X-ray scattering from multilayers of NbCu

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The question of coherent interfaces between multilayers of sputtered Nb and Cu is addressed using x-ray scattering. It is found that the interfaces are not coherent and the NbCu multilayers are composed of columns strongly layered along the growth direction. The height of the columns is about five times the modulation wavelength Λ. The resistivity increases monotonically with Λ^{-1}, reaching 72.6 $\mu\Omega$ cm for $\Lambda = 18.3$ Å. The superconducting transition temperatures are all below 4.2 K.

I. INTRODUCTION

We present results from x-ray-diffraction experiments on the multilayered system NbCu prepared by magnetron sputtering from two sources.[1] This particular multilayered system has precipitated recent interest with respect to its structural properties. It has been reported that for the first time an artificial superlattice structure has been manufactured where the constituents have different crystal structures and large differences in their lattice parameters.[2] This coherence is surprising in view of the difference in lattice type (bcc Nb and fcc Cu) and the 9% difference between the spacings of the close-packed Nb(110) and Cu(111) planes which form the interfaces. Previous x-ray-diffraction studies have consisted of θ-2θ scans with the scattering vector $\vec{q}$ perpendicular to the plane of the layers. In this report we extend the measurements to include the scattering vector orientations $\vec{q}_\perp$, $\vec{q}_\parallel$ and other general directions with respect to the plane of the layers. The x-ray-diffraction data from our samples show the same $\vec{q}_\perp$ behavior as reported.[2] However, from the additional data we conclude that our layers are in fact incoherent at the interface although they have strong texture in the growth direction.[3] Our data are in semiquantitative agreement with a model where regions of Nb(110) planes and Cu(111) planes are stacked along the growth direction and each region has the lattice parameter of elemental Nb and elemental Cu, respectively. For scattering not along the growth direction there are broad diffraction rings indicating that in the plane of the layers the sample consists of very small grains (less than 60 Å for large

TABLE I. Results of measurements made on samples of NbCu multilayers where Λ is the modulation wavelength. In these samples $\Lambda = t_{Nb} + t_{Cu}$ where t is the individual layer thickness of Nb and Cu, respectively. The total number of layers is increased as Λ is decreased to maintain a total multilayer thickness of 2 μm.

	Λ(Å)	Number of layers	ρ/ρ_0	ρ_0 ($\mu\Omega$ cm)	T_c (K) Sapphire	T_c (K) Kapton
A	18.3	1310	1.017	72.6	1.48	1.55
B	20.9	1070	1.025	55.2	1.85	
C	25.8	905	1.049	45.0	1.51	1.59
D	47.2	411	1.140	20.53	2.12	1.97
E	55.2	377	1.160	20.35	2.30	2.31

Λ) with random orientation. In addition to the diffraction measurements the samples were characterized by measuring their superconducting transition temperatures inductively and by measuring their resistivities using a four-point probe where the contacts were made at opposing sides of the perimeter of the $\frac{1}{4}$-in. sample. The measured characteristics of the samples are summarized in Table I.

II. SAMPLE PREPARATION

To study the possible coherency of any new structure that might be formed, we made samples simultaneously on substrates of sapphire $\langle 1\bar{1}2 \rangle$, Si $\langle 100 \rangle$, Si $\langle 111 \rangle$, and Kapton (Kapton is a trademark of Union Carbide). The latter low absorptive substrate permits transmission x-ray-diffraction studies to be done.

The samples were prepared by mangetron sputtering in a bell jar pumped to 1×10^{-6} torr before sputtering, and the sputtering gas pressure was 2×10^{-3} torr. The argon gas is boiled off from the liquid and then passed through a titanium purifier before entering the bell jar. Substrates are placed on a rotating table 3 in. below the sputtering targets. The sputtering rate for most of these samples was about 14 Å/sec on a stationary substrate. A combination of sputtering rate and table rotation speed determines individual layer thicknesses and thus modulation wavelength. We have found multilayers of NbCu made in this fashion maintain their layered structure over a period of 5 yr and show no significant differences in x-ray analysis as compared to recently deposited samples.

III. RESULTS AND DISCUSSION

Qualitative information about directions for $\vec{q} \neq \vec{q}_\perp$ can be gained through x-ray Read photographs. Photographs from samples A and C are shown in Fig. 1. The sample sits at the center of a 5×7-in.^{2} film rolled into a half cylinder, and the x-ray beam is incident on the sample at an angle α as shown in Fig. 1(c). Careful measurement of these photographs gives d_{hkl}, the interplanar spacing of the (hkl) planes, and χ_{hkl}, the angle the (hkl) planes make with the growth direction. The most obvious attribute of our photographs is the Debye-Scherrer-like arcs, indicative of a polycrystalline sample. The arc length of the diffraction lines is relatively short due to preferred orientation, and the width of the lines is 3 to 5° wide in 2θ. The sample is composed of rather small grains in preferred orientation, with their close-packed directions Nb(110) and Cu(111) roughly in the growth direction.

Quantitative measurements were made on a diffractometer equipped with a quarter circle Eulerian

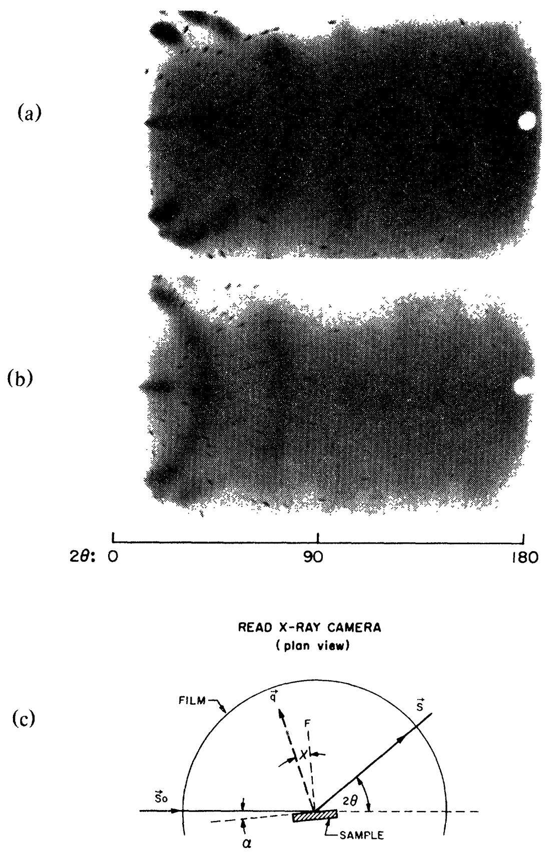

FIG. 1. X-ray Read photographs of sample C and sample A, respectively; the sharp spots are scattering from the single crystal substrate. (a) For $\Lambda = 25.8$ Å the scattering intensity from the sample appears broad and arclike. (b) When $\Lambda = 18.3$ Å the sample scattering becomes broader and the arc length increases considerably. Also note the broad diffuse band at low 2θ. (c) The geometry of the Read camera defined in the cross section of the camera.

Cradle so that θ-2θ scans could be made with the scattering vector at an angle χ from the growth direction and so that the scattering vector $\vec{q}$ could be rotated through an angle ϕ around the growth direction. χ scans around the average $\vec{q}$ of Cu(111) and the Nb(110) give a full width at half maximum (FWHM) varying between 6° and 10° for the samples in Table I. Once the growth axis is established, the direction of other reciprocal-lattice vectors can be calculated. To check for preferred orientation in the plane of the layers, a ϕ scan was made of the Nb(211) for which $\vec{q} = 4.79$ Å^{-1} and χ is equal to 30°. The scattered intensity was constant for the Nb(211) ϕ scan and for other ϕ scans made on all the samples, independent of Λ. The χ and θ scans show that these films are essentially like fibers showing strong texture in the growth direction but with the grains oriented randomly in the plane of the film. Below we discuss $\vec{q}$ scans made on samples A and C at $\chi=0°$, 70°, and 90° (see Fig. 2).

The scans with $q_\perp$ ($\chi=0$) were done on samples deposited on sapphire substrates. We found diffraction peak intensities from samples deposited on sapphire substrates to be slightly greater and sharper than samples on Kapton or silicon, but the positions of the diffraction maxima were independent of substrate type. Below $2\theta=10°$ we observed superlattice reflections of different orders for samples where $18.3 \leq \Lambda \leq 55.2$ Å. The modulation wavelength Λ determined from these superlattice reflections is in good agreement with that calculated from the sputtering rate and table rotation speed. It also agrees well with the Λ calculated from the position of the satellites.[4] At larger $\vec{q}_\perp$ the Bragg peaks and associated satellites evolve as shown in Ref. 4, and our results are in qualitative agreement with that data. The width of the Bragg peaks are resolution limited where the resolution of the diffractometer is $\Delta 2\theta=0.3°$ at

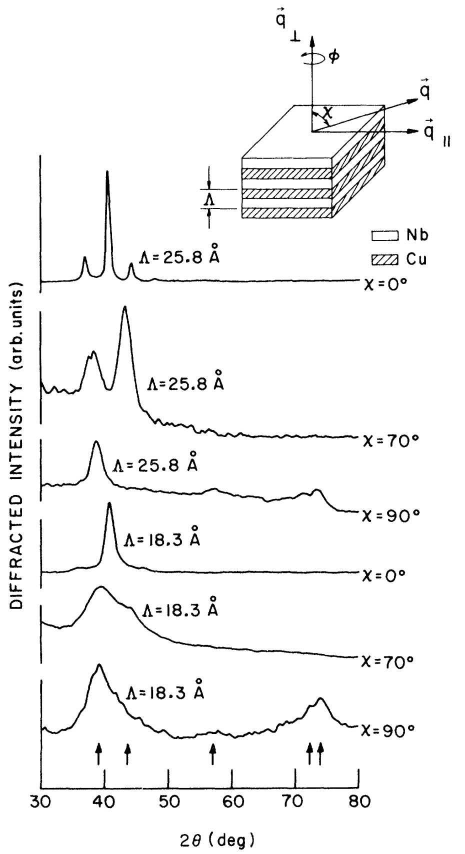

FIG. 2. Longitudinal θ-2θ scans in reciprocal space making angles $\chi=0°$, 70°, and 90° with the growth direction for sample C and sample A, respectively. The arrows point out the d values of the bulk Nb(110), Cu(111), Nb(200), Nb(211), and the Cu(220), respectively.

$2\theta=35°$. The satellites also become quite broad at short Λ indicating that the multilayers are becoming disordered and the grain size is decreasing. We estimate from our x-ray-peak width using the Scherrer equation the grain size to be about 60 Å in the growth direction for our smallest Λ. In all probability under current sputtering condition the sample would be even more disordered at shorter wavelengths.

Since the Cu(111) planes are along the growth direction the Cu$(1\bar{1}1)$ planes must make an angle of 71° with the growth direction. When χ is set to 70° (sample C) a θ-2θ scan shows two Bragg peaks (Fig. 2); the second of the peaks is indeed the Cu$(1\bar{1}1)$ and it gives a lattice constant $a_0=3.61$ Å. This is the lattice constant of bulk copper which indicates that the copper layers are not distorted from their bulk structure. The first peak at $\chi=70°$ and the peak at $\chi=90°$ are the Nb$(10\bar{1})$ and Nb$(1\bar{1}0)$ reflection, respectively, and they give $a_0=3.30$ Å, which indicates that the niobium also maintains its bulk lattice constant. The first peak would be centered at $\chi=60°$ but is also observed at $\chi=70°$ because of the poor texture.

Setting $\chi=90°$ and making θ-2θ scans allow us to study the correlation of the atomic planes perpendicular to the substrate. The x-ray linewidth shows the intraplane correlation to be about 10 Å when $\Lambda=18.3$ Å and about 70 Å when $\Lambda=55.2$Å. For sample C the Cu$(1\bar{1}1)$ does not appear since the copper is highly textured but in sample A the layers are becoming disordered and there are traces of Cu$(1\bar{1}1)$ at $\chi=90°$. We also measured other texture forbidden lines in sample A.

Coherency in a multilayer structure requires that there is a reciprocal-lattice vector $\vec{\tau}$ defined by the equation $\vec{\tau}=h\vec{a}^*+k\vec{b}^*+l\vec{c}^*+m\vec{w}$ where h, k, l, and m are integers, $\vec{a}^*$, $\vec{b}^*$, and $\vec{c}^*$ define the average lattice, and $\vec{w}$ the composition modulation. If there is a small mismatch between layers at the interface it can be accommodated by coherency strain to give a single $\vec{\tau}$ as, for example, in $GaAs_nAlAs_m$ and Cu_mNi_n.[5,6] The x-ray data obtained in directions with χ not equal to zero show no evidence for sizable lattice distortion from the d spacing expected for the pure elements. This suggests that there is no coherence at the interface and that the layers can be thought of as an incoherent stack of alternating slabs of Nb and Cu.

The resistivities and T_c's measured on all the samples are given in Table I. All the samples have critical temperatures below 4.2 K, so we were able to measure the residual resistivity conveniently at 4.2 K. The resistivities are very high for small values of Λ reaching as high as 72.6 $\mu\Omega$ cm in contrast with 14.5 $\mu\Omega$ cm for bulk Nb and 1.7 $\mu\Omega$ cm for bulk Cu. The resistivity of the samples increases with decreasing Λ and correspond to the previously reported results[7] (see Fig. 3). There are two probable causes for the excess resistivity: One is the electron scattering off

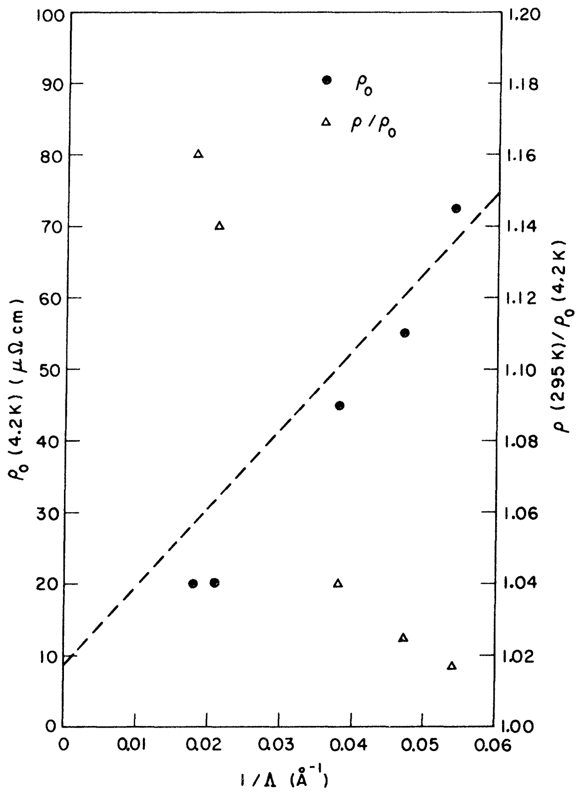

FIG. 3. The resistivity $\rho(4.2\ \mathrm{K})$, and the resistivity ratio $\rho(295\ \mathrm{K})/\rho_0(4.2\ \mathrm{K})$ vs reciprocal modulation wavelength. The broken line is a fit of resistivity vs inverse modulation wavelength taken from Ref. 7.

the interfaces which increase in number with decreasing Λ; the second is electron scattering at grain boundaries which also increase with decreasing Λ.

IV. SUMMARY

In summary, we have made, using the technique of magnetron sputtering, multilayers of NbCu with modulation wavelengths as short as 15.3 Å and containing as many as 1500 periods. It is also demonstrated that, by making x-ray photographs using a simple geometry, such questions as preferred orientation, structural coherence, and grain size can qualitatively be answered. We find from the x-ray photographs that the material is composed of small horizontal islands with texture; and, in the case of $\Lambda = 18.3$ Å, the islands are surrounded by disordered material. The islands exhibit a wire texture in which there is one axis of symmetry about which each island is randomly rotated through some angle ϕ. The diameter of the islands we estimate from our x-ray peak width to be not more than 60 Å, for $\Lambda = 55.2$Å. We have also shown that scattering in a plane containing the growth direction($\vec{q} = \vec{q}_\perp$) is vastly different than when the scattering plane is parallel to the multilayers, and that this vast difference can be attributed to small grain size and the fact that each layer of Nb and Cu maintains its bulk lattice parameter. The measured resistivities were high with a strong Λ dependence. The superconducting transition temperatures were all below 4.2 K and, in general, increased from 1.5 to 2.3 K as Λ increased from 18.3 to 55.2 Å. The T_c's measured from samples on Kapton agreed well with those measured on samples deposited on sapphire. We have not analyzed the T_c in accordance with proximity effect theories but note that the gradual increase in T_c with the increase in Λ is in general what should be expected.

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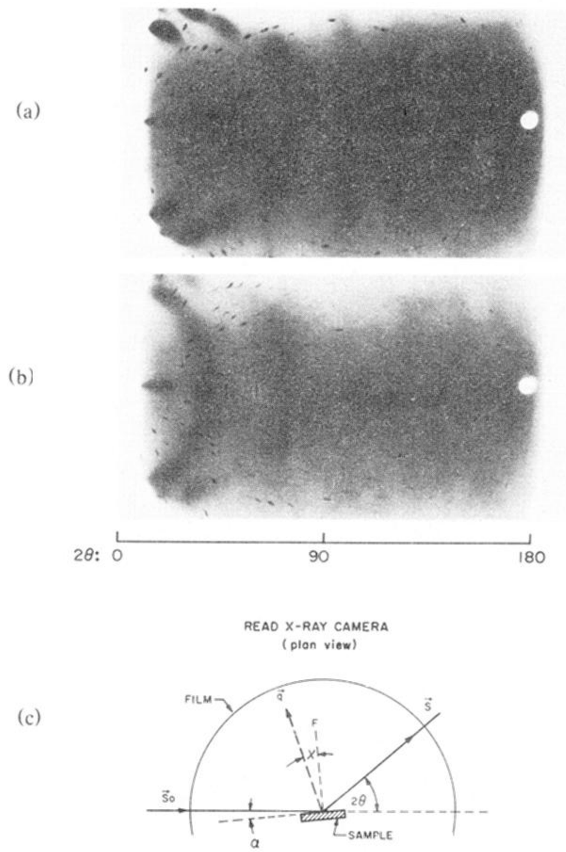

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