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

S. T. Ruggiero, D. W. Pace, and D. E. Prober
Section of Applied Physics, Section Center
Yale University
New Haven, CT 06520

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 ($\approx 10$ A) 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/NbO interface. 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, Al, and, more recently, Zr surface layers have been employed for this purpose. In addition, the use of Si barriers, in conjunction with SNAP, has proven quite successful.

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. 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.

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 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 a target and substrate holder. Our approach to the problem of obtaining high-quality Nb-based junctions is to deposit thin Ta layers on Nb. The substrate holder holds up to four 4" 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.)

As illustrated in Fig. 1, an ion gun is employed to sputter material from a target onto sample substrates. We use an Ion Tech 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 a 1-2 sec where it is 15 cm from and inclined 45° to the source. In this experiment we use 99.94% 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.

<table>
<thead>
<tr>
<th>Gas Pressure</th>
<th>Beam Voltage</th>
<th>Beam Current</th>
<th>Deposition Rate</th>
<th>$T_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Torr)</td>
<td>(V)</td>
<td>(mA)</td>
<td>($\AA$/sec)</td>
<td>(K)</td>
</tr>
<tr>
<td>Ar $1 \times 10^{-4}$</td>
<td>1300</td>
<td>57</td>
<td>2.2</td>
<td>8.3</td>
</tr>
<tr>
<td>Xe $1.7 \times 10^{-4}$</td>
<td>1500</td>
<td>35</td>
<td>2.4</td>
<td>9.1c</td>
</tr>
</tbody>
</table>

a. Uncompensated ion-gauge reading
b. 2000 A film
c. $s_0 = 3.333$ A

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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 $3 \times 10^{-7}$ Torr 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. 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, 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, 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 $\pm 10\%$ 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.

![Electrodiffraction pattern and TEM photograph of a 300 Å Nb film on a 300 Å carbon substrate using Xe deposition conditions in Table I. Results for bcc Ta films (see text) are similar.](image)

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 $\leq 150$ 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 $\leq 0.1$ 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 $T_c$ as a function of low temperature resistivity. Results for Nb-O from Ref. 12 and for irradiated Nb from Ref. 13.

![Superconducting transition temperature of Nb as a function of low temperature resistivity.](image)

![Residual resistance ratio (RRR) of Nb-O films as a function of low temperature resistivity.](image)

$T_c$ as a function of low temperature resistivity ($\rho_{10K}$) and residual resistance ratio (RRR $\equiv \rho_{300K}/\rho_{10K}$), for $\approx 2000$ 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 $\approx -0.1$ K/μΩcm. For comparison, we show results for oxygen-doped and irradiated Nb. For the Nb-O system, $\Delta T_c/\Delta \rho$ is considerably slower for our ion-beam Nb as compared to the Nb-O system which show slopes near 9.2 K whereas both the $\approx 2000$ Å thick Nb films of Testardi et al. (as shown) and the related work of Ischenko et al. on oxygen-irradiated bulk crystalline Nb show $\Delta T_c/\Delta \rho$ $\approx 0.25$ K/μΩcm. We have also noted that the rate of decrease of $T_c$ with lattice parameter, $\Delta T_c/\Delta a_0$, 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/Å and 0.25 K/Å respectively. 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 Hains and Kay, factors correlated with $T_c$ in sputter-deposited thin films include the energy...
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undesirable due to its low T
strates
where, we note that
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(30-100 A)
investigated.
and
pressure
x
Substrates
ubstrate temperature.
ally high quality can be
deposited by ion-beam sputtering under conditions which are
relatively "relaxed": a moderate background
pressure (3 x 10-7 T), low deposition rate (2.4 A/sec)
and low (<65°C) substrate temperature.

Using Xe gas and the corresponding deposition pa-
rameters listed in Table I, thick films of Ta were also
investigated. As previously reported in detail else-
where, it was initially found that on virgin sub-
strates Ta grows in the &-tetragonal phase, which is
undesirable due to its low T
(0.5 K) and high resist-
vivity (>1500Ocm). 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
= 3.224 Å) could be grown
with T
2
4.3 K and p
10K
5uOhm. It is likely, there-
fore, 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. Im-
mediately following this, the Ta target was simply ro-
tated 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 tempera-
ture 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,71Bi,29 counter-electrode. We note that only with
the thinnest masking layers did the counter-electrode
remain affixed upon thermal cycling.

Notwithstanding these detailed considerations, how-
ever, 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 x 10-7 T), low deposition rate (2.4 A/sec)
and low (<65°C) substrate temperature.

Fig. 4. Superconducting transition temperature of Nb
as a function of residual resistance ratio
(RRR = $\rho_{300K}/\rho_{10K}$). 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 sputter-
ing, a deconvolution of some participating factors may
ultimately be possible.

Fig. 5. Current-voltage characteristics of Nb-oxide-
Pb,71Bi,29 (top curve) and Nb/Ta-oxide-Pb,71Bi,29
junctions at 4.2 K. Indicated is
$\Delta_{PbBi} = 1.75$ 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 $\Delta_{PbBi}$ 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 ($\Delta_{PbBi} = 1.75$ meV, $\Delta_{Nb} = 1.55$ meV).
For $D_{Ta} = 10$ Å the sum gap increases perceptibly 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 zero 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 $R_S$ for the pure Nb junctions, $\approx 10^{-4}$ cm$^2$ ($8 \times 5 \times 10^{-3}$ cm$^3$), is representative of the characteristically slow oxidation of Nb, the junctions with Ta overlayers had $R_S$ values $\approx 10$ 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 affect a significant improvement in tunnel-junction characteristics.

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References
