RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 30, NUMBER 1 1 JULY 1984

# Interplay of superconductivity, magnetism, and localization in Mo/Ni superlattices

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(Received 13 April 1984)

A new experimental relationship between superconductivity, magnetism, and localization is explored in short-wavelength ($14 \text{ Å} \leq \Lambda \leq 40 \text{ Å}$) sputtered Mo/Ni superlattices. A crossover to a superconducting state is observed for $\Lambda < 9$ Å implying paramagnetic behavior when the Ni strata are four atomic layers thick, or less. All samples show localization effects at liquid-helium temperatures and nonsuperconducting samples develop an unusual resistance plateau below $T \approx 0.5$ K.

Artificial modulated heterostructures provide many interesting examples of physical phenomena that are difficult to realize in any other way. The major emphasis so far has been technological, directed at semiconducting superlattices based primarily on GaAs-GaAlAs.[1] However, the broader class of novel heterostructures, particularly *metallic superlattices,* is also of great interest for fundamental studies of collective effects in the restricted geometry of thin layers and interfaces.

In this Rapid Communication we present the results of a combined study of the structural and electronic properties of Mo/Ni superlattices grown by the sequential sputtering technique.[2] We focus on the region of short modulation wavelength ($14 \text{ Å} \lesssim \Lambda \lesssim 40 \text{ Å}$) where strong departures from normal bulk properties may be expected. In our experiments these are most clearly manifest in the competition between magnetic (Ni) and superconducting (Mo) behavior, and in the occurrence of localization effects at low temperatures. Although the samples show a surprisingly high degree of stacking coherence *normal to the layers* there is an intrinsic interfacial mismatch in these mixed bcc-fcc microstructures which is crucial to understanding their electrical properties, as we shall see.

The Mo/Ni superlattice samples prepared for our experiments were basically of two different types: in most of the samples studied, the flux of sputtered atoms was adjusted to give equal numbers of Mo and Ni layers; in the other kind, the number of Mo layers was chosen to be three times that of Ni layers. We refer to the two types of samples by, respectively, $\frac{1}{2}\Lambda$ and Mo$\frac{3}{4}\Lambda$/Ni$\frac{1}{4}\Lambda$, where $\Lambda$ is the modulation wavelength. The purpose of the latter type of sample was to promote coherent stacking while achieving very thin Ni layers. The superlattices were deposited on 90° sapphire substrates ($\sim$1-cm$^2$ area) held at a temperature of 20°C.[3] The total thickness of the superlattices was approximately 1 $\mu$m. Standard photolithographic techniques were used to etch out a four-point bridge pattern suitable for in-plane dc resistance measurements. In order to facilitate low-temperature measurements the substrates were mounted on, and in some cases immersed directly in, the mixing chamber of a dilution refrigerator. Electrical contacts were made by ultrasonic soldering with pure indium, and voltage measurements were sensed with a precision of 1 part in $10^5$ with excitation currents in the range 10–100 $\mu$A.

X-ray characterizations of the superlattices were carried out at room temperature on a four-circle diffractometer. The x-ray source was a 12-kW rotating anode tube (Mo $K\alpha$) monochromatized with a graphite crystal. Both $\theta$-$2\theta$ and $\omega$ (rocking) profiles were obtained. The former scans determine the degree of ordering perpendicular to the layers and the latter are used to probe undulations of the layers. In all cases there is polycrystalline texture within the layers.[3]

Figures 1 and 2 show the behavior of the in-plane resistance of various 1:1 and 3:1 Mo/Ni samples in the small-wavelength region. Several unexpected findings are evident: firstly, we note that structures with Ni layer thickness $\leq 9$ Å, i.e., less than nominally four atomic layers, show a sharp superconducting transition (see inset of Fig. 1). Referring now to Fig. 2, which shows the corresponding resistance behavior for several longer wavelengths, *no* transition is observed down to 15 mK on structures with Ni layers thicker than 9 Å. This observation of a crossover to superconducting behavior is interpreted as an unequivocal signature of the loss of ferromagnetic order. Indeed, direct measurements of the magnetization $M$ as a function of $\Lambda$, show that $M$ approaches zero at a nickel thickness of 9 Å.[4] It is tempting to ascribe such behavior to so-called "magnetically dead" layers of Ni; however, first one must inquire into the nature of the *interface* between Mo and Ni layers and consider the possibility that alloys are formed which may be nonmagnetic. Thus, we have carried out detailed x-ray structural characterization of the quality of the layering and its degree of coherence. This question becomes particularly relevant here since we are dealing with the interface of two distinct morphologies: bcc Mo(110) and fcc Ni(111).

In Fig. 3 we compare x-ray scattering profiles of three different samples which span the crossover to superconducting behavior. The two scans shown in the main body of Fig. 3, $\frac{1}{2}\Lambda = 11.7$ Å and Mo 15 Å/Ni 5 Å, are representative of the x-ray profiles of all samples in the range $8 \text{ Å} \lesssim \frac{1}{2}\Lambda \lesssim 150$ Å.[3] They consist of a sharp principal peak flanked by satellites at $\pm 2\pi n/\Lambda$, from which the modulation wavelength can be accurately determined. This is precisely what one would expect for scattering from a modulat-

 

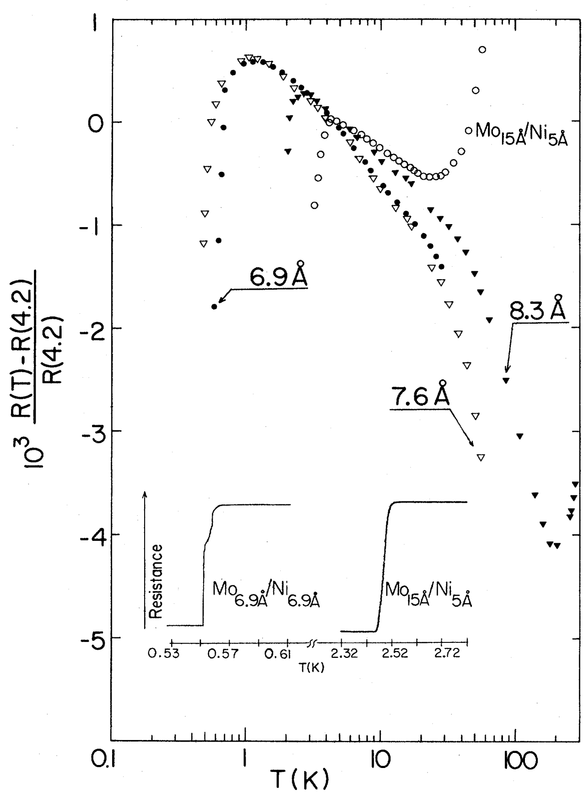


FIG. 1. Temperature dependence of in-plane resistance, normalized to values at 4.2 K. Data are shown for the short-wavelength Mo/Ni heterostructures which show superconductivity. The labels refer to $\frac{1}{2}\Lambda$ for samples with equal numbers of Ni and Mo layers. The inset shows the sharpness of the superconducting transition.

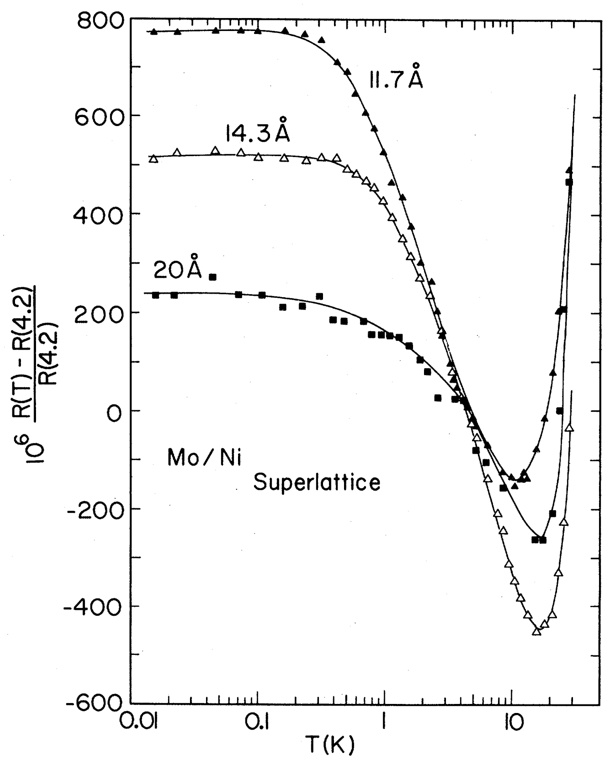


FIG. 2. Temperature dependence of normalized resistance for nonsuperconducting Mo/Ni structures. Again, the labels refer to $\frac{1}{2}\Lambda$.

ed structure that is substantially *coherent* perpendicular to the layers. The $2\theta$ position of the principal peak gives an average $d$ spacing in this direction of 2.14 Å consistent with Mo(110)/Ni(111) stacking. The inverse width of the peaks, after deconvoluting the instrumental resolution, can be used to determine the coherence length of the ordering perpendicular to the layers. In this way, the two samples referred to in Fig. 3, $\frac{1}{2}\Lambda = 11.7$ Å (nonsuperconducting) and Mo 15 Å/Ni 5 Å (superconducting), are found to have coherent stacking over lengths of at least 250 and 100 Å, respectively. Detailed modeling[3] shows that intermixing, if any, occurs at most on one interfacial atomic plane, confirming that for both of these samples the interfacial region has *well defined layering* with few faults, i.e., at most one every 5Λ. Small angle scattering results[3] have also confirmed independently the thickness of the layers and that there is uniform 100% composition modulation. The layering is further characterized by measurements of the vertical mosaic [∼8°, half-width at half maximum (HWHM)] which probe angular undulations of the layers across the sample [see Fig. 3, inset (a)]. Also, we can rule out interdiffusion at room temperature since the satellite intensities do not change over a period of many months.

Now, if *both* Mo and Ni constituent layers are made very thin, an interesting structural effect is observed. Referring to Fig. 3, inset (b), we see that the $\theta$-$2\theta$ diffraction profile for $\frac{1}{2}\Lambda = 6.9$ Å takes on a broad continuum form reminiscent of an amorphous structure. The short-range order is calculated to extend over only ∼20 Å for this sample. Thus, in the small wavelength limit $\frac{1}{2}\Lambda \lesssim 8$ Å, coherence is lost and the samples are more akin to metallic glasses than superlattices. One factor which may be responsible for the loss of coherence is the intrinsic limitation of the sputtering

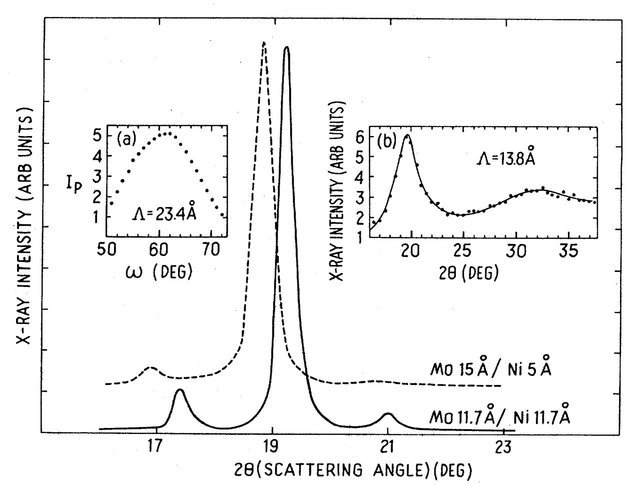


FIG. 3. 00$l$ x-ray diffraction profiles for two samples showing coherent satellites. Inset (a): Rocking curve for Mo 11.7 Å/Ni 11.7 Å. $I_p$ is the intensity of the principal peak near $2\theta = 19°$. Inset (b): Glassy behavior of Mo 6.9 Å/Ni 6.9 Å sample.

method. Another possibility is that these superlattices cannot sustain more than a critical amount of strain. If this limit is exceeded then it may become more favorable to form a random alloy, as in the $\frac{1}{2}\Lambda = 6.9$ Å sample shown in Fig. 3. This has been confirmed by recent molecular dynamics calculations.[5]

To summarize the structural data then, we observe quite coherent layering until the thickness of *both* constituents is reduced below $\sim 8$ Å, in which case a glassy structure is obtained. This structural crossover may be responsible for the grouping of $T_c$'s seen in Fig. 1, i.e., $T_c \approx 0.5$ K for the glassy structures and $T_c \approx 2$ K for those with coherent layering and thin Ni layers. As the thickness of the Ni component is reduced we find that the onset of superconductivity occurs somewhat *before* the coherence of the layering is lost. This result is suggestive of the existence of magnetically "dead," i.e., paramagnetic, layers when the Ni strata are less than four atoms thick. However, we cannot rule out the possibility that a small amount of intermixing could be responsible for this behavior.[6] The existence of magnetically "dead," i.e., paramagnetic, layers has been discussed extensively in the literature. The current consensus[7–10] is that *substrate effects* are very important. For example, a Ni monolayer is calculated to be ferromagnetic on Cu(100) but paramagnetic on Cu(111) and Tersoff and Falicov[9] concluded that for substrates which couple strongly to the Ni film, ferromagnetism is suppressed at around three atomic layers of Ni (by *sp-d* hybridization); our findings lend support to this conclusion.

Finally, we point out an interesting effect observed on the resistance curves in Figs. 1 and 2. In all cases, including the superconducting samples, the resistance shows a distinct upturn at low temperatures. The position of the resistance minimum deepens and shifts to a higher temperature with decreasing $\Lambda$; in fact, samples with the smallest $\Lambda$ (13.8 and 15.2 Å) have negative temperature coefficients up to at least room temperature. Absolute resistivities are in the range 60–160 $\mu\Omega$ cm, the actual value being inversely proportional to the modulation wavelength. This fact, coupled with the imperfect vertical mosaic mentioned above, demonstrates the dominance of boundary (interface) scattering in the low-temperature electronic transport. Of particular interest are the nonsuperconducting (ferromagnetic) samples, shown in Fig. 2, in which the resistivity levels off[11] in a plateau in the region below $T = 0.5$ K. Truncation of the resistance rise at low temperatures may signal the importance of *finite size effects,* such as the effective width of the conduction channel becoming comparable to the inelastic diffusion length. The latter phenomenon was recently searched for in thin, short films of $Au_{40}Pd_{60}$ but so far has not been observed.[12] Alternatively, at low enough temperatures the plateau may arise from the destructive influence of strong internal magnetic fields on the spin pairing.[13] It is interesting to note that superconductivity and the existence of the resistivity plateau seem to be mutually exclusive. The reason for this is not currently understood but such effects have been the subject of considerable recent theoretical interest.[14] We are now carrying out more detailed experiments to distinguish between the different possible mechanisms and this work will be the subject of a longer publication. In connection with the resistivity plateau, it is also interesting to note that an almost identical effect has been reported previously in amorphous ferromagnets[15] but, again, the actual mechanism is unclear.

We acknowledge stimulating discussions with Dr. H. Fukuyama and Dr. Y. Bruynseraede, and thank H. Homma and D. T. Morelli for help with x-ray characterizations and measurements. The work was supported in part by National Science Foundation Low Temperature Grant No. DMR 82-07449 (R.C.) and Grant No. DMR 83-04356 (C.U.), Army Research Office Grant No. DAAG-29-83-K-0131, U.S. Office of Naval Research Contract N00014-83-F-0031, and the U.S. Department of Energy.

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