Energy gap measurement of nanostructured aluminium thin films for single Cooper-pair devices

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Abstract
We report measurements of the superconducting gap in Al–Al₂O₃–Al tunnel junctions as a function of aluminium film thickness. Films of thickness 5, 7, 10 and 30 nm were used to form the small-area superconductor–insulator–superconductor tunnel junctions. In agreement with previous measurements we have observed an increase in the superconducting energy gap of aluminium with a decrease in film thickness. In addition, we find grain size in small-area films with thickness \( \geq 10 \) nm has no appreciable effect on the energy gap. Finally, we utilize 7 and 30 nm films in a single Cooper-pair transistor and observe the modification of the finite bias transport processes due to the engineered gap profile.

1. Introduction

By contacting superconducting materials with energy gaps it is possible to modify the energetics of quasiparticle states in different regions of a superconducting nanostructure. This allows quasiparticles to be either confined in, or excluded from, certain parts of a device. This principle of quasiparticle gap engineering is used in photon detection where it is desirable to confine excess quasiparticles resulting from a photon absorption event [1–3]. In addition, the interplay between Coulomb blockade and an engineered gap profile can be used to suppress unwanted quasiparticle tunnelling (QP poisoning) in single Cooper-pair transistors (SCPTs) and Cooper-pair boxes (CPBs) [4–6].

Methods of gap engineering include the use of different superconducting materials such as aluminium and tantalum [1, 3], superconductor–normal metal bilayers [2], oxygen-doped aluminium [4, 7] and more recently different thickness aluminium films [5, 6, 8]. This work focuses on the latter method in which changes in the superconducting gap of more than 50% may be achieved [9].

Early experiments on thin aluminium films observed the enhancement of the superconducting gap and transition temperatures well above bulk values. This enhancement has been attributed to disorder associated with varying grain size, average lattice constant, the presence of an oxide layer surrounding each grain [10–13] and film thickness [9, 10, 14]. More recently, experimental and theoretical studies have investigated aluminium nanowires and observed an increase in the transition temperature with decreasing radius [15–17].

By controlling the superconducting gap using aluminium films of different thicknesses we avoid the introduction of different materials. However, the use of thickness engineered films as thin as 5 nm in Coulomb blockade devices such as the SCPT [8] involves evaporation onto a liquid nitrogen cooled stage and requires repeatable fabrication parameters for electrically continuous films. In addition it is thought that added disorder in such films may induce unusual characteristics such as the presence of multiple superconducting gaps.

Parity of the supercurrent (whether the supercurrent occurs at odd or even integer gate charge) has recently been investigated by utilizing oxygen doping in a gap-engineered SCPT [4]. The parity was found to be strongly influenced by the difference in gap energies of the island and leads, \( \delta \Delta = \Delta_i - \Delta_l \). The explanation given for this behaviour involves the tunnelling of thermal, or non-equilibrium, quasiparticles from the leads onto the device island. The energy cost of transferring a quasiparticle from the leads to the island...
δΔ, consequently alters the corresponding quasiparticle tunnel rates and occupation probability. For the case of positive δΔ the quasiparticle tunnel rates can be suppressed, and the subsequent use of radio-frequency techniques has enabled several experiments in which quasiparticle tunnelling rates were studied on microsecond timescales [6, 18]. The ability to create well-known δΔ in single Cooper-pair devices is the main motivation behind this study.

Here we present measurements of small-area (≤100 × 100 nm²) aluminium superconductor–insulator–superconductor (SIS) junctions. From the maximum in differential conductance measurements we infer the peak density of states, and thus determine superconducting gap energies, Δ, of d = 5, 7, 10 and 30 nm aluminium films [19]. We focus on single thickness film (S1–I–S1) junctions for more reliability in the determination of gap energies. We also investigate films with different grain sizes, by changing evaporation temperatures, to ascertain if there is any appreciable effect on the energy gap. Finally, we briefly present measurements on a gap-engineered SCPT (30 nm–7 nm–30 nm). We discuss both the 2e supercurrent and the finite bias resonances.

2. Thin-film junction fabrication

A typical test sample with a junction area of ~50 × 75 nm² is shown in figure 1. A number of devices were measured with resistances ranging between ~20 kΩ and 2 MΩ. Devices were fabricated on a high resistivity (5 kΩ cm) silicon substrate with 200 nm of thermally grown silicon dioxide on the surface. To enable sufficient contact to the thinner films (≤10 nm) a single layer of PMMA was patterned to define thin overlap areas (~15–20 nm Au/Ti) between the optically defined Au/Ti bond pads and the electron beam lithography (EBL) defined device structure. For the SIS structures a Dolan bridge resist structure [20] was formed from a UV-sensitized resists structure and oxidized in situ with a junction size of a scanning electron micrograph (SEM) of a typical device structure, with a junction size of ~50 × 75 nm². SEM images of the grain structure of an aluminium thin film (thickness 10 nm) in (b) a room temperature evaporated device T ~ 293 K and (c) device evaporated onto a liquid nitrogen cooled stage at T ~ 173 K.

Figure 1. Thin-film Al–Al₂O₃–Al junctions were fabricated using a Dolan bridge resist structure and oxidized in situ. (a) shows a scanning electron micrograph (SEM) of a typical device structure, with a junction size of ~50 × 75 nm². SEM images of the grain structure of an aluminium thin film (thickness 10 nm) in (b) a room temperature evaporated device T ~ 293 K and (c) device evaporated onto a liquid nitrogen cooled stage at T ~ 173 K.

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3. Results and discussion

Measurements were performed in a dilution refrigerator with a electron temperature of approximately 100 mK. The differential conductance (dI/dV) was measured with a standard low-frequency ac lock-in technique with a 10 μV ac bias. Differential conductance as a function applied dc bias voltage is shown in figure 2 for four different thickness films (all evaporated at T_evap = 173 K). We note that no external magnetic field was used to suppress Josephson effects.

In a separate measurement, a thermocouple with similar dimensions (~1 cm²) was used in place of our substrate. From this we estimate the substrate temperature to be T_sub ~ 185 K for cooled evaporations and T_sub ~ T_evap for room temperature evaporations.
is also consistent with the critical temperature enhancements decreasing characteristic dimensions.

There is a clear dependence on the observed $2\Delta$ as a function of film thickness ranging from $2\Delta = 430 \, \mu\text{eV}$ for the 30 nm film (figure 2(a)) to an increase of almost 50% for 5 nm films with $2\Delta = 608 \, \mu\text{eV}$ (figure 2(d)). The differential conductance has been rescaled to a normalized resistance due to differences in junction area, as indicated by the normal state resistances given in figure 2. It is of interest to note that only singular peaks occur in the superconducting density of states. This is opposed to multiple gap structures that have previously been observed in disordered films [22].

Subgap structure appears in the 5 nm and not in thicker devices (see figure 2(d)), perhaps due to the lower junction resistance ($R = 26 \, \text{k}\Omega$) of this sample. We see negative differential resistance regions ($dI/dV < 0$) which lead to peaks in the integrated $dI/dV$ (not shown). Similar behaviour was also seen in a second device ($R = 21 \, \text{k}\Omega$). At present we cannot attribute this behaviour to specific transport processes. However, we note that multiple Andreev reflection is unlikely to be the origin since the positions do not coincide with integer multiples of $2\Delta/n$ [23].

The distribution of the measured energy gap $\Delta$ as a function of film thickness $d$, is shown in figure 3. We observe an increase in $\Delta$ as the thickness of the film is decreased. The magnitude of this increase is consistent with other studies which infer gap energies from the critical temperatures of films [10, 11, 14]. The observed increase is also consistent with the critical temperature enhancements seen experimentally for nanowires [15, 16] and predicted theoretically for nanowires [17] and nanofilms [24] with decreasing characteristic dimensions.

From the standard distribution of gap energies taken at each thickness we see a spread of up to 15 $\mu\text{eV}$ away from average values (table 1). The magnitude of this variation is likely to be due to changes in conditions (e.g. evaporation rate and substrate temperature) between successive evaporations. We support this by noting that junctions made in the same evaporation (for example the 5 nm and the 10 nm room temperature evaporated films) tend to have similar characteristics. The gap energies agree with those obtained in [5] and table 1 indicates the change in $\Delta$ that can be easily achieved both by room temperature and cooled substrate evaporations.

Films with larger grain structure (evaporated at $T_{\text{evap}} \simeq 293 \, \text{K}$, see table 1) have peaks at $eV = 2\Delta$ which fall within the standard deviation of energies for both the 10 and 30 nm films evaporated at $T_{\text{evap}} \simeq 173 \, \text{K}$. Consequently, we see no indication that the grain size has a strong influence on the energy gap. Our results agree with the conclusions of previous investigations [10, 12] that grain size alone does not account for the enhancement in energy gaps.

To review, we have presented differential conductance measurements of small-area SIS junctions which show an enhancement of superconducting gap with decreasing thickness films. A variation of up to 15 $\mu\text{eV}$ from average values for each thickness is accounted for due to variability in evaporation conditions. We see no significant dependence of gap energies on grain size for thicker films.

![Figure 2](image2.png)

*Figure 2.* Normalized differential conductance as a function of applied bias voltage for four different thickness films (evaporated $T_{\text{evap}} = 173 \, \text{K}$). $2\Delta$ was determined from the peak in differential conductance. The $4.2 \, \text{K}$ resistance of each device is also shown. (a) $d = 30 \, \text{nm}$, $2\Delta = 430 \, \mu\text{eV}$, (b) $d = 10 \, \text{nm}$, $2\Delta = 476 \, \mu\text{eV}$, (c) $d = 7 \, \text{nm}$, $2\Delta = 584 \, \mu\text{eV}$ and (d) $d = 5 \, \text{nm}$, $2\Delta = 608 \, \mu\text{eV}$.

![Figure 3](image3.png)

*Figure 3.* Observed superconducting energy gap $\Delta$ of aluminium as a function of film thickness. Each point represents a single junction whose gap energy was extracted via differential conductance measurements. Films were evaporated onto a substrate in contact with a liquid nitrogen cooled stage at a temperature of $T_{\text{evap}} \simeq 173 \, \text{K}$ with grain structure similar to that seen in figure 1(c).

| $d$ (nm) | Number of samples | Approx. Junct. area (nm)$^2$ | $T_{\text{evap}}$ (K) | $\Delta \pm \sigma \Delta$ (μeV) |
|---------|-------------------|-----------------------------|----------------------|----------------------------------|
| 5       | 2                 | $100 \times 100$            | 173                  | 307 ± 3.5                        |
| 7       | 4                 | $30 \times 30$              | 173                  | 298 ± 9.4                        |
| 10      | 8                 | $50 \times 70$              | 173                  | 250 ± 14.9                       |
| 10      | 2                 | $50 \times 70$              | 293                  | 236 ± 3.5                        |
| 30      | 6                 | $50 \times 70$              | 173                  | 209 ± 11.0                       |
| 30      | 1                 | $50 \times 50$              | 293                  | 208                              |

Table 1. SIS parameters for cold and room temperature evaporations: number of samples measured, evaporation temperature, evaluated gap energies and standard deviation for each thickness film.
B corresponds to the lower threshold for the JQP cycle occurring at 2\( \Delta \) of a SCPT device with a 7 nm island and 30 nm leads. thickness across the SCPT. (c) Observed superconducting Coulomb island and the 30 nm leads. (b) Schematic profile of the change in that measured showing the contrast difference between the 7 nm the leads. (a) Scanning electron micrograph of a device similar to eV

D shows the DJQP occurring at

(54 k

SCPT (figure 4(b)). The 4.2 K resistance of this device was determined from normal-state Coulomb diamonds measured at

the 4.2 K resistance and the Ambegoakar–Baratoff relation 

\( E_J \sim \frac{\delta \Delta}{2(\Delta_0 + \Delta)}\) we find \( E_J = 30 \mu eV \).

In the Coulomb diamonds (see figure 4(c)) we see peaks in the reflected power corresponding to 2e-periodic supercurrent in \( n_g \) at zero bias. Supercurrent peaks are not observed in the case of a device without gap engineering and their presence is due to a reduced quasiparticle occupation probability (\( P_{qp} \ll 1 \)) on the island. The appearance of the supercurrent due to the quasiparticle barrier on the island is consistent with previous studies [4, 5].

Coulomb blockade of quasiparticle tunnelling occurs for

\( eV_{ds} > 2\Delta_1 + 2\Delta_i \), and from the threshold of this process we can estimate the superconducting gaps of the leads and island. In this device 2\( \Delta_1 + 2\Delta_i = 1.08 \) meV and, taking \( \Delta_{30 \text{nm}} = 209 \) \( \mu eV \), then \( \Delta_{7 \text{nm}} = 331 \) \( \mu eV \). We note that this is larger than expected for a 7 nm film and is again likely due to be caused by a variation in evaporation conditions. Qualitatively we notice the presence of a large quasiparticle co-tunnelling current in the gap-engineered devices.

At finite bias, but for \( eV_{ds} < 2\Delta_1 + 2\Delta_i \), there are e-periodic features corresponding to the sequential tunnelling of both Cooper pairs and quasiparticles (figure 4(c)). We observe e-periodic peaks related to resonant Josephson quasiparticle (JQP) and double Josephson quasiparticle (DJQP) cycles [26–28]. The JQP cycle consists of the coherent tunnelling of a Cooper pair through one junction followed by two quasiparticles through the other. The condition for Cooper-pair tunnelling must be satisfied and, additionally, the energetics must permit the subsequent tunnelling of two quasiparticles. This results in e-periodic repetition of a pair of crossed lines in the range \( E_C + 2\Delta < eV_{ds} < 3E_C + 2\Delta \). In principle, the primary change to the JQP cycle due to the modified gap is that the thresholds change so that the cycle occurs in the range \( E_C + \Delta_1 + \Delta_i < eV_{ds} < 3E_C + \Delta_1 + \Delta_i \). The lower threshold is plotted in figure 4(c) with the estimated values for the superconducting gaps and appears to correspond to the start of the JQP cycle.

In the DJQP cycle, coherent tunnelling of Cooper pairs between the island and junctions is allowed with quasiparticle tunnelling events determining which junction is on resonance [29]. For the cycle to be possible, the quasiparticle events must be permitted to occur but, due to the need to satisfy Cooper-pair tunnelling across both junctions, its position must remain fixed at \( eV_{ds} = \pm 2E_C \). The DJQP cycle can be seen in figure 4(c).

Additional transport resonances appear just above the DJQP and parallel to the JQP. Unlike the previously described processes, these differ between devices and may be due to transport though higher-order bands in the SCPT or more complex cycles involving both Cooper-pair and quasiparticle tunnelling.

To summarize, the fabrication of a gap-engineered SCPT with a positive \( \delta \Delta \) has a number of different effects on the transport processes. The primary difference between devices with engineered \( \delta \Delta \) and those without is the appearance of a supercurrent which is discussed in greater detail in [4]. Furthermore, the thresholds for Coulomb blockade of quasiparticle tunnelling and the Josephson quasiparticle resonance are shifted. Explanation of the additional resonant features will require a more detailed study of the energetics, perhaps involving knowledge of higher-order bands.

4. Thin films in the single Cooper-pair transistor

The above work makes possible the design of superconducting single Cooper-pair devices with a well-controlled gap profile. In this section we briefly describe electrical transport measurements on such a sample. We made SCPTs using the same fabrication procedure and junction areas as the SIS tunnel junctions (see figure 4(a)). The measurements were performed at milliKelvin temperature in a dilution refrigerator by a radio-frequency reflectometry technique [25]. The SCPT was embedded in a resonant LC tank circuit and the reflection coefficient of an incident radio-frequency signal at the circuit position must remain fixed at

\( E_J = 30 \mu eV \).

4.2 K resistance and the Ambeegakar–Baratoff relation 

\( E_J \sim \frac{\delta \Delta}{2(\Delta_0 + \Delta)}\) we find \( E_J = 30 \mu eV \).

From the SIS results presented above we are able to estimate \( \delta \Delta = \Delta_1 - \Delta_1 \sim 90 \mu eV \) for a 30 nm–7 nm–30 nm SCPT (figure 4(b)). The 4.2 K resistance of this device was 54 k\Omega and the charging energy \( E_C = e^2/2C_S = 190 \mu eV \), as determined from normal-state Coulomb diamonds measured at \( B = 3 \) T. Estimating the Josephson energy per junction from the 4.2 K resistance and the Ambeegakar–Baratoff relation 

\( E_J \sim \frac{\delta \Delta}{2(\Delta_0 + \Delta)}\) we find \( E_J = 30 \mu eV \).
5. Conclusion

We have observed an increase in the superconducting energy gap of aluminium, as a function of decreasing film thickness in small-area SIS junctions. We find that our results are consistent with previous studies and that cold evaporation of thin films produces high-quality small-area junctions which can be used in superconducting gap-engineered devices. Further work needs to be performed to decrease the variability in the measured gap, especially for the thinnest films. For SCPT engineered from different thickness films we observe 2e-periodic supercurrent and a modification of the finite bias transport processes. In addition, for this technique to be used in quantum nanostructures, such as the Cooper pair box, a measurement of the charge noise in the cold-evaporated film and fluctuator density of the tunnel junctions remains to be undertaken.

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