ION-BEAM-DEPOSITED FILMS FOR REFRACTORY-METAL TUNNEL JUNCTIONS

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Abstract

We report on the application of a Kaufman ion source to the deposition of Nb and Ta thin films. We find that high quality Nb films (T_c = 9.1 K) can be produced by this technique under tolerant deposition conditions. In addition, substantial, systematic improvement in the I-V characteristics of Nb tunnel junctions has been realized by depositing, in situ, thin (≥10 Å) Ta layers on the Nb film surface.

Introduction

In this paper we report on the ion-beam deposition of Nb and Ta films. The deposition of refractory metal films is of considerable interest because of their desirable superconducting and material properties. In particular, Nb has the highest T_c of the elemental superconductors and is favored for superconducting device applications because of its physical integrity. However, it is well known that pure Nb tunnel junctions characteristically exhibit non-ideal behavior which has been attributed to suboxide growth at the Nb/Nb_2O_5 interface.[1] A very promising approach to ameliorating this situation is to cover the surface of the Nb film, in situ, with a thin overlayer of another metal which can "cap" the Nb surface and form an improved tunneling oxide. Rare-earth metals,[2] Al[3] and, more recently, Zr[4] surface layers have been employed for this purpose. In addition, the use of Si barriers, in conjunction with SNAP, has proven quite successful.[5]

Our approach to the problem of obtaining high-quality Nb-based junctions is to deposit thin Ta layers on Nb. Ta was chosen for this study because of its demonstrated ability to produce very high quality tunneling characteristics.[6] In addition, Ta is quite similar to Nb in its chemical, crystallographic, and normal-state properties. Therefore the overall character of Nb junctions with thin Ta overlayers is not expected to deviate far from those of pure Nb except with regard to an improvement in tunnel-barrier formation. This may be particularly important in obtaining devices with both high-quality I-V characteristics and large and/or reproducible critical-current densities.

We find that the ion-beam technique is particularly adaptable to studies of this kind because of the ease of implementation of a multiple target capability. In our system, any one of four targets can, in rapid succession, be presented to the ion beam for deposition. Many of the unique and promising applications of the ion-beam technique to thin-film studies - such as etching, oxidation, and stress modification - have recently been comprehensively reviewed.[7]

Experimental

As illustrated in Fig. 1, an ion gun is employed to sputter material from a target onto sample substrates. We use an Ion Tech[8] 2.5 cm ion source and a modified water-cooled target holder consisting of a copper carousel on which four 4" sputter targets can be mounted. A different target can be rotated into place under the beam within ∿1-2 sec where it is 15 cm from and inclined 45° to the source. In this experiment we use 99.9+% Nb and 99.95+% Ta targets. The substrate holder

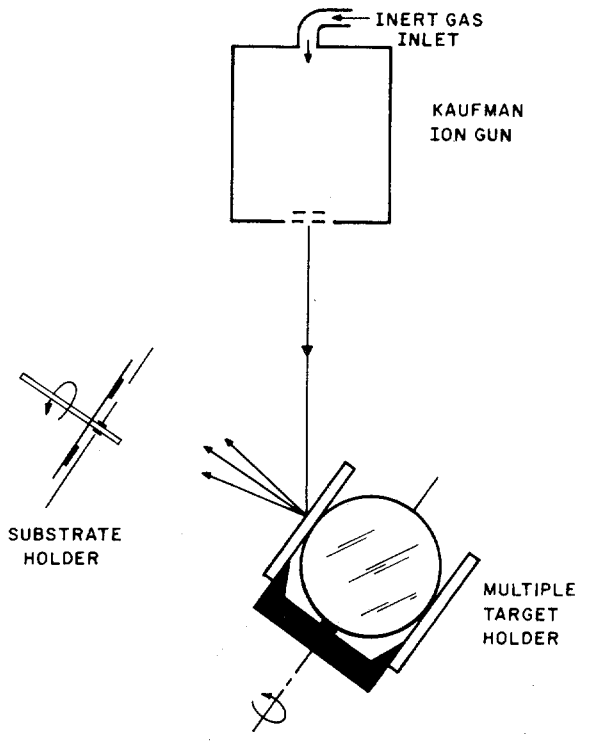

Fig. 1. Schematic of the deposition system, comprising a 2.5 cm Kaufman ion source, multiple target holder accommodating four 4 in. targets, and substrate holder. Not shown is the main sample shutter. Also not depicted is a liquid-nitrogen-cooled copper shroud which surrounds the above components. (Figures not to scale.)

used for thin-film studies is constructed of Nb and Mo to allow for future studies employing heated substrates. It holds up to 8 pair of individually selectable substrates and is 10 cm from the center of and parallel to the target. A similar holder, for depositing thin strips of metal on a number of substrates, is employed in tunnel junction fabrication. Not shown in the figure is a shutter which can be rotated in front of the samples, which typically are polished silicon substrates. During deposition the target and substrate temperatures rise to no more than 55°C and 65°C, respectively.

The apparatus described above and pictured in Fig. 1 is contained in an 18" viton-sealed pyrex cylinder.

Table I: Optimum beam condition for Nb film deposition.

Gas	Pressure[a] (Torr)	Beam Voltage (V)	Beam Current (mA)	Deposition Rate (Å/sec)	T_c[b] (K)
Ar	1×10^{-4}	1300	57	2.2	8.3
Xe	1.7×10^{-4}	1500	35	2.4	9.1[c]

a. Uncompensated ion-gauge reading
b. 2000 Å films
c. a_o = 3.335 Å

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Pumping is accomplished with a standard 6" diffusion pump and a liquid-nitrogen-cooled copper shield which fits just inside the glass cylinder and reduces the base pressure from 6 to 3 x 10^{-7}T when cold. The primary contaminant is water vapor. Research-purity gas was used for all depositions.

Thin Film Results

A fairly lengthy optimization process was followed to obtain the highest quality Nb films, as previously reported.[9] Film quality was judged by both T_c and low-temperature resistivity. The result of this investigation was first that Xe as opposed to Ar gave consistently better films. We note that this trend is consistent with the earlier work of Schmidt[10] in which a hollow-anode ion source was used. The new result here is that ion-beam-deposited Nb films have now been produced with T_c's approaching the bulk value. We note that Bouchier,[11] using a duoplasmatron source, has quoted comparable results for NbTi deposited on heated substrates. We have found that with both Ar and Xe the proper beam conditions are important in maximizing T_c. These optimum conditions are listed in Table I. To obtain high-T_c material we find it necessary to stay within ∿±10% of the listed beam power and to minimize the accelerator current and discharge voltage at a given pressure. We note that once determined, the deposition parameters listed were fully reproducible from run to run.

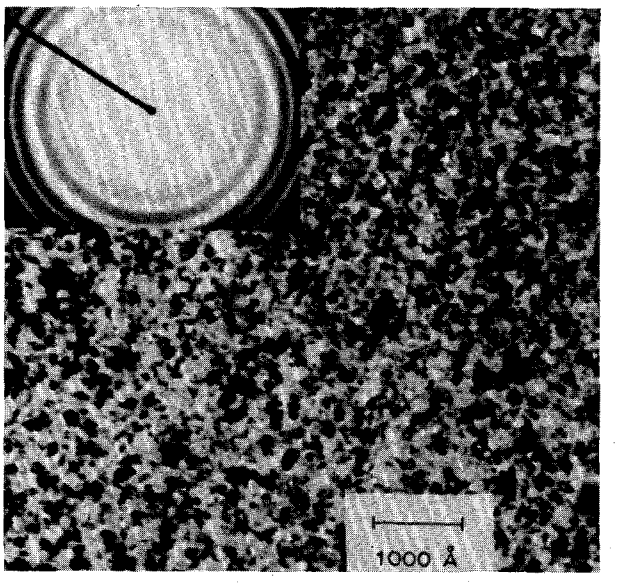

Fig. 2. Electron diffraction pattern and TEM photograph of a 300 Å Nb films on a 300 Å carbon substrate using the Xe deposition conditions in Table I. Results for bcc Ta films (see text) are similar.

Shown in Fig. 2 is an electron diffraction pattern and TEM photo of a 300 Å Nb film deposited on a 300 Å carbon film using the Xe deposition conditions listed in Table I. This photo is representative of films made under a variety of conditions. Based on standard x-ray diffraction scans, we see that our ion-beam-deposited Nb is bcc, (110) textured and has ∿150 Å grains with a random in-plane orientation. The films also appear to be homogeneous and free of voids. We note that preliminary investigations of the impurity content of our highest quality films by Auger analysis and SIMS show the content of C, N, and O to be ≲0.1 at.%. Also, electron microprobe and Auger analysis, with a 0.5 at.% sensitivity, found no detectable levels of Ar or Xe in films deposited with those gases.

We have also examined the transport properties of a number of films. Shown in Figs. 3 and 4 are plots of

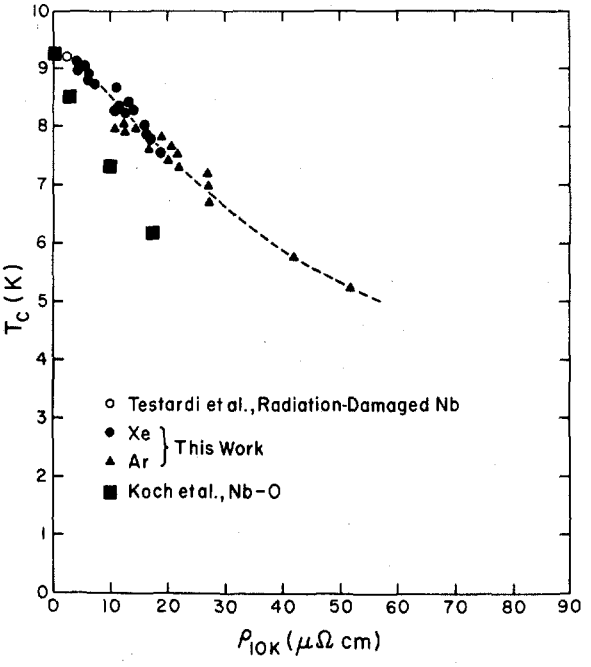

Fig. 3. Superconducting transition temperature of Nb as a function of low temperature resistivity. Results for Nb-O from Ref. 12 and for irradiated Nb from Ref. 13.

T_c as a function of low temperature resistivity (ρ_{10K}) and residual resistance ratio (RRR ≡ ρ_{300K}/ρ_{10K}), for ≳2000 Å thick Nb films. These data were accumulated during our optimization of beam parameters. We note that there is a well-defined, monotonic decrease in T_c with increasing resistivity with an initial rate, $\Delta T_c/\Delta\rho$, of ∿ -0.1 K/μΩcm. For comparison, we show results for oxygen-doped and irradiated Nb. For the Nb-O system[12] $\frac{\Delta T_c}{\Delta\rho}$ ∿ -0.2 K/μΩcm near 9.2 K whereas both the ^{4}He-irradiated sputtered Nb films of Testardi et al.[13] (as shown) and the related work of Ischenko et al.[14] on oxygen-irradiated bulk crystalline Nb show $\Delta T_c/\Delta\rho \lesssim$ -0.07 K/μΩcm. We have also noted[9] that the rate of decrease of T_c with lattice parameter, $\Delta T_c/\Delta a_o$, is considerably slower for our ion-beam Nb as compared to the Nb-O system which show slopes near 9.2 K of 0.05 K/mÅ and 0.25 K/mÅ respectively. These differences in behavior are also manifest in a comparison of T_c versus RRR where it is clear that T_c can vary widely for a given RRR, depending on the particular system of interest. These results illustrate the non-universal nature of the depression of T_c in Nb and suggest that specific mechanisms, such as interstitial gas incorporation in the Nb-O system as compared to radiation-induced lattice damage, must be considered.

These results further suggest a contribution and/or competition of these and perhaps other mechanisms in the decrease of T_c in our films. Indeed, as discussed in the work of Heim and Kay,[15] factors correlated with T_c in sputter-deposited thin films include the energy

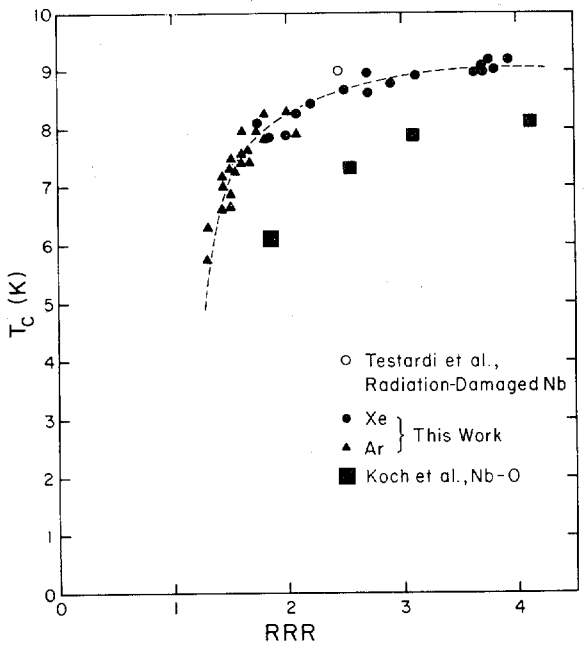

Fig. 4. Superconducting transition temperature of Nb as a function of residual resistance ratio (RRR $\equiv \rho_{300K}/\rho_{10K}$). References for other workers as in Fig. 3.

and species of sputtering gas, residual gas content, sputtering rate, lattice constant, crystallographic disorder, substrate temperature and other quantities which are typically interdependent. We note, however, that because of the independent control over a number of deposition parameters afforded by ion-beam sputtering, a deconvolution of some participating factors may ultimately be possible.

Notwithstanding these detailed considerations, however, we note that Nb of overall high quality can be deposited by ion-beam sputtering under conditions which are relatively "relaxed": a moderate background pressure (3×10^{-7} T), low deposition rate (2.4 Å/sec) and low (<65°C) substrate temperature.

Using Xe gas and the corresponding deposition parameters listed in Table I, thick films of Ta were also investigated. As previously reported in detail elsewhere,[9] it was initially found that on virgin substrates Ta grows in the β-tetragonal phase, which is undesirable due to its low T_c (0.5 K) and high resistivity (>150μΩcm). It was subsequently discovered that by first depositing, in situ, thin layers (∿30-100 Å) of Nb on the substrates, single-phase, (110) textured, bcc Ta (a_o = 3.324 Å) could be grown with $T_c \cong 4.3$ K and $\rho_{10K} \sim 5\mu\Omega$cm. It is likely, therefore, that thin Ta layers deposited on thick Nb films will be strongly predisposed to bcc growth.

Tunnel Junction Preparation

Films for tunnel junctions were deposited using the optimized parameters for Xe gas (see Table I). After a standard 2-3 hour presputter of Nb to getter the chamber, the Ta target (if used) was rotated under the beam for 15 minutes of surface cleaning. The Nb target was next rotated into place, the shutter opened, and 2000 Å of Nb was deposited on the substrates. Immediately following this, the Ta target was simply rotated back under the beam within ∿1 sec to deposit an overlayer of the desired thickness. After deposition, the xenon flow was terminated and dry gas run through the cold shield which returned to room temperature in 1-2 hours. Nitrogen gas was next bled into the chamber and samples were oxidized at room temperature by exposure to air. Composite base electrode edges were masked by painting them with highly diluted resin, preceding the evaporation of an ∿2000 Å thick $Pb_{.71}Bi_{.29}$ counter-electrode. We note that only with the thinnest masking layers did the counterelectrode remain affixed upon thermal cycling.

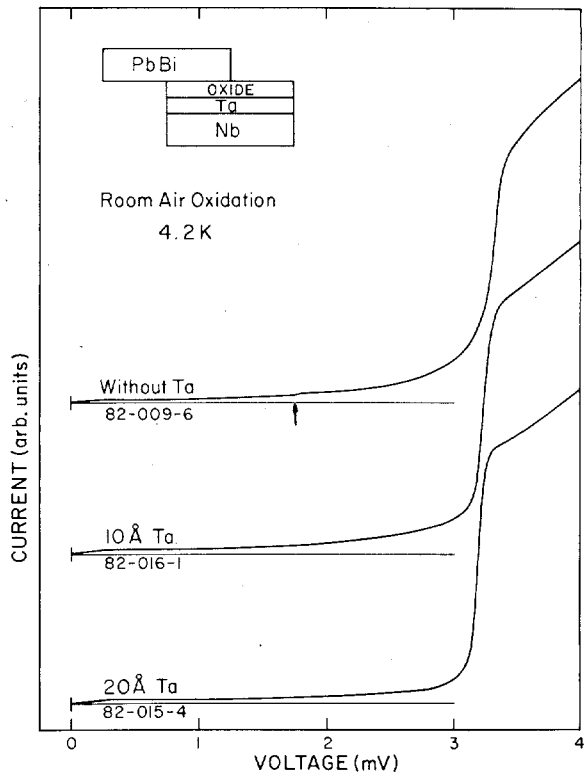

Fig. 5. Current-voltage characteristics of Nb-oxide-$Pb_{.71}Bi_{.29}$ (top curve) and Nb/Ta-oxide-$Pb_{.71}Bi_{.29}$ tunnel junctions at 4.2 K. Indicated is Δ_{PbBi} = 1.75 meV.

Results on Tunnel Junctions

Placing Ta layers over the Nb base-electrode films changed their tunneling properties considerably. Shown in Fig. 5 are junctions prepared with pure Nb base electrodes and Nb with 10 Å and 20 Å Ta surface layers. There is clearly a systematic decrease in the conductance below the sum gap as the thickness of the Ta surface layer is increased. There is also a strong suppression of a conductance increase at Δ_{PbBi} for $D_{Ta} \geq 10$ Å which suggests the inhibition of undesirable normal material formation in the vicinity of the base electrode surface. We note that for $D_{Ta} = 0$ the sum gap is ∿3.2 meV compared to the theoretical value of 3.3 meV (Δ_{PbBi} = 1.75 meV,[16] Δ_{Nb} = 1.55 meV[3]). For D_{Ta} = 10 Å the sum gap increases perceptably to 3.25 meV and then decreases back to 3.2 meV for D_{Ta} = 20 Å.

This suggests the beginning of a proximity-effect depression of the Nb gap. We also note, in this regard, a systematic increase in the strength of the "proximity" knee structure just above the sum gap, suggestive of the presence of residual metallic Ta.

It was found that tunneling characteristics also depended on oxidation time. Pure Nb junctions formed with a 24 hour oxidation were of much poorer quality than those oxidized for 2 hours. This and similar results for Ta layers favored 1-2 hr. oxidations. Although $R \cdot S$ for the pure Nb junctions, $\sim 10^{-4} \Omega cm^2$ ($S \sim 5 \times 10^{-3} cm^2$), is representative of the characteristically slow oxidation of Nb,[3] the junctions with Ta overlayers had $R \cdot S$ values $>1 \Omega cm^2$. This value appears to be representative of a more reactive metal and may be related to the detailed properties of the Ta surface layer. In any case this behavior demonstrates the general integrity of the Ta layer. A more complete understanding of this interesting result will require the adoption of improved procedures for junction definition.

In summary, we have produced both high-quality Nb and Ta on Nb films using ion-beam deposition. Also, thin Ta layers on Nb are shown to effect a significant improvement in tunnel-junction characteristics.

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