High quality superconducting tunnel junctions on Nb and Ta single crystals for radiation detection

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High quality Nb/Al-based tunnel junctions, fabricated with a superconducting interface onto thick single crystal x-ray absorbers of Nb and Ta are discussed. Current-voltage characteristics, recorded at 0.5 K, show a subgap current which is still dominated by thermally excited quasiparticles. The quality parameter $R_{\mathrm{subgap}}/R_{\mathrm{normal}}$ reaches a value of several million, which is unequalled for nonepitaxially sputtered tunnel junctions. The fabrication process and some development steps, such as preparation of ultrasmooth crystal surfaces are described. Observations of x-ray photons absorbed in Nb and Ta single crystals detected by the superconducting tunnel junctions are also presented. © *1997 American Institute of Physics.* [S0003-6951(97)00526-3]

Over the past decade high energy-resolution detectors for x-ray spectrometry based on superconducting tunnel junctions have been developed by several groups worldwide. A recent overview has been compiled by Booth.[1] Detectors for x-ray astrophysics require high spectral resolution combined with high quantum efficiency and spatial resolution. Nb-, Ta-, and Al-based devices, which survive repetitive thermal cycling, offer, in principle, a high energy resolution, but the thin vacuum deposited layers, used thus far, only provide a low quantum efficiency. Furthermore, imaging is cumbersome, since it can only be achieved by means of detector arrays. The use of superconducting single crystal absorbers, read out by a small number of tunnel junctions, provides a way to combine high spectral resolution with high quantum efficiency and spatial resolution.

The use of superconducting single crystals for cryogenic (particle) detectors has been studied in recent years. Up until now, the best results were obtained with junctions separated from the crystal by an insulating layer, using phonons as an intermediate.[2] Gaitskell[3] succeeded in fabricating tunnel junctions with Pb as a counterelectrode directly on top of a Nb crystal. The junctions however suffered from high subgap current due to normal metallike behavior of the interface layer next to the tunnel barrier. Also the quasiparticle transmission coefficients from the back-sputtered Nb crystal to Al trapping layers, deposited on the surface, were very low. On Ta crystals tunnel junctions with an Al trapping layer were reported, but they suffered from a high superconducting leakage current.[4]

This x-ray detector concept requires many difficult technological issues to be solved, such as growing and refining of high purity crystals, structuring of crystal edges which do not trap quasiparticles, preparation of well defined interfaces between superconducting materials, polishing to obtain smooth surfaces which are necessary for high quality tunnel barriers, and deposition of high quality insulating layers. In this letter we report on significant progress in the development of a process for fabrication of tunnel junctions directly onto a superconducting crystal. Our tunnel junction process,[5,6] developed on Si wafers, forms the basis of the process. We focus on some of the issues which are necessary to obtain proper current-voltage (I-V) characteristics: preparation of smooth surfaces, a good superconducting contact, and proper insulating layers.

The principle of x-ray detection with tunnel junctions can be found in Ref. 1, as well as the mechanism of quasiparticle trapping. In the detector we develop (see Fig. 1) the x-ray photon is absorbed in the single crystal. The created quasiparticles diffuse through the crystal and are trapped in one or two stages near the tunnel barriers, where they can tunnel and be detected. In principle the position of impact can be inferred from the ratio of the tunneled charge in the junctions and the energy of the absorbed photon from the sum of signals.

The Nb and Ta single crystals we used are commercially

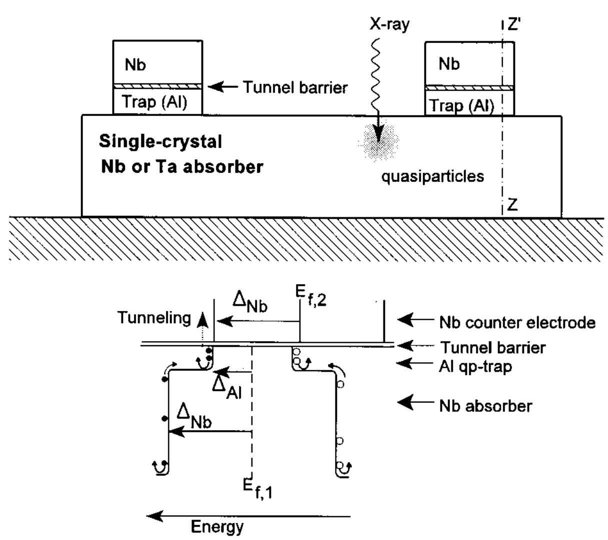

FIG. 1. Schematic representation of the detector concept and quasiparticle potential as a function of vertical position (along a line ZZ' through a junction). The Fermi levels E_f below and above the barrier are different because of the applied bias voltage.

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available 12-mm-diam rods. Slices of typically 300 μm thickness were cut by spark erosion from these crystals. Mechanical polishing was done in several steps with decreasing diamond powder size. The final step contained a slurry with 50 nm Al_2O_3 powder. A rms roughness [determined using atomic force microscopy (AFM)] of about 1 nm has been reached (over an area of 5 μm×5 μm). Since we did not succeed in obtaining a proper superconducting contact to these surfaces, we removed the top layer by chemical polishing. Starting from the etching procedure for Nb of Gaitskell,[3] we determined the mixtures which generated the smoothest surfaces on Nb and Ta. The mixture used for Nb was HF(40%):HNO_3(65%)=1:3 and for Ta HF(40%): HNO_3(65%):H_2SO_4(96%)=1:1:1. Extremely smooth surfaces were obtained by 2–8 min polishing with an etch rate of about 25 μm/min. After polishing the crystals were degassed under vacuum (1×10^{-7} Torr, 860 °C) to prevent formation of surface structures during cooldown to helium temperatures.[3]

Measurements by AFM revealed a wavy surface with height differences of 5 nm for periods of about 10 μm. If we filter out periods larger than 1 μm, a rms roughness of 0.6 nm for the Ta surface and 0.4 nm for the Nb surface is obtained. The residual resistance ratios (RRRs) of these slices were measured with a dc four-probe method. The values for these crystals are still very low, RRR=110 for Ta and RRR=60 for Nb.

Before deposition of sputtered films the native oxide layers were removed by sputter erosion in an Ar plasma. This was done in three steps, the last step with a low bias voltage (ion energy) to ensure that the thickness of the damaged layer is as small as possible. Measurements using micron sized contact areas showed critical current densities $J_c>3\times10^5$ A/cm^2 and $J_c>2\times10^5$ A/cm^2 for Nb and Ta crystals, respectively.

On the polished, degassed and back-sputtered crystal surfaces the following layer sequences were deposited:

On Nb: Nb(200)/Al(25)/AlO_x/Al(5)/Nb(200)/Ta(10).

On Ta: Al(40)/Ta(5)/Al(5)/AlO_x/Al(5)/Nb(200)/Ta(10).

The values between brackets are layer thicknesses in nm. The junction pattern was formed by reactive ion etching and wet etching. On the Nb crystal only the counterelectrode and barrier were structured, so that the base electrode covers the entire crystal (see Fig. 4). On the Ta crystal two etching steps were performed to create a trapping surface (40 nm Al) with a larger area than the junction. The 5 nm Ta on top of the trap serves as an etch stop layer. The wiring is insulated from the base electrode by a SiO layer, structured using lift-off. Because the bonding pads for the counter electrodes were positioned on top of the single crystal the insulation layer must be pinhole free and able to withstand ultrasonic wire bonding. We found that a layer of 400 nm SiO, evaporated at 2 nm/s in two consecutive runs, is sufficient for this task. The substrate must be rotated during deposition. The counterelectrode wiring pattern [Nb(800 nm)/Ta(10 nm)] was deposited using lift-off. The samples were glued to a copper holder with cryogenic varnish and electrical contact was made by Al wire bonding.

I-V measurements were performed in pumped ^{3}He and ^{4}He cryostats at temperatures of 4.2, 1.4, and 0.5 K. Typical results for the Nb crystal are shown in Fig. 2. All but one of the ten junctions show comparable behavior. The current levels scale well with the area, which varies from 300 to 20000 μm^2. The uniformity is better than obtained with the process on standard Si wafers. The gap voltages ($\Delta_1/e=1.4$ mV and $\Delta_2/e=1.1$ mV) can be explained by the proximity effect in the Al layers. The critical current density, calculated from the normal state resistance, is 138 A/cm^2, a value that we also obtain for junctions on Si wafers, oxidized under the same condition.

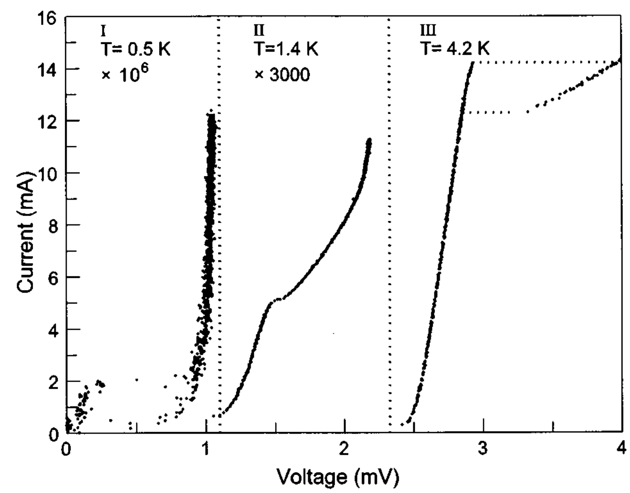

FIG. 2. I-V characteristics of a typical superconducting tunnel junction on a Nb single crystal surface. The area of this junction is 5000 μm^2. Note that the current scales and temperatures T differ in the three regions.

At 1.4 K no ohmic or superconducting leakages could be observed, the junction shows completely thermal behavior. The current increase at 1.1 mV is attributed to multi-particle tunneling. At 0.5 K the subgap current still shows no ohmic behavior, but is three to five orders of magnitude higher than expected on the basis of a simple BCS model for thermally excited quasiparticles. The quality parameter R_{subgap}/R_{normal} at $V=0.4$ mV reaches a value of 5×10^6. To our knowledge this is the highest value ever measured for nonepitaxially sputtered Nb/Al-based tunnel junctions.

The I-V curve of a junction on a Ta crystal at $T=0.5$ K is shown in Fig. 3. Again nine out of ten junctions showed thermal behavior. A clear difference-gap structure is present at 1.0 mV. The gap voltage of the base electrode ($\Delta_1/e=0.3$ mV) is due to the proximitized 40 nm Al trapping layer on the Ta crystal. R_{subgap}/R_{normal} at $V=0.2$ mV is typically $>1\times10^5$. The critical current density is 50 A/cm^2.

For both crystals we have measured the response to irradiation with 5.9 keV x-ray photons. The setup for these measurements has been described elsewhere.[7] X-ray measurements on the Ta sample revealed two kinds of pulses, one with a positive sign from the crystal and another with a negative sign from the counterelectrode, because the bias voltage was below the difference gap voltage. The maximum positive pulse height was 6% of the number of excess quasiparticles created in Ta. A pulse height spectrum measured with a 20 000 μm^2 junction on top of the Nb crystal is

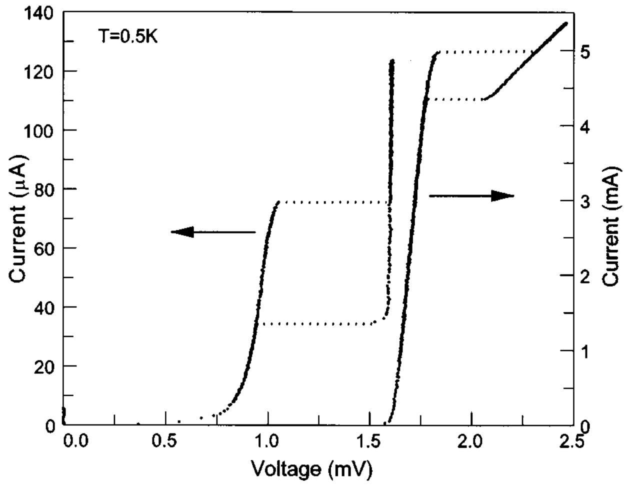

FIG. 3. I-V characteristics at $T = 0.5$ K of a superconducting tunnel junction on a Ta single crystal surface. The area of this junction is 5000 μm^2.

shown in Fig. 4. Based on the count rate in comparison with junctions on Si wafers we attribute the peak at lower channels to x rays absorbed in the crystal. From the count rate ratio of the crystal and the counterelectrode it can be inferred that the sensitive volume of the crystal is about 1×10^5 μm^3. Taking 3.6 μm for the absorption length of 5.9 keV x rays in Nb,[8] we find an effective sensitive area of the crystal 35% larger than the junction area. The effective area is limited by the average time it takes for the quasiparticles to diffuse to the junction. If this time is much larger than the charge amplifier integration time or the quasiparticle lifetime, then the x-ray impact does not yield any observable signal. The sensitive volume is therefore limited by the low RRR of the crystal, i.e., the short mean free path, which gives rise to long diffusion times. For the same reason simultaneous signals in more than one junction from a single x-ray impact were not observed.

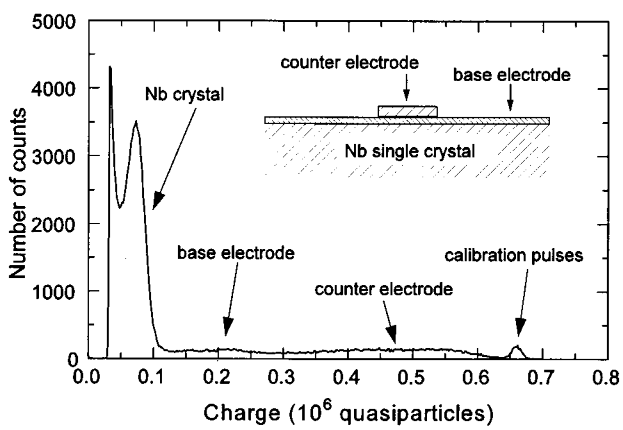

FIG. 4. Pulse height spectrum of 5.9 keV x rays, as detected with a superconducting tunnel junction on a Nb crystal.

A high energy resolution can neither be expected when the diffusion process is too slow. Our next step will therefore be to increase the RRR value of the crystals by ultrahigh vacuum (UHV) annealing. It has been shown by Schulze[9] that it is possible to improve the RRR up to two orders of magnitude, which should be sufficient to collect quasiparticles from the entire crystal within ~ 100 μs. Hereafter only the trapping configuration has to be optimized, since the tunneling time is already of the order of 1 μs. For the realization of a sufficiently long quasiparticle lifetime in the Nb crystal passivation of its surfaces in the annealing system is possibly required.

We have shown that it is possible to fabricate high quality superconducting tunnel junctions directly onto polished single crystal surfaces of Nb and Ta and detect quasiparticles created after absorption of radiation. This is a very important step towards a detector for x-ray astrophysics which combines high energy resolution with high quantum efficiency and imaging properties.

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