Temperature dependence of the resistivity in Cu/Ti superlattices

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In this paper we report resistivity measurements as a function of temperature for Cu/Ti superlattices of periods 20, 50, 100, 200, and 400 Å. We find that the in-plane resistivity is inversely proportional to the layer period; in the sample with the smallest period, there is a sharp drop to a negative value of the temperature coefficient of resistivity, which is positive and roughly constant for the other layer periods. This agrees with results for the Cu/Nb superlattice and indicates weak localization in the 20-Å sample.

I. INTRODUCTION

Recently there has been extensive interest in the properties of metallic superlattices[1-4] since they are new types of materials with novel structural, electronic, and mechanical properties. Of particular interest are the transport properties and their dependence on superlattice periodicity. Temperature and layer-period dependence of the resistivity have been studied in Pd/Au (Ref. 5) and Nb/Al.[6] Werner *et al.* reported the first observation of a negative temperature coefficient of resistivity (TCR) in Nb/Cu superlattices.[3] The TCR, in fact, was found to change sign sharply as the period λ was decreased to the lowest values. For these ultrathin layers ($\lambda \approx 10-20$ Å) the superlattice resistivity was found to be close to 150 $\mu\Omega$ cm, which is the critical resistivity separating positive and negative TCR's in the Mooij plot.[7] To our knowledge no other evidence of negative TCR in metallic superlattices has been published. According to this result, it would seem that ultrathin superlattice films feature the same phenomenology as amorphous metallic alloys.[8]

In order to confirm and extend this result, we have studied the resistivity of Cu/Ti superlattices as a function of period and temperature. We find that the resistivity ρ varies inversely with the period λ, in agreement with previous results; furthermore, the TCR for the 20-Å superlattice is negative, switching from the essentially constant positive values it had for all the other superlattices we have studied.

These data are interpreted in terms of current theory; in particular, for the more standard superlattice behavior we use a modified[9] Soffer theory.[10] The more anomalous behavior is discussed in the framework of weak localization[11,12] in disordered metals.

II. EXPERIMENT

The samples were grown on glass substrates by magnetron sputtering; the initial vacuum was 10^{-6} Torr and sputtering was done in an ambient pressure of 7.5×10^{-3} Torr of argon gas. Deposition rates were 20 Å/sec for both Cu and Ti. The Cu and Ti layers were of equal thickness. Five samples were prepared, with period varying from 20 to 400 Å, for a total thickness of 2 μm for all samples with the exception of the λ=400 Å sample, which was 0.5 μm thick.

The in-plane electrical resistivity was measured by a balanced-bridge ac four-point probe technique at different temperatures ranging from 3.7 K to room temperature. The samples were immersed in liquid helium or in the helium exchange gas of a cryostat; temperature was controlled with a microprocessor-based system and temperature stability was better than 0.1 K. Measurements were performed both upon heating and cooling and no appreciable difference was observed in the data.

All the samples were examined by x-ray diffraction. The results show that Cu and Ti layers had strong (111) and (002) textures, respectively. There was apparently very little interdiffusion and the interfaces were sharp for all samples. Detailed results are described elsewhere.[13,14]

III. RESULTS AND DISCUSSION

In Fig. 1 we show the temperature behavior of the in-plane resistivity for Cu/Ti superlattices with different periods λ. We have chosen to show the behavior of $\Delta\rho = \rho - \rho_0$, where ρ_0 is the residual resistivity, since on the scale of the absolute resistivity the temperature variation was too small to be observed clearly in the figure. Note the change in slope between the λ=50 and 20 Å samples

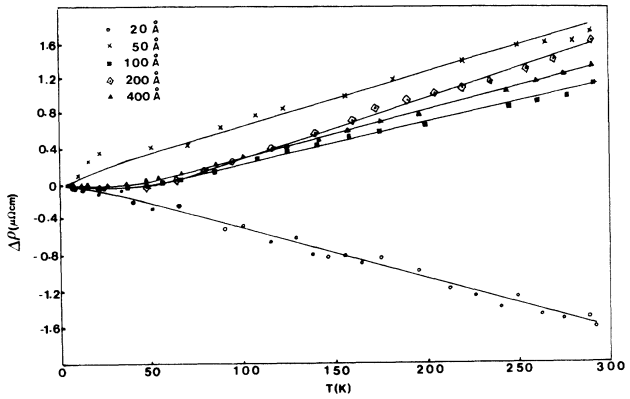

FIG. 1. Temperature dependence of resistivity in Cu/Ti superlattices.

for $T \geq 30$ K. In Fig. 2 we show the behavior of the residual resistivity ρ_0 versus λ^{-1}. The proportionality to the inverse period is clearly verified by our data.

In Fig. 3 we show the results for the TCR. Although we believe the most physically significant quantity is the absolute TCR (i.e., in units of $\mu\Omega\,\mathrm{cm\,K^{-1}}$), for comparison purposes we have also plotted the relative TCR (in units of $\mathrm{K^{-1}}$). Whereas the relative TCR varies smoothly with λ, in agreement with results for Pd/Au superlattices,[2] the absolute TCR is essentially independent of λ, but sharply drops to a negative value for the smallest period investigated, in good agreement with the behavior observed in Cu/Nb superlattices.[3]

In order to interpret our results, we start using modified Soffer theory as extended to superlattices by Shiroishi *et al.*[1] Thus the resistivity of the metallic superlattice can be written as

$$\rho = \frac{m v_F}{n e^2}\left\{\frac{1}{\bar{\ell}} + B + \frac{3\tilde{\sigma}}{4\sigma_\infty} \times \left[\left(\frac{\bar{\sigma}}{\tilde{\sigma}} - 1\right) + \frac{C}{3}\left(\frac{4\pi}{\lambda_e}\right)^2 \Delta^2\right]\frac{1}{\lambda}\right\}, \tag{1}$$

where C is a constant of the order unity, λ_e is the electron wavelength, Δ is an electrical interface roughness which is assumed proportional to the crystallographic interface roughness, and

$$\tilde{\sigma} = (\sigma_1 \ell_2 + \sigma_2 \ell_1)/(\ell_1 + \ell_2)\,, \qquad \bar{\sigma} = (\sigma_1 \ell_1 + \sigma_2 \ell_2)/(\ell_1 + \ell_2)\,, \tag{2}$$

where σ_1 (σ_2) and ℓ_1 (ℓ_2) are, respectively, the conductivities and mean free paths in metals 1 (in metal 2). In Eq. (1) the first term on the right side is due to phonon scattering and the second term is due to grain-boundary scattering; in a metallic superlattice we expect the latter term to be small and weakly dependent on λ, and shall heretofore neglect it. The third term, due to interface scattering, yields the dominant contribution to the residual resistivity. For a Cu/Ti superlattice, given the large difference between σ_1 and σ_2, we expect[6]

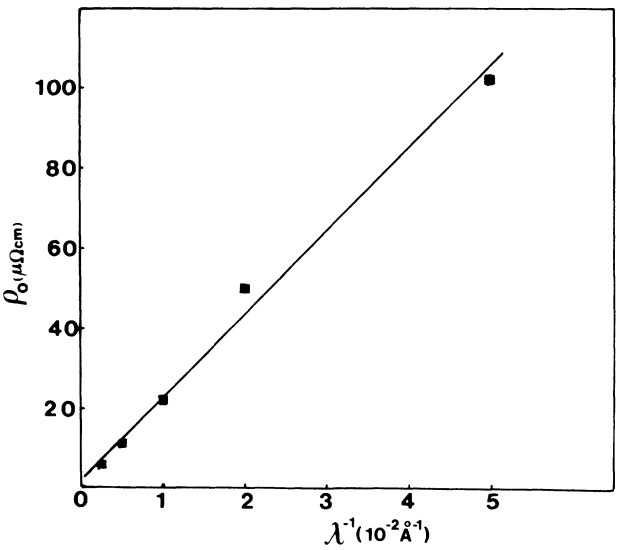

FIG. 2. Residual resistivity vs inverse superlattice period.

$$\frac{\bar{\sigma}}{\tilde{\sigma}} - 1 \gg C(4\pi)^2\Delta^2/3\lambda_e^2\,, \tag{3}$$

and therefore

$$\rho \approx \frac{1}{\lambda}\,. \tag{4}$$

Equation (4) is consistent with our results; however, the fit to the data yields a slope which is twice the expected value if approximation (3) were valid. In fact, a specific calculation shows that in Cu/Ti the two terms in (3) have roughly the same value, in agreement with our experimental result. This does not mean, however, that Eq. (4) is not followed, as might be expected from the known dependence of Δ on λ. In fact, this dependence saturates for $\lambda \gtrsim 20$ Å.

The measured resistivity of our samples is 3 times as high as in Pd/Au, and twice as high as in Fe/Cr superlattices. From the point of view of materials, a reason for this can be found in the fact that Pd and Au have

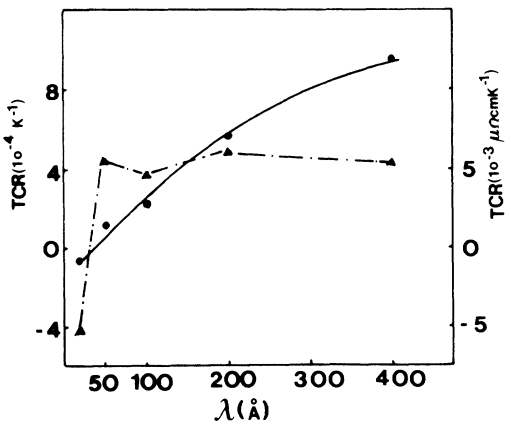

FIG. 3. Superlattice-period dependence of absolute (dash-dotted line) and relative (solid line) TCR.

face-centered-cubic structures with very similar lattice parameters; thus in this system the interfaces are well matched by a coherent strain field. The same holds for the Fe/Cr superlattice. On the other hand, there is a comparatively much larger difference in atomic distances between the Cu(111) and Ti(002) textures, and therefore the defect structure at the Cu/Ti interfaces should be much more pronounced. Thus it is reasonable to expect that because of dislocations, etc., the interfacial scattering will be more intense, and have a larger effect on the resistivity.

From a more fundamental point of view, we note the extreme similarity of our data with the phenomenology in disordered metals. We shall now try to interpret our results in terms of current ideas.

We start by evaluating the electron mean free path. Due to the relatively large resistivity ratio between Ti and Cu (≈60 at room temperature), our superlattices may be assumed to be parallel resistor networks in which the titanium layers may be neglected; thus we have

$$\rho_{Cu} \approx \rho/2 , \tag{5}$$

where ρ_{Cu} is the resistivity of the Cu layer in the Cu/Ti superlattice, and ρ is the measured resistivity of the sample. In the Drude approximation, we have

$$\bar{\ell} = \frac{A}{\rho} , \tag{6}$$

where the constant $A = mv_F/ne^2$ has the value of 850 $\mu\Omega$ cm Å for a Cu layer.[6] The ℓ^1_{Cu} values obtained this way are shown in Table I.

We now compare these results with the prediction of a simple theoretical model[15] in which the effect of interfaces on the resistivity is included, but under the assumption that the relaxation is the same as for the bulk. In this case Eq. (6) is still expected to hold. Using the value of 500 Å for the bulk Cu electron mean free path, we obtain the ℓ^2_{Cu} values also reported in Table I.

There are two noticeable features in these results; the first is that our experimental mean free path is larger than the layer thickness, and this differs from the behavior found for Al/Nb and Cu/Nb,[6] in which just the opposite happens. This difference is probably due to better sample quality in our case, and has the interesting implication of making, at least in principle, the samples with the thinnest layers two-dimensional electron systems. The second and related feature is the increasing difference between ℓ^1_{Cu} and ℓ^2_{Cu} as the superlattice period decreases. We feel that this behavior is direct proof of differences in the relaxation processes in interface scattering relative to those in the bulk. Such differences will play a bigger role as the layer thickness decreases. Thus we expect relaxation in the λ=50 Å and especially 20 Å samples to be essentially dominated by interface scattering.

TABLE I. Values of electronic mean free path obtained in two different ways from the experimental data (see text). Resistivity values are for room temperature.

λ (Å)	ρ ($\mu\Omega$ cm)	ℓ_1 (Å)	ℓ_2 (Å)
400	3.5	242	240
200	6.3	135	150
100	11.6	73	102
50	26	33	68
20	50.6	17	33

The resistivity of the 20 Å sample is ≈ 110$\mu\Omega$ cm, which is close to the critical Mooij value of 150$\mu\Omega$ cm separating metallic from "nonmetallic" behavior. The negative TCR may thus be explained in terms of defect-induced quantum-interference effects.[12] According to this general model of transport in strongly disordered metals, the temperature-dependent part of the resistivity may be written in terms of the usual expression involving elastic scattering plus another inelastic term, due to defect-induced scattering. This latter term, which may lead to a decrease of the resistivity as temperature increases, may dominate the actual temperature dependence over a wide temperature range. In fact, the temperature dependence of the quantum-interference effect may be parametrized by the so-called inelastic scattering time τ_i, which is expected to scale with temperature as T^{-n}, with n=2 if phonons provide the dominant inelasticity. In terms of the conductivity σ, for a three dimensional disordered metal we have

$$\Delta\sigma = \frac{\Delta\rho}{\rho^2} \approx \frac{e^2}{2\pi^2\hbar}(D\tau_i)^{-1/2} \approx T \tag{7}$$

if $\tau_i = aT^{-2}$.

In Fig. 4 we replot the behavior of $\Delta\sigma$ on an expanded scale for the 20 Å sample to compare with Eq. (7). For $T \geq 30$ K we clearly observe the linear dependence predicted by Eq. (7). From a fit, we may obtain τ_i, provided that the diffusion coefficient D is known. Using $D = \approx 0.2$ cm^{2}s^{-1} obtained for Cu/Nb superlattices[3] and $Cu_{0.5}Ti_{0.5}$ alloys,[16] we obtain $\tau_i = aT^{-2} = 3.6 \times 10^{-9}T^{-2}$. Thus τ_i varies between

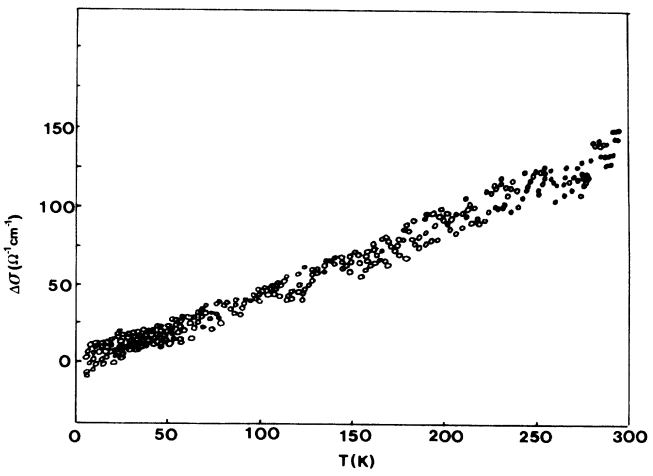

FIG. 4. Precise temperature dependence of $\Delta\sigma$ for the λ=20 Å sample.

$\approx 10^{-12}$ s at 20 K to $\approx 10^{-14}$ s at 200 K, in good agreement with results on the Cu/Nb superlattices and Cu-Ti alloys.

For $T \leq 30$ K we note a slight deviation from linearity. Replotting the data on a logarithm T scale, we observe linear behavior in this low-temperature range. This would imply a crossover from three-dimensional to two-dimensional behavior in the 20 Å sample. Actually, from the measured values of the mean free path we should expect two dimensionality over a wider temperature range. The three dimensionality over most of the temperature range may imply phonon-assisted tunneling of the electrons from one Cu layer to the next. At low temperatures such tunneling is hindered and the layers tend to behave two dimensionally, although we cannot draw quantitative conclusions, given the very weak temperature dependence observed.

In conclusion, we have confirmed weak-localization effects in ultrathin Cu/Ti superlattices. The observed phenomenology of resistivity versus temperature and lattice period is very similar to that of amorphous metals and ultrathin single layers, including the Mooij correlation. This may be intrinsically connected to interface roughness, which not only dominates the behavior of the conductivity within a single layer, but may assist the tunneling by bringing two adjacent interfaces randomly closer together.

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