

SUPERCONDUCTING AL-TRILAYER TUNNEL JUNCTIONS FOR USE AS X-RAY DETECTORS

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Abstract – We have developed photolithographic techniques to fabricate high quality Al-Al Oxide-Al superconducting tunnel junctions for use in x-ray detectors. These devices are designed to incorporate $\approx 1\mu\text{m}$ thick superconducting x-ray absorbers for the detection of $<10\text{ keV}$ single photons. In an effort to increase energy resolution, superconductor "bandgap engineering" with lateral and vertical trapping has been used to shorten quasiparticle tunneling times and diffusion lengths and to prevent quasiparticle diffusion away from the tunnel junction. Methods have been developed for overcoming materials incompatibility and device degradation upon thermal cycling; these are reported. We also report on the use of a non-rectangular tunnel junction geometry which reduces the magnetic field needed to suppress the Josephson current for stable biasing. Work in progress to measure the energy resolution of these x-ray detectors at 0.35 K is also discussed.*

polycrystalline or epitaxial Nb exhibit surprisingly short quasiparticle recombination times [3]. There is also evidence that quasiparticle self-recombination in nonequilibrium "hotspots" is the dominant loss mechanism here, and will prohibit the use of Nb x-ray absorbers solely because of niobium's intrinsic properties [4,5]. Much success has been realized with Sn absorbers and Al traps [2], but poor film adhesion prevents thermal cycling or even short-term storage of the devices [6]. In addition, prior to our work, photoresist processing techniques had not been developed for Sn-Al devices; cumbersome mechanical masks were used for that earlier work, restricting geometrical design options. Our work incorporates the best features from the well developed body of work on Nb trilayers and from the Sn-Al work. Refractory tantalum ($T_C \approx 4.5\text{ K}$) is used as an absorber; Al forms the quasiparticle trap. Our devices are thermally cyclable, and theoretically should perform well at temperatures up to 0.3 K .

I. INTRODUCTION

Improved x-ray detectors exhibiting single photon efficiency and very good energy resolution ($\Delta E/E \approx 10^{-3}$) will provide answers to many questions in particle physics and astrophysics. We are concerned primarily with the development of cryogenic detectors for x-ray astronomy applications. Such detectors in satellites like NASA's AXAF will observe K-shell x-rays in the range 100 eV to 8 keV from abundant elements. One of the more promising detector schemes, superconducting tunnel junctions (STJs), exhibits the single photon efficiency needed for faint celestial sources, while potentially offering the energy resolution necessary for determination of atomic ionization structure, source velocity structure, and plasma temperatures, densities, and elemental abundances.

X-rays impinging on STJs create quasiparticle excitations from Cooper pairs, thus producing an increased tunnel current. Although operating in much the same way as a semiconducting p-n junction detector, the STJ creates a far greater number of charge carriers per x-ray (10^6 vs. 10^3), thus resulting in a factor of ≈ 30 better energy resolution. Tunnel junctions may be preferred to cryogenic calorimeter detectors because of faster response times, greater design flexibility, and potentially higher operating temperature. The theoretical [1] and experimental [2] feasibility of STJs as x-ray detectors has been demonstrated, yet further advances must be made before a robust detector is available which fulfills theoretical performance expectations. Niobium-based STJs would be good candidates for x-ray detectors, but several problems have prevented the success of this technology. Thin films of either

II. DEVICE DESIGN AND FABRICATION

Important considerations for the x-ray absorbing thin film include thermal cyclability, x-ray absorption depth, quasiparticle recombination time and diffusion length, and nonequilibrium (hotspot) behavior. Tantalum emerges as the material of choice, although little, if any, previous work has been done with Ta in this application. A refractory metal, Ta exhibits excellent thermal cyclability, thus permitting repeated testing prior to flying in a satellite experiment. Its high atomic number allows one to use films of only moderate thickness to achieve the necessary x-ray absorption efficiency; only $4000\text{ \AA}$ of Ta will stop 20% of 6 keV x-rays. Thin film processing techniques are thus useful for detectors incorporating Ta absorbers. Tantalum's moderately high T_C translates to theoretically long quasiparticle lifetimes at moderately low temperatures of ≈ 0.1 to 0.35 K . Intrinsic modes of quasiparticle recombination in the x-ray absorber as a loss mechanism should therefore not be detrimental to energy resolution. (Unfortunately, little is known about the actual behavior of thin film Ta in this respect, and it could conceivably present problems such as those found in niobium x-ray detectors.) Sufficiently pure Ta can be deposited such that quasiparticles should *theoretically* be able to diffuse across $\approx 0.5\text{ mm}$ absorbers to tunnel junctions before recombining. Lastly, based on quasiparticle-phonon coupling strength, Goldie *et al.* [5] argue that Ta should effectively (i.e., with small quasiparticle losses) dissipate any nonequilibrium regions ("hotspots") caused by the large energy density deposited by an x-ray, with most of the photon energy ending up in the desired quasiparticle excitations.

The tunnel junction used for quasiparticle detection must also meet some stringent requirements: low subgap current (of order or less than that due to the expected signal current

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pulse) for low shot noise, low capacitance for low electronic noise, and good thermal cyclability. In addition, it is highly desirable for the tunnel junction design to be compatible with standard thin film photolithography and trilayer fabrication techniques, mainly for design flexibility and uniform characteristics of many detector junctions in an anticipated 2-dimensional array. Each of these requirements can be satisfied with the use of Al-Al Oxide-Al tunnel junctions.

The use of an Al base electrode and a Ta x-ray absorber also allows us to make effective use of quasiparticle trapping [7] to ensure the best electrical performance. By utilizing quasiparticle trapping to separate the functions of the x-ray absorber and the tunnel junction detector, all requirements can be satisfied for both functions. While optimizing the Ta for x-ray absorption, one can tailor the Al-Al Oxide-Al tunnel junction for optimal tunneling time (which should be much less than the quasiparticle recombination time), capacitance, and readout configuration [2]. Quasiparticle trapping is also used in our devices to inhibit quasiparticle diffusion out the device leads. The leads are made of niobium; its high gap should effectively eliminate the loss mechanism of quasiparticles in the Ta diffusing out the leads [2].

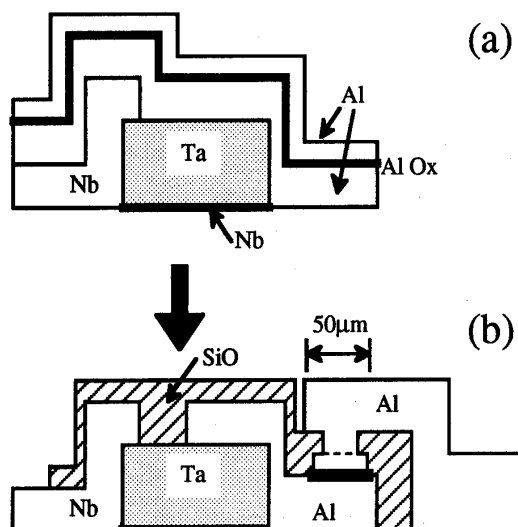


Figure 1: Fabrication of STJ X-ray Detectors (side view)
 a. Al-Al Oxide-Al trilayer deposited over Ta absorber and Nb wiring lead.
 b. Al counterelectrode wiring connected to Al-Al Oxide-Al tunnel junction to form completed device.

The fabrication of the STJ x-ray detectors is outlined in Figure 1 along with approximate dimensions. Tantalum absorber deposition takes place first, with 4000 Å sputter deposited on low resistivity Si in a good vacuum (10^{-7} torr) at $\approx 500^\circ\text{C}$. A thin ≈ 25 Å Nb layer is deposited immediately prior to the Ta to ensure nucleation and stability of the desired bcc Ta phase [8]. This procedure results in Ta with $T_C \geq 4.4$ K and a residual resistance ratio (RRR) of $R_{300\text{K}}/R_{10\text{K}} \approx 6$. This is not outstanding film quality, but should be adequate for our purposes. The Ta absorber dimensions are $\approx 200 \mu\text{m} \times 100 \mu\text{m}$, large enough to allow the use of a FeCl_3 wet etch to

define the structure. Note that the features in these devices are all large enough to allow wet etching with photoresist masks. Also, all materials are compatible with photoresist processing. Device fabrication thus does not require the complexity of reactive ion etching, yet has significantly more flexibility and control than with the use of mechanical masks.

The Nb wiring layer is deposited next, sputtered without intentional heating to a thickness of ≈ 2000 Å. Its RRR of ≈ 4 and T_C of ≈ 9.2 K provide effective trapping against quasiparticle outdiffusion. To prevent unwanted tunnel barrier formation at the absorber-wiring interface, the Ta surface is ion beam cleaned immediately prior to Nb deposition. The Nb wiring features are defined with an etch of 6 HNO_3 : 1 HF : 20 H_2O which does not attack Ta. After this etch we ion beam clean the Nb and deposit the Al-Al Oxide-Al trilayer. The ≈ 2000 Å thick Al base electrode is thermally evaporated at >200 Å/s for best film purity and anticipated long quasiparticle recombination time. We oxidize to form the tunnel barrier in 500 mtorr of pure O_2 for ≈ 2 hours, then evaporate ≈ 800 Å Al at <20 Å/s. (We find that sputtered Al counterelectrodes tend to produce junction short circuits). The Al quasiparticle trap is then defined by a 50°C phosphoric acid etch (PAE), which etches the full trilayer in about one minute. The junction area is defined by a ≈ 2.5 minute room temperature PAE etch, removing unwanted counterelectrode material as well as a small amount of the Al base electrode. The tunnel junction area is approximately $1500 \mu\text{m}^2$, with capacitance ≈ 75 pF, less than that of the cables.

Approximately 1000 Å of thermally evaporated SiO provides junction isolation and a protective coating. It is patterned by photoresist liftoff. The last step is another ion beam cleaning, followed by sputter deposition of a thick (3000 Å) Al counterelectrode wiring layer. This layer is intentionally made thick to reduce unwanted backtunneling effects [9]. This sputtered Al film forms a more reliable short circuit contact to the counterelectrode than would a thermally evaporated film, whose deposition is more gentle. The wiring layer is also patterned by photoresist liftoff. To date we have fabricated only single junction devices, but new masks will soon be developed to make full use of the double tunnel junction geometry of Kraus *et al.* [2]. The fabrication procedures need not be altered.

The large amount of overlap of the Al and the Ta takes advantage of the *experimentally* established [2] long quasiparticle lifetime in Al, and reduces the dependence of device performance on *experimentally* untested Ta quasiparticle lifetime. This novel combination of vertical and lateral trapping makes the quasiparticle diffusion length required to be of order $1 \mu\text{m}$ in the Ta (for x-rays absorbed in the Ta under the Al vertical trapping region) rather than $100 \mu\text{m}$. The Ta area not covered by Al in the present design will be made smaller in a future mask design. The placement of the junction – tens of microns to the side of the large-gap absorber – should provide for very effective lateral trapping, as the Al gap far from the absorber will not be increased by proximity to the Ta film.

III. DEVICE CHARACTERISTICS

Current-voltage characteristics from a device of the type shown in Figure 1 are given in Figure 2. Traces are obtained

in a pumped ^3He dewar with a 0.37 K base temperature. These tunnel junctions exhibit very low subgap currents at low voltages, and have Al base and counterelectrode gaps consistent with very pure Al thin films. An interesting feature is the current rise at $\approx 200 \mu\text{V}$ in the $B=0$ curve. This is most likely a Fiske mode or an unwanted magnetic field penetrating normal to the junction. This feature will be examined in more detail in the future; however, it is effectively suppressed in a parallel (possibly misaligned) magnetic field, so it should not prohibit use of these devices as x-ray detectors.

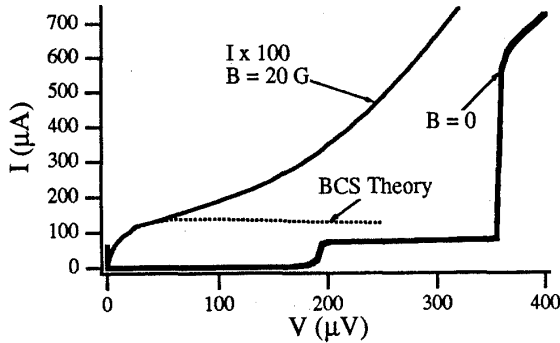


Figure 2: Current-Voltage Trace of Al-Al Oxide-Al STJ at $T = 0.37 \text{ K}$ ($J_c \approx 40 \text{ A/sq. cm}$).

The Josephson current evident at $B_{\text{applied}}=0$ is less than the expected value of $\approx 500 \mu\text{A}$. This is due to either a substantial unintentional parallel magnetic field trapped in the device, or to RF noise; as yet, no attempt has been made to shield the devices from such magnetic fields, but filters have been installed to reduce unwanted RF noise. Other identical tunnel junctions have shown higher critical currents in different runs. Normal state junction resistances of approximately 0.5Ω ($J_c \approx 40 \text{ A/cm}^2$) are typical, making the quasiparticle tunneling time of order $1 \mu\text{s}$. This value is nearly ideal for our purposes when compared with diffusion and recombination times, and when balanced against the need for large subgap resistances. The expanded trace in Figure 2 is obtained in a magnetic field of 20 G from a Helmholtz pair located inside the dewar. The feature near $200 \mu\text{V}$ has been completely suppressed, and subgap currents near $V=0$ are small enough to be attributed to thermally generated quasiparticles (as opposed to barrier defects). The magnitude of the current at low voltages is fit well by an approximation to the standard SIS tunneling current integral [10]. The SIS approximation at higher voltages differs from the experimental trace by an additional exponential term with a SIN-like temperature dependence. There is an increase of the exponential term with magnetic field, indicating that lower fields, better field alignment, or nulling of perpendicular fields should result in more SIS-like behavior. In any case, the exponential term does not dominate below $\approx 50 \mu\text{V}$, and at our bias voltage of $\approx 25 \mu\text{V}$, the tunnel current is sufficiently low. For x-ray detection at 0.37 K, we can stably bias the junction at $1 \mu\text{A}$. Shot noise will dominate the intrinsic resolution of the tunnel junction at this quiescent current, giving an energy broadening of order $\Delta E/E \approx 10^{-2}$.

To achieve theoretically predicted energy resolutions, the subgap current must be reduced. As Figure 3 indicates, one

need only operate at lower temperatures to reduce the subgap current. Measured subgap currents decrease according with temperature according to BCS theory down to 0.37 K, and therefore are not due to tunnel barrier defects. The subgap current should thus continue to decrease in this manner down to dilution refrigerator temperatures [11].

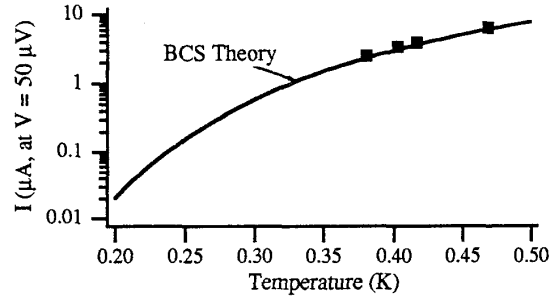


Figure 3: Temperature dependence of subgap current.

IV. MAGNETIC FIELD DEPENDENCE

Externally imposed magnetic fields parallel to the plane of the tunnel junction are essential for stable junction biasing with current sources. This is because, for best electrical performance, junctions are biased at the lowest practical dc current. However, the horizontal load line of a current source may then permit noise-induced switching of the bias point to the Josephson branch, where STJs do not operate as x-ray detectors. A magnetic field parallel to the tunnel junction can suppress the Josephson current and eliminate this branch as a stable bias point. Unfortunately, quasiparticle recombination times are severely shortened by an applied magnetic field with a perpendicular component, as the thin films in STJs have very low critical fields for fields aligned perpendicularly to the film. Such field penetration creates regions of depressed gap, and (undesirable) enhanced quasiparticle recombination. It is thus very important to minimize the magnitude of the perpendicular field. Careful magnet alignment is not necessarily a reliable solution because of fringing fields and undesired diamagnetic effects from nearby superconducting films. Perpendicular nulling coils can be used, but significantly increase the complexity of operation.

The simplest way to minimize problems of misaligned fields is to reduce their magnitude – by reducing the (nominally) parallel field used to suppress the Josephson current, and thereby reducing any associated perpendicular component. One can increase the junction's cross-sectional area, or use non-standard junction shapes [12] (e.g., diamonds rather than rectangles). We have experimented with an approximately quartic-shaped junction (edges defined by a fourth order polynomial) [13]. Figure 4 shows a picture of one of these junctions.

In Figure 5, the dramatic decrease in Josephson current with magnetic field measured for a quartic junction is contrasted with the $\sin(x)/x$ dependence of a rectangular junction. Here, $x = 2\pi\phi/\phi_0$, with ϕ the flux. The Josephson critical current has a zero field value of $380 \mu\text{A}$, and decreases to less than 50 nA at approximately 7 G. This 7 G field is consistent with having one flux quantum through the $\approx 50 \mu\text{m}$ wide junction. The critical current remains at a very low

value for a wide range around 20 G, permitting stable biasing of junctions even with slight drifts in magnetic field. The magnitude of the parallel field is as much as an order of magnitude less than that required for comparable square junctions, as one often has to bias rectangular junctions at higher zeros than the first to ensure a sufficiently small critical current.

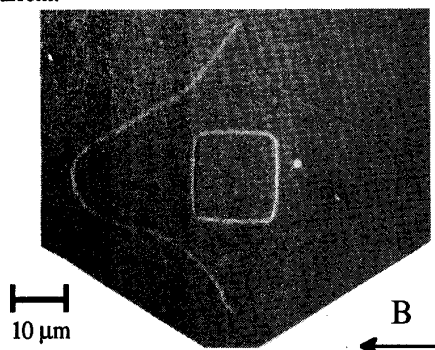


Figure 4: Quartic-Shaped Tunnel Junction. The square feature is a via through the SiO₂.

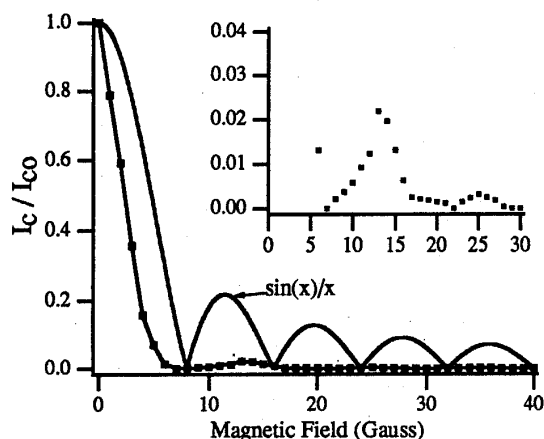


Figure 5: Suppression of I_c by a magnetic field, measured for our quartic junction, compared to the expected $\sin(x)/x$ behavior of a rectangular junction.

In future work, computer generated photolithographic masks will make the shape more precisely quartic. We will also expand the junction dimension in the direction perpendicular to the applied field while keeping the total junction area constant. This will maintain the junction shape important for magnetic field dependence, while lowering the necessary magnetic field density to thread a flux quantum through the junction.

V. DISCUSSION AND CONCLUSIONS

We have successfully fabricated and tested STJs for use as x-ray detectors. Features on these devices are photolithographically defined, making possible great flexibility and ease of device design. Devices with Ta

absorbers have been thermally cycled with no change in current-voltage characteristics. In addition, non-standard junction shapes have been successfully implemented to reduce magnetic fields necessary for stable current biasing.

Preliminary tests of x-ray detection have been performed using a ⁵⁵Fe source (≈ 6 keV) to illuminate the device described in Figure 1. Some evidence of x-ray induced current pulses has been observed, but signal to noise ratios of only ≈ 20 have prohibited more detailed examination of the x-ray response characteristics.

Several avenues are being pursued to increase the collected charge with these devices and the signal to noise ratio. It is apparent that perpendicular magnetic fields are present in our devices, and are certainly detrimental to performance. Perpendicular nulling coils, external field shielding, and new junction mask designs will be implemented to reduce these problems. Sn absorbers will also be studied. Our use of photolithographic processing techniques and a Ti adhesion-promoting layer between Sn and Al should result in useful devices. Tin is not an ideal choice as an absorber material (see the discussion under Device Design and Fabrication), but it has been shown to work rather well [2], and will be useful as a diagnostic test of our tunnel junctions. A recently acquired dilution refrigerator will also be used to ensure low subgap currents and long quasiparticle lifetimes. Finally, changes will be made in the readout electronics and the tunnel junctions to ensure low noise operation, and elimination of signal loss through the dynamic resistance of the junctions.

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