, one can categorize the crystals by their vibration mode, their quartz plate orientation (defined by the cutting angle) and their resonance frequency range. Commonly used are AT-cut crystals, which have a good temperature stability and a cutting angle of around 35°. Their vibration mode is a thickness shear and they can be driven at their overtones to reach higher frequencies.

A piezoelectric quartz crystal sensor consists of a sandwich of two metallic electrodes with a piece of quartz in-between (cf. Fig. 2.23). By connecting the electrodes, one is able to drive the quartz at its resonance frequency. We use crystals with a double anchor regarding the bottom electrode. By trapping the oscillation energy in the center of the quartz, it has the advantage of minimizing unwanted oscillation modes and therefore maximizing resonance stability and crystal life [42].

In our case, the quartz crystal is driven with 6 MHz. The device is mounted inside the evaporation chamber in a slightly larger distance from the evaporation source as the sample. The larger distance and the different solid angle is corrected for by a compensation factor. All evaporation rates stated in this work relate to the quartz crystal sensor [36].

Additional mass  $\Delta m$  on the crystal changes the resonance frequency according to the Sauerbrey equation

$$\Delta f = -c \cdot \Delta \varphi = -c \cdot \frac{\Delta m}{A_{\rm q}} \tag{2.5}$$

where  $\Delta \varphi$  is the surface mass and  $A_q$  the active oscillation area of the circular crystal. c



**Figure 2.23:** Exploded view of double anchored quartz "Inficon 008-010-G10 gold coated 6 MHz" with a diameter of 14 mm. The circular holder (white ring) keeps the crystal from undesired oscillations. An adhesion layer (gray) helps to improve the electrode-to-quartz bonding and reduces micro-tears under evaporation stress. Such tears can cause the deposited film to be unattached from the quartz and therefore not being measured. During process, a film is being deposited on the plain side of the quartz [42].

is the crystal sensitivity, which is defined as

$$c = \frac{f_{\rm q}^2}{N_{\rm AT} \cdot \rho_{\rm q}} \approx 81.8 \cdot 10^6 \, {\rm Hz \, cm^2/g}$$

with  $N_{\rm AT} = 166\,100\,{\rm Hz\,cm}$  being the frequency constant of an AT-cut quartz and  $\rho_{\rm q} = 2.649\,{\rm g/cm^3}$  the density.  $f_{\rm q} = 6\,{\rm MHz}$  is the oscillation frequency of the bare quartz without any additional mass deposited onto it.

The higher the sensitivity, the better the resolution. If one wanted to detect a single aluminum atom with a change of 0.1 Hz, the required sensitivity would assume an exorbitant value of  $2.2 \cdot 10^{21}$  Hz cm<sup>2</sup>/g, resulting in a quartz frequency of 31 THz. With our system, we are able to detect surface mass changes in the range of approximately  $1 \cdot 10^{-9}$  g/cm<sup>2</sup>.

Starting from Eq. (2.5) and assuming homogeneous material deposition, the thickness  $d_{\text{evap}}$  depends on the frequency change  $\Delta f$  as

$$d_{\rm evap} = -\frac{1}{c \cdot \rho_{\rm f}} \cdot \Delta f \,. \tag{2.6}$$

Material parameters enter in  $\rho_{\rm f}$ , the density of the evaporated film. For aluminum,  $\rho_{\rm f} = 2.77 \,{\rm g/cm^3}$ . All in all, an 1 Å layer of aluminum causes the quartz to shift its frequency by well measurable  $-2.27 \,{\rm Hz}$ .

#### Temperature dependence

The system at the WMI is already calibrated very well [36]. However, we experience the problem of temperature instabilities which cause the crystal to oscillate differently. In general, the temperature dependence of an AT-cut quartz can be seen in Fig. 10a in Ref. [40].

An optimum temperature stability can be achieved by taking crystals with a suitable

cutting angle. For room temperature usage, this implies an angle of 35°10′.

For the reason that quartz crystals are quite sensitive to temperature changes, we mounted a new temperature sensor on the outside of the evaporation chamber hull to acquire temperature data. It becomes apparent that our system suffers from temperature fluctuations which destabilize the crystal frequency in the range of approximately 10 Hz per  $1 \,^{\circ}C$  (cf. Fig. 2.24). Therefore, it is absolutely necessary to maintain a stable temperature by water cooling the crystal, in order to resolve frequency changes during oxidation. Also, a phenomenon called thermal shock can appear when the shutter of the evaporation source is opened quickly and the quartz is exposed to the molten aluminum which has a temperature of at least 660 °C in the liner. Hence, before oxidation, when the shutter is closed again, we wait for several minutes until the crystal temperature has stabilized. During this time, also the pressure reaches an equilibrium state again after evaporation.



Figure 2.24: Temperature dependence of quartz crystal (6 MHz AT-cut quartz): The data is taken overnight, when the room temperature slowly changes by 1 °C (water cooling is turned off). Temperature resolution is limited to 0.06 °C by the sensor.

#### Evaluation of measured aluminum thickness

In our case, we first evaporate a layer of 40 nm aluminum and after oxidation another layer of 50 nm. The frequency change can clearly be seen in Fig. 2.25, since it is in the range of 1 kHz to 2 kHz.

Independent thickness measurements using an AFM (cf. Fig. 2.26) confirm the results obtained with the quartz crystal. This proves a correct thickness measurement during evaporation.



**Figure 2.25:** Changes of the quartz crystal's resonance frequency during one evaporation run: The first evaporation of 40 nm aluminum causes the frequency to decrease by 1271 Hz and the second evaporation of 50 nm by 1524 Hz.

## 2.4.4 Quartz data during oxidation

Until now, the quartz sensor has only been used for monitoring the thickness during the first and second evaporation step. Since the quartz crystal data is also recorded during oxidation, we plot the data and analyze it in order to find a method to determine oxidation parameters, such as time and pressure.

#### Model of oxygen uptake

Oxidation occurs when oxygen molecules are brought in contact with the aluminum surface caused by weak Van der Waals forces. This physisorption step is followed by chemisorption, where the molecules dissociate into two oxygen ions with electrons from the metal. Now, one oxygen and one aluminum ion change places and form a polycrystalline oxide  $Al_2O_3$ , which then separates the remaining aluminum from the oxygen. After a monolayer has formed, further oxidation is only possible with the tunneling of aluminum



**Figure 2.26:** Plotted data of the height profile of a Josephson junction in the overlap area measured using an AFM: Thicknesses of evaporated aluminum layers (40 nm and 50 nm). The yellow dotted line in the inset denotes the position of the slice.

ions through the oxide. The probability for aluminum ions to tunnel is much higher than for oxygen ions, since the former ones are smaller. In order to keep up charge neutrality, free electrons in the metal concentrate at the metal-oxide interface and leave positively charged aluminum ions behind. Thus, an electric field is created, which is a driving force for tunneling together with the concentration gradient [43].

Concerning the quartz crystal, a fresh aluminum layer is evaporated onto it in the same way as it is onto the sample. This aluminum film is now exposed to oxygen and experiences an uptake in mass. Due to the light weight oxygen atoms and the resulting vanishingly small changes in the resonance frequency, it is a difficult task to monitor the oxide thickness during oxidation. We measure frequency shifts in the range of -10 Hz to -20 Hz. These could only be made visible, since other sources for frequency fluctuations are eliminated, such as strong temperature variations.

The response of the quartz crystal to the oxidation process can be seen in Fig. 2.27. First, we can see a very fast gain in mass, which refers to oxygen molecules sticking to the metal surface and forming a monolayer by chemisorption. Then, when every site is occupied by oxide, the tunneling process starts. The further mass uptake of oxygen is much slower, since aluminum ions have to tunnel to the surface first. A similar result is also shown in Ref. [44].



Figure 2.27: Frequency of the quartz crystal (black) and oxygen pressure (red) as a function of time during the oxidation process.

After the oxygen flow stops and the valve to the turbo pump is fully opened again, we observe a small rise in the quartz frequency. Actually, a rise in frequency means that the quartz has lost some of its mass according to the Sauerbrey equation (2.5). This loss can be explained by taking into account, that some of the oxygen molecules are only surface bound to the quartz crystal. When stopping the oxygen flow and therefore reducing the pressure significantly, these molecules are withdrawn from the surface and lead to a loss in mass.

At this point, we investigate the speed of the monolayer formation first and then focus on the second part of the oxidation, the tunneling process.

#### Monolayer formation speed

The beginning of the oxidation is governed by a very fast monolayer formation. In this case, fast means in the range of about 60 s as compared to a total oxidation time of 1500 s. In order to describe this event, we take the time derivative of the quartz data graphs (cf. Fig. 2.28a). The derivative's interpretation is the rate of oxygen mass uptake during oxidation. Hence, it can be viewed as an oxidation speed. From the derivatives of the quartz data we can conclude that the initial oxidation speed is similar in each run (cf. Fig. 2.28b). In our opinion, this is an explicit hint for the monolayer formation, since the formation is expected to be uniform in time for each run.



Figure 2.28: (a) As Fig. 2.27. The orange line indicates the derivative evaluated for (b), which is proportional to the oxidation speed. (b) Maximum derivatives near the position indicated in (a). In the end, we obtain an average value of 1.47 ± 0.11 Hz s<sup>-1</sup>. The average oxidation pressure varies between 2.87 · 10<sup>-4</sup> mbar and 3.87 · 10<sup>-4</sup> mbar in these runs.

For the evaluation of the quartz crystal data in the monolayer formation regime, we need to take several aspects into account. During oxidation, the quartz oscillates in oxygen and not in vacuum anymore. This can result in a different oscillatory behavior and a changed heat exchange between the quartz and the environment. Especially for the beginning of the oxidation, these changes might have an impact. Since these effects have not been studied in detail yet, we simply sketch the present situation.

An analysis of the correlation between the oxidation speed and the initial or average oxygen pressure, respectively, does not reveal any characteristic feature. The average oxygen pressure during a single oxidation process lies between  $2.87 \cdot 10^{-4}$  mbar and  $3.87 \cdot 10^{-4}$  mbar as seen exemplary in Fig. 2.27. Nonetheless, the average oxygen pressure during the process plays a rather insignificant role in the beginning of the oxidation, that is the monolayer formation.

Expectedly, the initial pressure influences the initial oxidation more. It ranges from  $3.00 \cdot 10^{-5}$  mbar to  $1.00 \cdot 10^{-4}$  mbar. However, it is an extremely difficult task to determine the initial pressure correctly due to the finite time resolution of the data log. When the oxygen valve is opened, the oxygen pressure rises much faster than the monolayer formation is completed. A pressure equilibrium is achieved after approximately 10 s, whereas the monolayer formation takes approximately 60 s to 100 s. Hence, it does not appear meaningful to calculate correlations between the oxygen pressure rise and the monolayer formation speed.

Summing up, the similar oxidation speeds in the beginning indicate a monolayer formation. This monolayer formation does not have a measurable correlation to the average oxygen pressure in the range used for fabrication. However, for a working Josephson junction one monolayer of oxide is not sufficient. Thus, the subsequent oxidation process after the monolayer formation needs to be analyzed.

#### Tunneling regime characteristics

After an oxygen monolayer has formed rather quickly on the evaporated aluminum, the oxygen atoms are separated from the metallic aluminum. Hence, a slow tunneling process starts to govern the oxidation. Since the resulting oxide thickness strongly depends on this part of the oxidation, we need to investigate this process.



**Figure 2.29:** Plot of quartz frequency data during oxidation from different oxidation runs: All curves are manually aligned to zero, in order to clarify the frequency change  $\Delta f$ . The end of the curves marks the end of the oxidation process. Compared to the monolayer formation regime, the tunneling regime is reflected in a high variety of curves. Oscillations are due to small variations of the temperature of the cooling water.

In the course of this work, we investigate various oxidation times, but keep the pressure constant at approximately  $3.4 \cdot 10^{-4}$  mbar to  $3.6 \cdot 10^{-4}$  mbar. The logged data from the quartz sensor is plotted in Fig. 2.29. Contrarily to the monolayer formation, which causes the frequency to decrease steadily by approximately 7 Hz, the tunneling regime shows a high variety of curve progressions. For this reason, we concentrate on the obvious trends visible in the data.

Firstly, the oxidation is much slower in the tunneling regime than in the monolayer formation regime. This can be seen in the significantly slower mass uptake on the basis of a slower frequency change. Although we cannot identify a general uptake rate, we are still able to make a judgment according to the slopes. For some of the graphs, the change from the monolayer formation to the tunneling is quite abrupt, such as for the orange, the green, the pink and the purple one. Contrarily, the red, dark green and yellow one have a rather smooth transition. This means that the oxidation does not decrease immediately when the oxide monolayer has formed completely. We can guess that this can be traced back to a rate dependent surface roughness. It is obvious that rougher surfaces oxidize faster due to a larger effective surface. Although the rate is always set to the same value of  $1.2 \,\mathrm{nm}\,\mathrm{s}^{-1}$ , fluctuations in the rate might come from the filling level of the liner or from newly refilled aluminum. As a result, the surface of the deposited aluminum may be different from case to case, leading to a peculiar oxidation.

Secondly, the maximum frequency change levels off at around 14 Hz, except for the upper three plots (olive, pink and green). A similar result has also be obtained in [44]. We expect that the three outliers come from different initial conditions, such as pre-pressure inside the vacuum chamber after aluminum evaporation or a higher room temperature. All three of them are carried out at room temperatures around 24 °C to 25 °C, whereas the other ones are carried out at temperatures below 22 °C. The higher room temperature might have an influence on the quartz crystal sensitivity, even if it is water cooled. Though the water temperature could not be recorded, we guess that the temperature in the water cooling system was also higher, since the cooling system is connected to ambient air via a heat exchanger. Therefore, the quartz crystal sensor worked at a different temperature with a different sensitivity. Nevertheless, a clear trend is observable for the majority of the data plots.

Thirdly, most of the graphs contain oscillations. These may come from slight temperature oscillations in the cooling water. The oscillations also appear, if we log the quartz when there is no process running. Moreover, for some process runs, the oscillations do not appear at all. From this, we conclude that they have an external origin. The cooling system also cools other fabrication devices at the WMI, such as the sputtering chamber or the pulsed laser depositor close by. These may cause small variations in temperature. From the existing quartz frequency data we can estimate the amplitude of these temperature oscillations. The frequency variations are between 2 Hz to 4 Hz. This corresponds to a temperature variation of approximately  $0.2 \,^{\circ}\text{C}$  to  $0.4 \,^{\circ}\text{C}$ , which seems to be a realistic range.

If we take these aspects into account, the next task is to stabilize the quartz crystal sensor's cooling water. We recommend to set up an independent cooling cycle and to log the water temperature for keeping it constant. This may lead to more reliable data and make the crystal sensor a better tool for monitoring the oxidation.

#### Comparison of L-product and quartz crystal data

The L-product is the product of the oxygen pressure during oxidation and the oxidation time. Until now, this is used to estimate the Josephson junctions' critical current densities  $J_c$ . A higher L-product should result in a thicker oxide barrier and therefore in a lower current density. This general trend is shown in [18] for Nb/AlO<sub>x</sub>/Nb junctions, but large

deviations occur.

By trial and error, one is able to map the L-product to certain critical current density values. At the WMI, we usually use an L-product of about  $1500 \text{ s} \cdot 3.4 \cdot 10^{-4} \text{ mbar} = 0.51 \text{ s}$  mbar which results in a critical current density of approximately  $1 \text{ kA/cm}^2$ . Additionally, the same L-product can be achieved with various parameters, such as lowering the pressure and simultaneously increasing the oxidation time.

We find out that the frequency changes of the quartz crystal give a better estimation of the expected  $J_c$  than the L-product. In Fig. 2.30 the L-product, the maximum frequency change during oxidation and the final critical current density are plotted for several samples. Low critical current densities result from a thick oxide barrier. Hence, the L-product or the quartz frequency change, respectively, are high. In this range, both give a reasonable trend for the expected critical current density.

When the barrier becomes thinner and the critical current density higher, the L-product does not become lower, whereas the quartz frequency change does indeed. For this reason, we claim the quartz crystal to be a better method for estimating the critical current.



**Figure 2.30:** Critical current density, L-product and quartz frequency for different fabrication runs. The error bars of the  $J_c$  data points come from the fact, that we take measurements from several samples fabricated in a single fabrication run.

An explanation for the advantage of the quartz sensor may be the better temperature stability compared to the L-product. For the L-product, the room temperature needs to be the same in every run, otherwise the pressure inside the evaporation chamber will be different, resulting in a faster or slower oxidation. Contrarily, the quartz crystal's maximum change in frequency already contains the temperature.

Nevertheless, the maximum frequency change is covered by temperature oscillations and measures have to be taken to stabilize the quartz even better. This would increase the read-out precision by around 10%. For the future fabrication process, a quartz based oxidation control is to be preferred.

A stable oxide barrier with a correct thickness is one of the main elements of a Josephson junction. In the next chapter, we present the functionality of the oxide barrier by characterizing the Josephson junctions in a DC SQUID configuration.

# Chapter 3

# Results

After having described the fabrication routine, we now investigate the fabricated Josephson junctions in this chapter. We present the most important results from our investigations. They exemplify the main outcomes of this work.

At first, we will have a look at the quality of the samples. The estimated thickness of the oxide barrier, surface roughness and the influence of reactive ion etching are the topics in this section. It is our aim to point out how these parameters affect the quality of the DC SQUIDs. Additionally, an overview over the steps towards a better reproducibility is given.

This is followed by the cryogenic characterization of the DC SQUIDs where we describe our measurement setup and present the I-V characteristics. Of particular interest is the critical current  $I_c$  and its statistical spread for different samples.

Finally, we introduce a new pre-characterization method for nanometer-sized superconducting circuits. With the AFM as the instrument of choice, we are able to image the Josephson junctions nondestructively. The nondestructiveness is demonstrated by recording the DC SQUIDs' I-V characteristics before and after the AFM investigation.

# 3.1 Quality of samples

Several aspects define a sample of high quality. Our main interest is put on the Josephson junctions which are quite sensitive to the following parameters.

The oxide barrier thickness is of particular concern, since it has an exponentially large impact on the critical currents of our circuits. In this case, the quality of the sample is defined by the reproducibility of the oxide barrier within the limits set by our currently available equipment. Regarding the oxide barrier thickness, a direct measurement is not possible, since it sits underneath a layer of aluminum. Nevertheless, we can obtain data about the oxide barrier indirectly by measuring the critical current.

Second, the roughness of the aluminum layer underneath influences the oxide barrier thickness in a very complex manner. Hence, we demand highly uniform surfaces from sample to sample in order to keep up reproducibility. We estimate that the main influences on the roughness of the deposited layer come from the prior etching of the substrate surface and the evaporation process. As already discussed, the evaporation rate can cause roughness differences (cf. Sec. 2.4.2). Roughness analysis is mainly done with AFM, since SEM can hardly resolve the surface structure. AFM, however, records three-dimensional data. By tapping the surface line by line, even small grains and bumps can be made visible. Summing up, the surface roughness is expected to be the same all over the wafer, but can vary from sample to sample.

When it comes to scaling the numbers of superconducting circuits, the junctions will be distributed over a much larger area on the wafer. Still, the junction parameters should remain identical. Hence, a homogeneously deposited aluminum all over the wafer becomes indispensable. It is due to the resist system that deviations might occur. Optimizations on this topic have been done in the course of [21].

At last, the sample quality is defined by the geometric precision of the Josephson junctions. Especially the overlap area of a Josephson junction is of major importance, since identical junctions are characterized by equally large junction overlaps. A sample quality criterion is the reproducibility of such identical junctions. For the investigation of this, we can use SEM, but the risk of destroying the junctions by charging effects is quite high. Therefore, AFM is the better choice in this case.

## 3.1.1 Oxide barrier thickness estimations using the quartz crystal

We are very interested to know the precise thickness of the oxide barrier, since this is the crucial part of the Josephson junction which defines the quantum mechanical properties. The critical current density  $J_c$  relies exponentially on the thickness d according to Eq. (1.7). Since  $J_c$  appears in several other terms which describe the Josephson junction physics, such as the coupling energy  $E_J$  (cf. Eq. (1.9)) or the Stewart-McCumber parameter  $\beta_C$  (cf. Eq. (1.17)), a controlled thickness is of prime importance.

As we point out in Sec. 2.4.4, the quartz crystal frequency change includes more accurate information about the oxidation process than the L-product. Nevertheless, the oxide thickness can only be roughly estimated using the obtainable data from the quartz crystal sensor.

Due to the setup and working principle of the quartz sensor, it measures the mass uptake. If we take Eq. (2.6) to calculate the thickness, we need the density of the oxide film. In contrast to a regular deposition process, oxygen is deposited onto the crystal and then reacts chemically with aluminum and forms an oxide which is chemically and physically different from pure oxygen. Particularly, it has a different density. For a rough estimation, we assume an effective film density of

$$\rho_{\rm eff} = \rho_{\rm AlO_x} - \rho_{\rm Al} = 1.18 \,\mathrm{g/cm^3}.$$

This is directly related to the effective mass uptake during oxidation.

In Fig. 3.1, the thicknesses are calculated for different samples according to Eq. (2.6).

For some samples, the critical current density values can be obtained as described in Sec. 3.2.



Figure 3.1: Critical current density from SQUID *I-V* curve measurements as a function of the oxide layer thickness determined from quartz crystal measurements: Black squares: data. Red line: fit using Eq. (3.1). The error bars in y-direction are of statistical nature, whereas the error bars in x-direction are of systematical origin as described in the text. At the bottom, further thickness data is plotted in blue from samples which could not be measured.

From the graph, it is obvious that Josephson junctions with a thin barrier have a higher critical current density, whereas thick barriers result in low current densities. In this regard, our thickness estimation seems to be reasonable.

Another indication for the quality of our estimation is the fit which converges on these data points. The fit is applied on the foundation of

$$J_{\rm c} = \frac{e\hbar\kappa}{m_e} \frac{\sqrt{n_1 n_2}}{\sinh(2\kappa d)} = a \cdot \frac{\kappa}{\sinh(2\kappa d)}$$
(3.1)

with a being a constant factor which includes the physical constants and the Cooper pair density. The values of the fitting parameters are summarized in Tab. 3.1.

Although we state here only a rough estimation, the fit still converges. This tells us that up to an arbitrary factor, which can be related to an incorrect quartz crystal calibration, the estimated thickness is reasonable. For even more data points, these errors could be ruled out and the estimation could be tested. However, on the basis of the fit and the

|          | Fit for $J_{\rm c}$ and $d$          |  |
|----------|--------------------------------------|--|
| a        | $19.69 \pm 4.31 \mathrm{A  cm^{-1}}$ |  |
| $\kappa$ | $1.93 \pm 0.11  \mathrm{nm^{-1}}$    |  |

**Table 3.1:** Fitting parameters of  $J_c$  fit function

value for the characteristic decay constant  $\kappa = \sqrt{4m_{\rm e}(V_0 - E_0)/\hbar}$ , we can calculate the potential difference  $V_0 - E_0$  by inserting the mass of the Cooper pairs and  $\hbar$ . We obtain

$$V_0 - E_0 = 71 \,\mathrm{meV}\,.$$

Compared to the Josephson junction coupling energy  $E_{J0} = \Phi_0 I_c/2\pi$  which is in the order of 1 meV for our junctions, this approximation for the potential difference appears to be reasonable.

From the parameter a, the Cooper pair density  $n = \sqrt{n_1 n_2}$  can be calculated.

$$n = a \cdot \frac{m_{\rm e}}{e\hbar} = 1.061 \cdot 10^{16} \, 1/{\rm cm}^3$$

This value is two orders of magnitude smaller than the typical value for a bulk superconductor [45]. But considering the case of a thin superconductor where boundary effects play a significant role at the edges of the insulating barrier of the Josephson junctions, the number of Cooper pairs is diminished. Hence, the order of magnitude of the calculated Cooper pair density seems to be correct.

As both values lie in the correct order of magnitude, the foundations of our model seem justified. We are now able to relate the critical current density to the oxide layer thickness. For a desirable critical current density of  $1 \text{ kA/cm}^2$ , a thickness of  $d \approx 1.12 \text{ nm}$  is necessary, as can be seen in Fig. 3.1.

Since we are dealing with an estimation, we also need to estimate the errors. The error of the maximum quartz frequency is  $\Delta f_{\text{max}} = 2 \text{ Hz}$  due to oscillations, as discussed in Sec. 2.4.4, which obscure the maximum value. Regarding the error of the effective density, we take it as  $\Delta \rho_{\text{eff}} = 0.5 \text{ g/cm}^3$ . We neglect the error of the crystal sensitivity, as it is much smaller than the error of the frequency change. This results in a systematic error of 0.26 nm.

We can now substitute the values into Eq. (2.6) and obtain

$$d_{\text{oxid}} = -\frac{1}{81.8 \cdot 10^6 \,\text{Hz}\,\text{cm}^2/\text{g} \cdot 1.18 \,\text{g/cm}^3} \cdot \Delta f$$
  

$$\approx -0.1036 \,\text{nm}\,\text{Hz}^{-1} \cdot \Delta f \,. \qquad (3.2)$$

#### Thickness of an aluminum oxide monolayer

The thickness of a single aluminum oxide monolayer can be estimated as follows. First, we calculate the number of aluminum atoms per area. As an approximation, we use the vertical distance  $d_{\rm Al}$  of two aluminum atoms in an aluminum lattice as the thickness of one aluminum monolayer [46].

$$\rho_{\rm Al} = \frac{m_{\rm Al}}{V}$$
$$\frac{N_{\rm Al}}{A} = \rho_{\rm Al} \cdot \frac{d_{\rm Al}}{M_{\rm Al}} = 2.77 \,\text{g/cm}^3 \cdot \frac{404 \,\text{pm}}{26.98 \,\text{g mol}^{-1}} \approx 2.44 \cdot 10^{13} \,\text{cm}^{-2}$$
(3.3)

From the molecular formula of aluminum oxide  $Al_2O_3$ , we know that two aluminum atoms are required to form a single oxide molecule. Using Eq. (3.3), but with the properties of aluminum oxide, we can guess the thickness of an  $Al_2O_3$  monolayer.

$$d_{\text{AlO}_x} = \frac{N_{\text{Al}}}{A} \cdot \frac{1}{2} \cdot \frac{m_{\text{AlO}_x}}{\rho_{\text{AlO}_x}} = 1.22 \cdot 10^{13} \,\text{cm}^{-2} \cdot \frac{101.96 \,\text{g} \,\text{mol}^{-1}}{3.94 \,\text{g/cm}^3} \approx 525 \,\text{pm}$$
(3.4)

Hence, the estimated thickness of the oxide barrier, approximately 1.1 nm to 1.8 nm, refers to about 2 to 3 stacked aluminum oxide monolayers. In this calculation, we assume the monolayers to be ideally flat which is not the case in reality due to the surface roughness of the layer underneath. Moreover, the calculation is based on a very rough approximation regardless of the aluminum oxide crystal properties and chemical bonds. However, it is not linked to the quartz crystal sensor but is applicable in general.

## 3.1.2 Surface roughness and impact on the critical current density

As already mentioned in Sec. 2.4.2, the surface roughness has an impact on the critical current density. Thus, we need to study it in detail. We approach this topic by taking into account three different areas of a Josephson junction (cf. Fig. 3.2). From top to bottom, we first have the top aluminum layer which has been evaporated last. Underneath is the primarily evaporated aluminum layer. The bottommost layer is the silicon substrate with silicon oxide on top.

From a former investigation on the surface roughness of Josephson junctions [37], we know the following:

- The substrate layer roughness is similar for all samples.
- The top layer roughness is higher than the bottom layer roughness, which itself is higher than the substrate roughness.
- Etching of the substrate surface increases its roughness.
- Residual resist can alter the measured surface roughness of the substrate.



Figure 3.2: Micrograph obtained with AFM: Roughnesses at different positions of the sample.

Understanding the factors which are responsible for the surface roughness is of great importance. Basically, a rough wafer surface is imprinted into the first evaporated aluminum layer, since aluminum is a wetting material. Figure 3.2 demonstrates the similarity of the aluminum layers to the wafer substrate. The substrate roughness has increased due to residual resist. The bottom aluminum layer has a significantly higher surface roughness than the substrate, since distinct features of the substrate can be found imprinted into the metal layer and are thereby enlarged. Regarding the two aluminum layers, the top layer has a slightly higher surface roughness than the bottom layer. This result coincides with the result from [37].

The evaluation of the dependency of  $J_c$  on the surface roughness does not reveal any profitable information, since the number of cryogenically analyzed samples is too small. Still, a roughness comparison of many fabricated junctions, although their *I-V* characteristic could not be recorded, shows that the roughness increases from bottom to top (cf. Fig. 3.3). The difference between the bottom layer and the top layer of aluminum is far smaller than the difference between the substrate and the bottom layer. We can also see that etching increases the roughness significantly as we will describe in detail in the next section (cf. Sec. 3.1.2). More on surface roughness investigations of Josephson junctions can be found in [37].

As the oxidation is very sensitive to the surface potential of the aluminum, a varying surface roughness can cause the oxidation to take place differently.

Especially so called grains, which we assume are relatively large crystal clusters with a typical size of several tens of nanometers and typical heights of approximately 3 nm, may alter the surface potential locally. Since the oxide barrier is less than 2 nm thin, variations



Figure 3.3: Plot of RMS roughnesses of different samples from various positions on the surface: The black dots refer to the substrate surface, the red ones to the bottom aluminum layer and the blue ones to the top aluminum layer as depicted in Fig. 3.2. Sigma denotes the standard deviation. [37]

in the range of 2 nm to 3 nm already can cause significant changes. Although these grains might not have an impact on the average oxide layer thickness, due to the exponential dependence on this thickness of  $J_c$ , very thin spots in the barrier may drastically shift the quantum properties of the junction. Also, the surface roughness increases the effective area of a junction and hence decreases the effective critical current density. An inhomogeneous oxide barrier has an effect on the capacitive energy  $E_{\rm C}$  and the coupling energy  $E_{\rm J}$  as described in Sec. 1.2.2.

#### Influence of reactive ion etching

In our fabrication routine, reactive ion etching (RIE) is compulsory for some of our samples (cf. Sec. 2.1). This type of treatment provides for very well-defined and vertical edges of metal structures, such as resonators, but it also changes the surface roughness of the silicon wafer. The reason is that after the niobium has been etched off, the wafer's silicon dioxide surface is etched slightly. In Tab. 3.2, the roughness of etched and non-etched wafers is compared. The etching parameters can be found in App. A.1.

| RMS-roughness of $SiO_2$ wafer surface (nm) |   |                               |   |                   |  |  |  |  |
|---|---|-------------------------------|---|-------------------|--|--|--|--|
| Etched (with residual resist): 1.819        | > | Etched <sup>1</sup> : $0.416$ | > | Non-etched: 0.392 |  |  |  |  |

Table 3.2: Comparison of etched and non-etched SiO<sub>2</sub> surfaces

In a RIE system, a plasma is ignited. The plasma consists of gases such as argon, sulfur hexafluoride or oxygen. Ions from the plasma are accelerated towards the sample where

<sup>&</sup>lt;sup>1</sup>This sample has been etched with a pure  $SF_6$  plasma without Ar.

they cause chemical and physical etching. The RIE process is highly anisotropic, that is, the direction of the etching can be adjusted. Hence, vertical edges of metal resonators can be achieved.

It is important for resonators or other circuits that all spare metal is etched off at the edges in order to guarantee high isolation. Therefore, the etching process is configured in such a manner, that it also etches slightly into the silicon oxide surface of the wafer. This, of course, alters the surface structure of the silicon oxide. Indeed, this is of significance, since in the subsequent step, the aluminum superconducting circuit is placed onto this kind of modified surface. As already pointed out in the previous section, the aluminum as a wetting layer adopts the surface features of the layer underneath.

From the surface roughness data, we can see in Fig. 3.4, that the height distribution of an etched surface fits much better to a Gaussian. This indicates a more homogeneous surface. Both samples are prepared similarly except for the etching. The etching process parameters are given in App. A.1. After the etching, we evaporate an aluminum layer with a thickness of 40 nm onto the wafer. No further treatment follows this evaporation. The surface data has been obtained with the help of an AFM.



Figure 3.4: Height distribution plot of etched and non-etched sample with aluminum on top: A 40 nm aluminum layer is on top of the silicon oxide wafer.

We conclude on the basis of this evaluation that the RIE etching leads to a slightly rougher but more homogeneous surface all over the wafer. The surface's homogeneity is advantageous for fabricating Josephson junctions.



Figure 3.5: Plot of critical current density for etched and non-etched samples: In total, six DC SQUIDs from fabrication run number five are measured and show a similar critical current density. Number one's huge spread can be explained by a lower oxidation pressure.

Figure 3.5 shows a trend of a decreasing standard deviation for decreasing critical current densities. An explanation for this can be found in the thickness of the oxide barrier. Smaller critical currents come from thicker barriers which are easier to fabricate and hence lead to a more uniform thickness from junction to junction. For thin barriers and high critical currents the impact of surface features, such as grains, is higher. A spread in the critical currents is to be expected as depicted in the plot.

From the data, we see that etching diminishes the variance even more. If we compare fabrication run four and five, which have a similar critical current density, we conclude that the variance in the critical current density is by a factor of 3.5 smaller for the etched sample. This indicates that etching has the effect of creating a more homogeneous surface which is a first indication towards the uniformity of the critical current density.

Similar critical current densities are a key to reliable fabrication of identical circuits. In the next section, we focus on the aspects and measures for reproducibility.

## 3.1.3 Reproducibility and geometric precision benchmark

One goal of this thesis is to achieve reproducibility in the fabrication process of identical DC SQUIDs or qubits, respectively. Reproducibility is absolutely necessary for scaling

the number of superconducting circuits.

In this section, we examine the reproducibility of geometry parameters of the Josephson junction. Referring to the theory in Ch. 1, the overlap area of the junction has a significant influence on the quantum behavior of a DC SQUID. For this reason a clearly defined area with rectangular borders is desirable.

Following steps are taken to guarantee reproducibility. Most parts of the process are automated or contain a cross-check possibility:

- 1. Fully automated spin coating program with same amount of resist to maintain resist thickness
- 2. Dose calibration during e-beam lithography
- 3. Iterative development method for sufficient undercut
- 4. Fully automated evaporation procedure to guarantee same evaporation and oxidation

As far as the geometrical requirements are considered, it is possible for a first look to use the SEM to investigate the overlap of the Josephson junctions. With the items mentioned above, one can fabricate junctions which look very identical (cf. Fig. 3.6).



Figure 3.6: Josephson junction reproducibility over time.

A very crucial geometric property of a Josephson junction is its overlap area. The width B is given by the width of the openings in the top layer mask. However, the overlap length L is defined by the angle of the shadow evaporation as discussed in Sec. 2.4.1.



Figure 3.7: Scheme of evaporation angle and resulting overlap: L is the overlap length we want to calculate,  $\alpha = 17^{\circ}$  is the evaporation angle, H = 670 nm is the height of the resist,  $h_{Al} = 40$  nm is the height of the first evaporated aluminum layer and W = 292 nm is the width of the suspended resist bridge.

From Fig. 3.7 we can deduce the following formula for the overlap length:

$$L = H \cdot \tan(\alpha) + (H - h_{\rm Al}) \cdot \tan(\alpha) - W$$
  
=  $(2H - h_{\rm Al}) \cdot \tan(\alpha) - W$  (3.5)

Finally, the designed overlap area A can be calculated as

$$A = B \cdot L \approx 220 \,\mathrm{nm} \cdot 105 \,\mathrm{nm} = 23\,100 \,\mathrm{nm}^2 \,. \tag{3.6}$$

This serves as a benchmark for the junction overlap. It is the aim to fabricate this part of the junction reproducibly. In Ref. [37], the overlap areas are analyzed with an AFM. The results can be seen in Fig. 3.8. For the x-size we have a mean value of  $168 \pm 24$  nm and for the y-size a value of  $278 \pm 23$  nm. These errors are of statistical nature and depend on the accuracy of the top layer mask which suffers from the dose distribution of the e-beam inside the resist. It results in a slight blur of the designed structures. We can view this blur as the main source for deviations in the overlap size. Based on the statistical errors above, we receive a deviation in the area size of 23%.

The measured real overlap size is approximately twice as large as the designed overlap area size. Most of this can be traced back to systematical measurement aberrations, such as the finite diameter of the AFM tip and thermal drifts which occur during scanning.

## 3.2 Cryogenic characterization of DC SQUIDs

Now it comes to a test of the fabricated superconducting circuits. A DC SQUID shows a specific behavior as described in Sec. 1.4. Depending on the applied current, one can



Figure 3.8: X- and y-overlap length of the junctions measured with the AFM: In the AFM micrograph the x-size and y-size of the junctions are marked. The designed size is  $105 \times 220 \text{ nm}^2$ . Sigma denotes the standard deviation. The color code indicates the sample number [37].

distinguish between the resistive and the superconducting state, separated by the critical current  $I_c$ . For further experiments, the focus of interest lies on the critical current density  $J_c$ . In order to use these fabricated Josephson junctions for qubits, a value for  $J_c$  of approximately  $1 \text{ kA/cm}^2$  is desirable. From simulations done at the WMI, we know that this results in a qubit energy range which is easily accessible with microwaves.

Additionally, the dependency of  $I_c$  on the magnetic field is to be investigated. In this case, the DC SQUID acts as a magnetometer and is able to measure single magnetic flux quanta  $\Phi_0$ . The penetrating magnetic flux modulates the critical current of the DC SQUID loop periodically according to Eq. (1.24).

First, we describe the setup for the cryogenic measurement and the measurement method for both the I-V characteristics and the critical current in dependency of the magnetic field. A room temperature pre-check is presented, which is helpful for estimating the critical current density.

Second, we show the recorded I-V-characteristics of our sample and discuss the results. In this section, the progress towards identical critical currents in the desired range is explained. In addition, we illustrate the impact of the oxidation time and of the surface texture on the critical current densities.

At last, a new kind of pre-characterization is introduced. Here, we are able to use the AFM as an instrument of choice. Valuable data can be obtained by scanning the sample. A demonstration for the nondestructiveness of this method will be given by measuring the samples before and after the AFM treatment.

### 3.2.1 Setup and measurement method

Since the critical temperature of bulk aluminum is at 1.2 K, the characterization of these DC SQUIDs can only be done at temperatures lower than that. Therefore a liquid-He3cryostat is used (cf. Fig. 3.9a), which consists of several stages. The outermost stage is an isolation vacuum. A Joule-Thompson process at the liquid-He4 stage lowers the temperature of liquid-He4 to 1.5 K in order to condensate liquid-He3 in the inside. Then, evaporation cooling is performed by pumping at the liquid-He3 volume. In the end, it is possible to reach approximately 500 mK in the innermost stage, where the sample is placed. For a more detailed description of the cryostat see Ref. [18].

#### Pre-characterization at room temperature

Before cooldown, the sample has to be pre-characterized at room temperature. One does not expect to see any quantum phenomena, but it is crucial to check, if all the connections, especially the aluminum bonds, work and if the junctions show an adequate tunnel resistance.

The resistance, which we are able to detect by a 4-point measurement, results from the normal conducting DC SQUID loop and the Josephson tunnel junctions. Measurement feed lines are not included. Moreover, the junctions' overlap area size is approximately the same for all samples. When comparing the resistance at room temperature  $R_{\rm RT}$  with the successively recorded superconducting critical current density  $J_{\rm c}$  (cf. Fig. 3.10), we can clearly observe an exponential relation:

$$R_{\rm RT} \approx 300.4\,\Omega \cdot e^{-J_{\rm c}/1.2} + 96.8\,\Omega \tag{3.7}$$

with  $J_c$  in kA/cm<sup>2</sup>. The thickness of the oxide barrier, which is responsible for the resistance, is small (large) for high (low) critical current densities. As expected, the resistance is governed by an exponential term indicating single electron tunneling. We interpret the offset of 96.8  $\Omega$  to come from the feed lines on the chip which are normal conducting at room temperature. Even though we carry out a 4-point measurement, parts of the feed lines still contribute to the total resistance.

Functional junctions usually show a room temperature resistance of approximately  $100 \Omega$  or higher according to the graph. For future experiments, this may serve as an estimation for critical current densities. Values higher than  $500 \Omega$  indicate a vanishingly small  $J_c$ , whereas values in the range of several k $\Omega$  usually relate to an open circuit.

### Determining the current-voltage characteristic

During cryogenic measurements, the setup is placed inside a shielding room to protect it from noise caused by electromagnetic waves. Furthermore, the outer part of the cryostat consists of a magnetic shielding to prevent parasitic magnetic fields from penetrating the



Figure 3.9: (a) Photograph of cryostat, measurement devices and screening chamber. (b) Photograph of mounted sample in newly designed sample holder: In total, there are 16 feed lines available inside the cryostat. A superconducting coil is placed approximately 2 mm to 3 mm above the DC SQUIDs. (c) Schematic drawing of measurement setup: The DC SQUID is in the innermost part of the cryostat at 500 mK. To perform a 4-point-measurement, the green and blue circuits are needed. The "detect" mode is made possible with the additional purple wires. Furthermore, the temperature at the sample is recorded (orange circuit) and the superconducting coil can be controlled (brown circuit). All the data is collected with a PC running LabView. FGEN: arbitrary function generator, MULT: digital multimeter, CCS: constant current source, VAMP: voltage amplifier, OSC: oscilloscope, LP: low-pass filter, DAK 3k: calibrated resistance for temperature measurements.


Figure 3.10: Room temperature resistance as a function of the critical current density at 500 mK: Red line: Exponential fit. The colors indicate samples from different fabrication runs.

DC SQUID loop.

Figure 3.9b shows the sample holder. At the bottom of the sample holder, a superconducting coil sits directly above the DC SQUIDs with a distance of a few millimeters. Each DC SQUID is connected with four cables. The aim is to perform a 4-point-measurement, which allows to measure the I-V characteristic of the Josephson junction only, without any resistive parts coming from the cryostat leads (cf. Fig. 3.9c). Again the sample is protected with a magnetic shielding cover made of cryoperm.

The circuit is fed with a constant current which splits into the two arms of the DC SQUID (dotted red line). We measure the voltage drop across the DC SQUID. This measurement has to be carried out for several current values in a continuous interval. To this end, an arbitrary function generator drives a constant current source (CCS). In the end, this results in the I-V characteristic of the DC SQUID.

In summary, only a CCS and a voltmeter are needed for determining the critical current, but a detailed investigation of the experimental procedure reveals that low-pass filters are also necessary, because high frequencies inside the cables tend to excite the Josephson junctions and alter the measured results.

#### Recording the critical current as a function of the magnetic field

Another characteristic quantity is  $I_c(\Phi)$ , the critical current in dependency of the magnetic flux. A distinct, periodic pattern of  $I_c(\Phi)$  is to be expected (cf. Fig. 1.6a). The arbitrary function generator sweeps the magnet coil, which produces a magnetic flux penetrating the DC SQUID. In the "detect" mode, the CCS ramps up the current. During this ramping, the measured signal of the voltmeter is fed back into the CCS. When the critical current is reached, the voltage signal disappears due to the superconducting state of the DC SQUID and the CCS stops ramping the current. Now this value of the critical current is recorded by the PC for each value of the magnetic flux.

#### 3.2.2 Current-voltage characteristics and critical current

As described in Sec. 1.4, the I-V characteristics contain three distinct parts. Around zero voltage, we find a supercurrent. Next to it lies the hysteresis part where the current jumps, and on the outermost is the resistive part separated by the gap voltage. Very often, some small steps or wiggles appear in the curve close to the gap voltage value. These can be explained by the existence of quasiparticles and will not be discussed any further in the course of this work.

From an I-V plot we are able to calculate the  $I_c R_n$  product. This has to coincide with the Ambegaokar-Baratoff relation (1.13). Since we use aluminum as our superconductor with  $V_{\rm g, Al} = 360 \,\mu\text{V}$ , we can write

 $I_{\rm c}R_{\rm n} \approx 282.7\,\mu{\rm V}$ 

where we approximate  $\tanh\left(\frac{V_g(T)e}{4k_BT}\right) \approx 1$  for very low temperatures, in our case 500 mK. This is valid for a circuit of two identical Josephson junctions in parallel, that is a DC SQUID, as well as for a single junction, since the prefactors for current and resistance cancel each other in the parallel circuit case.

Moreover, from the hysteresis the Stewart-McCumber parameter  $\beta_{\rm C}$  can be calculated with the use of Eq. (1.19). It describes how strongly the circuit is damped.

First of all, we would like to start this section with the problem of a high critical current spread, which occurred in the beginning of our sample fabrication. Then, we present samples with almost identical current-voltage characteristics and explain the reasons for this result. In the end, we deal with critical currents in the required range.

#### High variance of the critical current

Our first few samples show a very high variance in the critical current of about 500 % (cf. Fig. 3.11). These DC SQUIDs are on the same wafer and come from the same fabrication run regarding evaporation and oxidation (at  $2.18 \cdot 10^{-4}$  mbar for 1530 s). Compared with other oxygen pressures used in this work, this is smaller by a factor of about 0.6. This

leads to a thinner oxide barrier. Essentially, variances in the critical current are generally larger for samples with a higher  $J_c$  due to a thinner oxide barrier.

Moreover, as described in Sec. 2.4.4, oxidation is a complex and sensitive process which may be influenced by a lower pressure. At lower pressures, the errors of the pressure sensor are more pronounced. Hence, in the progress of this work, we move to higher pressures.

A second reason for the high variance is the position of the DC SQUIDs on the wafer which is of importance. Since it is one of our first fabricated samples, a lack of fabrication routine is responsible for a high variance. The developing method is not yet stabilized by the iterative development and therefore causes spatially different results.



Figure 3.11: *I-V* characteristic with high variance in the critical current: All three DC SQUIDs are from the same fabrication run. Inset:  $I_c$  in dependence of magnetic flux which is created by a current  $I_{coil}$  through a superconducting coil.

The almost non-existing hysteresis in the green graph indicates a strongly overdamped circuit. This overdamping can be calculated as seen in Eq. (1.17). The  $I_cR_n$  product is quite small for this DC SQUID. Moreover, the normal resistance is relatively small, as well, resulting in a vanishing  $\beta_c$ . Small normal resistance are found in systems with large normal conducting currents. Hence, this particular DC SQUID might suffer from elevated temperatures or parasitic magnetic fields which cause the supercurrent to be small inside the superconducting aluminum.

The  $I_c R_n$  product is similar for the black and red curve. Compared to the ideal value of 282 µV, it is relatively high. Actually, the theoretical value is the maximum case, since the

voltage gap can only decrease for elevated temperatures. However, a higher  $I_cR_n$  product can be explained with impurities or foreign atoms incorporated into the aluminum layers during evaporation. This causes a higher  $V_g$  and consequently a higher  $I_cR_n$  product which is proportional to the gap voltage.

Another suggestion is that residual resist get incorporated into the aluminum during evaporation. A thin layer of resist might always remain due to the rather weak isopropanol solvent. The chemical process always leaves traces behind, for it is a minimization of the free enthalpy G until  $\Delta G = 0$ . Due to the influence of the wafer's surface potential, we expect small amounts of resist to stay on the wafer.

An indication for this resist layer can be seen in the increased substrate surface roughness (cf. Sec. 3.1.2). A second evidence for residual resist is the fact that higher  $I_cR_n$  products are measurable for all different feed line materials: Gold, platinum and niobium. Hence, it cannot be related to contact difficulties. A further confirmation is the *I-V* characteristic of a sample which is cleaned with the ion gun directly before evaporation. Figure 3.12 shows an *I-V* graph with a much better  $I_cR_n$  product of 269.7 µV. An  $I_cR_n$  product close to the ideal value predicted by the Ambegaokar-Baratoff relation demonstrates a strong Cooper pair tunneling. We conclude that ion gun treatment removes residual resist, which leads to purer aluminum layers.

The modulation of the critical current in dependency of the magnetic flux does not reach zero as it is described by theory (cf. Sec. 1.4). This is an indication that the spatial critical current distribution  $J_c(x,y)$  for each Josephson junction barrier differs from the ideal case. This refers directly to the junction quality. In the picture of the interferometer, the interference pattern becomes blurred due to non-ideal slits.

Another reason for the imperfect modulation can be found in the finite screening parameter  $\beta_L$  of the DC SQUID circuit which is larger than  $\pi/2$  in this case. Due to the intrinsic inductivity of the loop, a screening current causes an external flux and diminishes the supercurrent. Although we are in the case of intermediate screening  $(\beta_L \approx 1)$ , we still use the approximation Eq. (1.26) as an estimation of the screening parameter. Furthermore, the asymmetric design of the DC SQUID (cf. Fig. 2.8) is responsible for a certain amount of flux quanta which do not cancel each other out. This remaining magnetic flux is added to the external flux and hence disturbs the measurement. For higher supercurrents, the disturbance is also higher. Although the modulation is not ideal, still, the maximum supercurrent  $I_{\rm s}^{\rm max} \approx I_{\rm c}$  is reflected in the  $I_{\rm c}(\Phi)$  graph.

From Eq. (1.26) and the minimum and maximum values of the critical current, we can calculate the screening parameter. The calculated values for  $\beta_L$  are indicated in the graphs. Since the DC SQUID loop theoretically remains the same for all fabricated DC SQUIDs, the variance of the screening parameter can only be explained by different inhomogeneities in the superconducting aluminum or in the oxide barrier.

Another reason for the weak modulation is the voltage state of the DC SQUID. If the supercurrent cannot carry the total current, a voltage starts to develop, which leads to

rounded modulation curves as depicted in Fig. 1.7.

Summing up, we find indications for inhomogeneities in the oxide barrier and impurities to be responsible for the anomalous behavior of the DC SQUIDs. This is one of the main obstacles for fabricating identical DC SQUIDs with high reproducibility.



Figure 3.12: I-V characteristic of a DC SQUID on an ion gun cleaned substrate.

#### Towards identical current-voltage curves

The aim of this work is to fabricate Josephson junctions with identical critical currents. Due to problems of the pressure sensor during oxidation with low oxygen pressure, we increased the oxygen pressure to  $3.45 \cdot 10^{-4}$  mbar. In the end, we obtain much smaller critical currents.

The I-V characteristics are shown in Fig. 3.13. Within the limits of accuracy, we can tell that the critical current values, the normal resistances and also the behavior in a magnetic field are identical.

Reasons for this identity are found in the higher oxygen pressure and longer oxidation time. The process becomes less time sensitive, since the beginning and end of the oxidation with their less controlled pressures contribute less. Furthermore, the DC SQUIDs are located much closer to each other on the 1 in wafer than in the former fabrication runs. The distance between the DC SQUIDs is around 1.4 mm as it can be seen in the layout of the on-chip feed lines (cf. Fig. 2.2b). DC SQUIDs which are close together tend to show



Figure 3.13: Identical *I-V* characteristics: DC SQUID 1 and DC SQUID 2 differ only in the quasiparticle tunneling at the critical point. Critical currents and the normal resistance are identical within the limits of the measurement accuracy. The graph, actually a 3-point measurement, is corrected by a constant resistive offset. The original data is plotted as dotted lines. The  $I_{\rm c}(\Phi)$  values are also identical as can be seen in the inset. They exactly fit the theory (green dashed line). Oxidation parameters are 2300 s at  $3.45 \cdot 10^{-4}$  mbar.

a similar behavior. Moreover, the substrate is etched beforehand, which causes a more homogeneous surface structure as described in Sec. 3.1.2.

With 231 µV, the  $I_c R_n$  product is lower than the expected value. This can be traced back to the residual resist incorporated into the aluminum and the non-vertical edges of the overlap area, which result in sloped potential walls. Still, the barrier seems to be homogeneous. This we can tell from the  $I_c(\Phi)$  graph which matches the theoretical fit excellently (green dashed line). We see a reason for the homogeneous on the etched substrate surface which is rougher on the one hand but more homogeneous on the other. Hence, the deposited aluminum and the oxide barrier contain less grains and bumps.

Finally, the very small critical currents come from a thick oxide barrier (approximately  $1.52 \pm 0.26$  nm, cf. Sec. 3.1.1) which is much easier to fabricate in a controlled manner than very thin barriers. Hence, the challenge is to obtain identical DC SQUIDs with a critical current of approximately one order of magnitude higher.

#### Critical currents for flux qubit experiments

For qubit experiments, critical current densities in the range of approximately  $1 \text{ kA/cm}^2$  are required. Beside the correct critical current density value for one Josephson junction, the other prerequisite of identical junctions on one and the same chip holds. If we remind ourselves of the junction geometry, we understand why this is a challenge. The barrier becomes thinner and thinner for higher critical current densities. For this reason, grains or other uneven surface features have a much larger influence on the effective barrier thickness. This constrains the reproducibility of junctions on one wafer.

A method for achieving such thin barriers is to decrease the oxidation time and keep the pressure the same. For this fabrication run, we take less than half of the oxidation time of the former run (900 s instead of 2300 s). After the oxidation, the quartz sensor gave out a frequency change of 11.8 Hz instead of 13.62 Hz. Regarding the thickness estimation, this is 1.23 nm compared to 1.52 nm.



Figure 3.14: I-V characteristic of DC SQUIDs with  $I_c$  in the required range: The variance is significant at such critical current values. It is  $0.92 \,\mu$ A.

In Fig. 3.14, we find measurements of four samples. The critical current densities are around  $1 \text{ kA/cm}^2$ . Some of the samples have proper  $I_c R_n$  products (green and red). Particularly, the red graph comes close to the ideal case. The higher  $I_c R_n$  product of the black curve is related to inhomogeneities inside the aluminum lines, which restrain superconductivity to a certain point and lead to a higher normal current.

For the blue graph we obtain a very low  $I_c R_n$  product. It is the sample with the lowest critical current and hence, in our opinion, it suffers most from flux flow resistances which lead to an adulterated critical current value. Further investigations on the topic of flux flow goes beyond the the scope of this work. A detailed analysis is given in Ref. [47]. In fact, we can eliminate this effect by cleaning the feed line surfaces before evaporation, the critical currents become larger and closer to the real case, leading to a more suitable  $I_c R_n$ product.

The Stewart-McCumber parameter of the samples shown in Fig. 3.14 lies in the intermediate to slightly overdamped case. For the blue curve, the value needs to be adjusted on the basis of a flux flow resistance compensation.

The blue  $I_{\rm c}(\Phi)$  curve has some jumps in the sweep which may come from an incorrectly set threshold value of the "detect" mode. The other curves do not modulate in the full range for the reason of significant screening. From the screening parameter values, we gather that it increases for higher critical currents. This makes sense because larger supercurrents cause a higher self inducted field and hence an enhanced screening.



Figure 3.15: Plot of the screening parameter for different critical currents: A linear relationship between  $\beta_L$  and  $I_c$  can clearly be seen. The loop's asymmetric geometry causes the offset.

In order to analyze the screening of the samples from Fig. 3.14, we plot the screening parameter in dependence of the critical current in Fig. 3.15. We yield the screening parameter values from Eq. (1.26). However, we need to consider, that this equation only holds for  $\beta_{\rm L} \gg 1$ . Still, it serves as an approximation in our case.

According to Eq. (1.25), there is a linear relation between  $\beta_L$  and  $I_c$  with a slope of  $2L/\Phi_0$ . In our case, a linear fit can be applied to the data. The slope is  $566 \pm 157 \,\mathrm{mA^{-1}}$ . From this, we can calculate the inductance which is  $L = 58.5 \pm 16.2 \,\mathrm{nH}$ . With the use of Eq. (1.11) we estimate the inductances' order of magnitude to be around nH for critical currents of 1 µA. This tells us that our estimation of L for our samples is reasonable.

The offset of  $0.84 \pm 0.28$  in the plot comes from the asymmetric DC SQUID design which causes a certain amount of self induced magnetic flux.

#### **Optimizing** $I_{c}R_{n}$ products

For optimizing the junction quality regarding the  $I_c R_n$  product, we have fabricated DC SQUIDs which are cleaned with the ion gun before evaporation. They all show proper  $I_c R_n$  products ranging from 237 µV to 297 µV (cf. Fig. 3.16). Additionally, their critical current density is suitable for flux qubit experiments with a relatively low standard deviation. We obtain  $J_c = 2.9 \pm 0.7 \text{ kA/cm}^2$  with junctions in the size of around 0.085 µm<sup>2</sup> each.



Figure 3.16: *I-V* characteristic of DC SQUIDs which are purged with an ion gun: The critical currents lie in the required range. We obtain critical current densities of  $J_c = 2.9 \pm 0.7 \text{ kA/cm}^2$ . Due to the ion gun treatment during fabrication, the  $I_c R_n$  products are close to the ideal value.

Summing up, it is now possible to fabricate nm-sized Josephson junctions with similar critical current densities which are suitable for flux qubit experiments.

### 3.3 Nondestructive pre-characterization via AFM

It is of great importance that a sample is pre-characterized before a cooldown. One needs to know beforehand if the junctions are fabricated properly and meet the requirements, since cryostat cycling times are in the order of weeks or even months. We are particularly interested in the size of the overlap, which defines the critical current density, the roughness and amount of grains of the aluminum surface and if any breakthroughs result in malfunctioning junctions.

An AFM micrograph contains useful data for the analysis of Josephson junctions. We can tell, if the edges are torn off, and therefore cause a junction to fail due to nonconductive connections (cf. Fig. 3.17d). Besides, the contrary case of short circuited junctions is clearly visible in Fig. 3.17b. This comes from a faulty shadow evaporation, when the top layer mask is broken and too much metal is deposited onto the wafer. In the last case, grains in sensitive regions can be spotted easily (cf. Fig. 3.17c). According to our experience, these grains have an impact on the oxide barrier and hence on the quantum mechanical properties of the junctions. Furthermore, the surface roughness and the geometric properties of the sample can be obtained, such as the overlap area size.



2 µm

Figure 3.17: AFM micrographs of Josephson junctions (faults are marked in red): (a) Flawless junction with rectangular shape. (b) Junction with breakthrough. (c) Junction with grain in the overlap area. (d) Insufficient undercut causes junction with no overlap. Due to the scan method, sometimes line scan artifacts occur, such as in (b).

#### 3.3.1 AFM as the instrument of choice

So far at the WMI, functional junctions have never been directly observed by SEM without destroying them. On the one hand, the risks of using SEM and in this way charging the junctions like a capacitor are too high, since the charge will spoil the thin oxide barrier. On the other hand, an optical microscope does not provide the resolution necessary to see the submicron junctions.

For this reason, atomic force microscopy as a mechanical imaging method seems to be the method of choice. We expect that in tapping mode, the tip of the AFM will not alter the junctions, specifically the oxide barrier. In this section, this hypothesis is to be investigated. Moreover, several other parameters can be gained out of an AFM analysis.

#### 3.3.2 Demonstration of nondestructiveness

In order to demonstrate that an AFM investigation does not alter the quantum properties of a Josephson junction, we perform a cryogenic measurement before and after the AFM investigation. First, we determine if the DC SQUID works at all and measure an I-Vcharacteristics. Then, we carefully disconnect the wafer from the bonds, place it under the AFM and record a micrograph. After all, the sample is reconnected to the sample holder and measured again in the cryostat. Usually, several days lie between these measurements. This could lead to an aging of the oxide barrier.

Figure 3.18 shows the *I-V* graphs of five different DC SQUIDs before and after the AFM investigation. In all cases, the critical currents change only slightly or even not at all. Slight changes may be due to the aging of the oxide barrier of the Josephson junctions. This still has to be verified. The normal resistance value changes a little in graphs (a), (b) and (e). If it changes, the resistance always decreases. According to the Ambegaokar-Baratoff relation, the relation between the normal resistance and the critical current can be described as  $R_n \propto 1/I_c$ . Hence, we see a reason for the lower resistance in an increase of the critical current, which is clearly visible in (a) and only barely visible in (b) and (e). We guess, this is related to a flux phenomenon, where a different amount of flux is trapped during the first and the second cooldown, respectively, and leads to a different critical current [48].

At the moment, a downside of this investigation is the lack of AFM data from qubits. Contrary to DC SQUIDs, qubits have metallic feed line islands which are separated by a Josephson junction on each side. The size of such islands is around  $5 \times 0.5 \,\mu\text{m}^2$  [48]. Hence, charging effects due to the AFM tip, which subsequently destroy the junctions' oxide barrier, cannot be ruled out without testing.

Taking everything into account, we may conclude that AFM is indeed a suitable method for analyzing Josephson junctions before cooldown. One is able to determine the size and the roughness of the overlap region of the junction without altering the oxide barrier significantly.



**Figure 3.18:** *I-V* characteristics of five different DC SQUIDs before and after AFM: (a) and (b) are from one fabrication run and (c) - (e) from another.

## **Chapter 4**

# **Summary and Outlook**

Our main focus in this work is the optimization of the fabrication process regarding reliability and accuracy. We achieve this aim by investigating several aspects of the fabrication process and by establishing techniques to overcome the obstacles.

#### **Resist development**

First, we analyze the development of the e-beam resist. Now, we are able to describe the temperature and time dependence. We find out that if we increase the developer temperature by  $10 \,^{\circ}$ C, it results in a 4.8 times faster development. Hot developer also starts to dissolve the top layer mask. For this reason, we recommend a development at temperatures lower than the room temperature. In order to realize this, we set up a peltier cooling device to maintain a stable development temperature of  $4 \,^{\circ}$ C. At these temperatures, the development does not affect the unexposed parts of the resist and stops automatically. A sufficient undercut can be easily achieved without sacrificing the top layer mask. Our studies show that with this kind of new development technique the precision of the developed structures is only limited by the e-beam resist's resolution and the dose distribution inside the resist.

We analyze the response of exposed resist on IPA developer in dependency of the used dose during e-beam lithography. At different development temperatures different clearing doses are necessary in order to develop an undercut. For room temperature, it requires at least 900  $\mu/\text{cm}^2$ . Furthermore, a larger undercut can be achieved with higher development temperatures at the same dose. In our data, we identify a transition in development speed at a dose of  $1020 \,\mu\text{C/cm}^2$ . We argue that this stems from a change in the molecular structure of the resist. This is supported by a biphasic dose response function.

Another aspect of the new development method is its iterative approach. The development is stopped after a short period of time in order to check the undercut under the optical microscope. During this investigation we discover, that the isotropy of the development is strongly influenced by the evaporation of the developer. Therefore, it is necessary to wash off the developer with deionized water and thus stop the development, before blow-drying the wafer with nitrogen.

#### Evaporation and oxidation

Regarding the evaporation and oxidation, we first analyze the surface roughness dependent on the evaporation rate. In our case, the surface becomes smoother for higher evaporation rates. A smooth and homogeneous surface is an essential basis for the further process. Moreover, the roughness increases from bottom to top regarding the stacking of the layers. Additional homogeneity of the surface of a specific wafer can be obtained by etching the substrate beforehand, although, it increases the overall roughness slightly. This homogeneity of a surface can be observed in the spread of the critical current densities. A low variance of critical current densities can be traced back to a homogeneous surface.

In the course of this work, the mechanisms of the in-situ oxidation of aluminum are studied. We interpret the data logs from the piezoelectric quartz crystal sensor, which is usually used for evaporation rate determination, and discover that it fits the theoretical description of oxidation. First, a monolayer forms, then a tunneling and diffusion process governs the oxidation. The speed of the monolayer formation has been determined to a quartz frequency change of  $1.47 \,\mathrm{Hz}\,\mathrm{s}^{-1}$ . In total, the monolayer formation is accomplished in about 10 s at a pressure of  $3.45 \cdot 10^{-4} \,\mathrm{mbar}$ 

In the tunneling regime, the oxidation takes place much slower. Here, temperature variations in the cooling water cause the graph to oscillate. A temperature change by 1 °C refers to a frequency change of around 10 Hz. For future processes, this has to be stabilized. All in all, the maximum frequency change of one oxidation process levels off around 14 Hz. This refers to 2 to 3 layers of oxide. The surface roughness may also have an influence on the oxidation speed and resulting barrier thickness, since grain boundaries can alter the tunneling.

Compared to the L-product, which has been used for estimating the critical current until now, the quartz data display the trend of the critical currents more accurately. It is the aim for further fabrication to use a quartz based system for oxidation control.

#### Solve connecting issues

In order to investigate connection difficulties, we try three different materials. Gold turns out to be less advantageous for its low adhesion on the silicon wafer. Niobium, which is usually used for resonators and transmission lines, can be used but needs to be cleaned from its oxide right before evaporation in the UHV. For DC experiments with superconducting circuits, platinum is the material of choice because it is easy to handle and sufficiently adhesive. Furthermore, it does not oxidize on the surface, what makes the ion gun treatment omissible. Lift-off problems can be solved by covering the platinum with an additional gold layer. This has to be tried in future.

For the bond to copper pad transition we recommend to use formic acid to clean the copper from its oxide. Then, a strong adhesion between the aluminum bond and the copper pad is guaranteed.

#### Cryogenic measurements

In order to pre-characterize a sample before cooldown, we implement two methods. First, it is possible to 4-point measure the room temperature resistance of the sample. Working junctions follow the relation  $R_{\rm RT} \approx 300.4 \,\Omega \cdot \exp(-J_c/1.2) + 96.8 \,\Omega$  with  $J_c$  in kA/cm<sup>2</sup>. Depending on the auxiliary feed line thicknesses – that is, their resistance – this relation has to be adjusted correctly.

Second, AFM seems to be a versatile tool for investigating Josephson junctions nondestructively. We gain knowledge about surface roughness parameters, geometrical aberrations and breakthroughs from a single micrograph. Moreover, the *I-V* characteristic of the DC SQUID does not change significantly.

During cooldown, we unveil several aspects of the DC SQUIDs' DC-properties. The I-V characteristics reveal that we are able to fabricate identical DC SQUIDs and thus diminish the spread of critical currents. The isotropic development method and the homogeneity by etching play an important role for this. Furthermore, we increase the oxidation pressure in order to improve the signal to noise ratio of the pressure sensor.

The plotted graphs from our DC SQUID samples reveal an offset of the linear branches in the normal conducting regime. We find that single-electron tunneling and flux-flow-like characteristics can be successfully avoided by ion gun cleaning. This removes oxide from the contact pads and residual resist from the silicon surface.

#### Outlook

As an outlook for future fabrication processes, we want to investigate treatments for the already fabricated Josephson junctions, such as an annealing process, for equalizing the critical current densities.

While having recorded the AFM data, one is able to try simulations for predictions of the characteristic parameters, such as  $J_c$ ,  $E_J$  or  $E_C$ . By modeling a Josephson junction as a capacitor on the basis of the surface roughness and taking into account the calculated oxide thickness, the retrieved simulation properties can be compared with the measured properties in order to verify and improve the simulation.

Finally, we aim at applying the new techniques on the fabrication of qubit-resonator or qubit-transmission-line systems. The obtained reproducibility enables us to place multiple qubits on one and the same wafer for investigations on quantum communication or quantum storage. On the basis of this work, where the yield of working junctions has been improved significantly, we want to approach the goal of a quantum computer with multiple qubits with well-controlled properties.

## **Appendix A**

# **Fabrication parameters**

### A.1 Reactive ion etching process

|                  | Reactive ion etching process    |
|------------------|---------------------------------|
| RIE system       | Oxford Instruments Plasmalab 80 |
| $O_2$ flow       | 0 sccm                          |
| Ar flow          | 10 sccm                         |
| $SF_6$ flow      | 20 sccm                         |
| RF power         | $100\mathrm{W}$                 |
| ICP power        | $50\mathrm{W}$                  |
| He backing       | 10 sccm                         |
| Chamber pressure | 15 mTorr                        |
| Strike pressure  | 30 mTorr                        |
| Ramp rate        | 5 mTorr/s                       |

 Table A.1: RIE process parameters

## A.2 Spin coating optical and e-beam resists

Before spin coating, the silicon wafer needs to be cleaned thoroughly with acetone at 70 °C for at least 10 min and then two times with isopropanol in the ultrasonic bath for 2 min at level 9.

|                    | Optical resist with undercut                 |
|--------------------|--|
| Resist             | AZ 5214E resist                              |
| Amount of resist   | wafer fully covered                          |
| Spin speed         | $8000\mathrm{rpm}$                           |
| Spin duration      | 60 s   |
| 1st baking step    | $110^{\circ}\mathrm{C}$ for $70\mathrm{s}$   |
| Flood exposure     | $\sim 3{ m mJ/cm^2}$                         |
| 2nd baking step    | 130 °C for 120 s                             |
| Exposure with mask | $42\mathrm{mJ/cm^2}$                         |
| Developer          | AZ Developer                                 |
| Development time   | $6 \min$                                     |
| Stopping           | 1. water $(2x) / 2$ . blow-dry with nitrogen |

 Table A.2: Spincoating optical resist parameters

|                         | Double layer e-beam resist with undercut                                  |
|-------------------------|---|
| Bottom resist           | PMMA-MA 33 %  |
| Amount of resist        | $440\mu\mathrm{L}$  |
| Acceleration time       | 0.2 s   |
| Rotation speed          | 2000 rpm  |
| Spin duration           | 120 s   |
| Baking                  | 160 °C for 10 min   |
| Top resist              | PMMA 950K A2  |
| Amount of resist        | 220 µL  |
| Acceleration            | $0.2\mathrm{s}$   |
| Rotation speed          | 4000 rpm  |
| Spin duration           | 120 s   |
| Baking                  | $160 ^{\circ}\mathrm{C}$ for $10 \mathrm{min}$                            |
| E-beam dose             | $1020\mu\text{C/cm}^2$ to $1600\mu\text{C/cm}^2$ (depending on structure) |
| 1st development         | MIBK  |
| Development temperature | room temperature  |
| Development time        | 60 s  |
| Stopping                | 1. IPA / 2. water / 3. blow-dry with nitrogen                             |
| 2nd development         | IPA   |
| Development temperature | 4 °C  |
| Development time        | 30-60 s (iteratively)   |
| Stopping                | 1. water / 2. blow-dry with nitrogen                                      |

**Table A.3:** Spincoating e-beam resist parameters

### A.3 Iterative development method

After the sample has been developed with MIBK already, the iterative development with IPA can be applied. First, use the peltier cooling device to cool down the IPA to 4 °C in a clean beaker glass (10 min beforehand). Put your sample into the beaker glass and take it out after about 30 s to 60 s. Use distilled water in a beaker glass to stop the development. Then, dry-blow the sample with nitrogen. Investigate the sample under the optical microscope and check if the undercut is sufficient. An example for an sufficient undercut can be seen in Fig. A.1. If the undercut is insufficient, repeat the IPA step until the undercut is sufficient.



Figure A.1: Controlled undercut and pin sharp top-resist: (a) Micrograph obtained with optical microscope: The dark violet lines represent the openings in the top layer, whereas the light blue parts show the undercut in the bottom layer. Particularly, the regions around the junctions (marked in green) are of special interest, since there the undercut is necessary for shadow evaporation. In this case the undercut is sufficient. (b) Micrograph obtained with SEM: The edges of the junction fabricated with this new method look very rectangular due to a sufficient undercut. For better visibility the edges of the undercut are marked in orange.

|                     | 30-40 nm of niobium                    | $30-40\mathrm{nm}$ of gold or platinum |
|---------------------|--|--|
| Sputtering system   | UHV sputter cluster                    | BAL-TEC MED 020 Coating System         |
| Pre-pressure        | $\sim 10 \cdot 10^{-10} \mathrm{mbar}$ | $< 2.0 \cdot 10^{-5} \mathrm{mbar}$    |
| Ar pressure         | $2.73 \cdot 10^{-3} \mathrm{mbar}$     | $5 \cdot 10^{-2} \mathrm{mbar}$        |
| Ar flow             | $10\mathrm{sccm}$                      | manually so that pressure is correct   |
| Power               | $200\mathrm{W}$                        | _                                      |
| Current             | -                                      | 60 mA                                  |
| Pre-sputtering time | $60\mathrm{s}$                         | -                                      |
| Sputtering time     | $50 \mathrm{s}$ to $60 \mathrm{s}$     | 60 s                                   |

## A.4 Sputter process

Table A.4: Sputter parameters

### A.5 Evaporation and oxidation parameters

Use the correct sample holder and remember the orientation or your sample for the evaporation angle tilt. Load it into the load lock chamber and evacuate it until  $5 \cdot 10^{-7}$  mbar. Be careful not to vent the turbo pump from its exhaust. Open the shutter to the evaporation chamber and slide your sample into the evaporation chamber. Be very careful with the retainer. Lift the sample from the sliding arm with the z-manipulator. Pull back the sliding arm and close the valve to the load lock.

Open the taps for the water cooling first and then turn on the compressor. Turn on the high voltage device. Start the LabView program and use the parameters stated in the table below.

|                         | Shadow evaporation                |
|-------------------------|-----------------------------------|
| Pre-pressure            | $< 10^{-8}\mathrm{mbar}$          |
| Angle                   | $\pm 17^{\circ}$                  |
| Evaporation rate        | $12\mathrm{\AAs^{-1}}$            |
| Thickness bottom layer  | 40 nm                             |
| Thickness top layer     | $50\mathrm{nm}$                   |
| Oxidation time          | around 900 s                      |
| Oxidation pressure      | $3.4 \cdot 10^{-4} \mathrm{mbar}$ |
| Quartz frequency change | around 12 Hz                      |

Table A.5: Evaporation and oxidation process parameters

## A.6 Ion gun cleaning parameters

|                      | Removing resist residuals                       |
|----------------------|---|
| Ion gun              | tectra IonEtch Sputter Gun                      |
| Ar flow              | $0.5\mathrm{sccm}$                              |
| Arm rotation in-axis | 45°   |
| Arm tilt             | $-20^\circ$ off target and $70^\circ$ on target |
| MW power ion gun     | $20\mathrm{mA}$                                 |
| Extraction voltage   | $-600\mathrm{V}$                                |
| Acceleration voltage | $2.4\mathrm{kV}$                                |
| Exposure time        | 60 s  |

 Table A.6: Ion gun cleaning parameters

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