Oscillatory Behavior of the Transport Properties in Ni/Co Multilayers: A Superlattice Effect

J. M. Gallego, D. Lederman, S. Kim, and Ivan K. Schuller
Physics Department 0319, University of California - San Diego, La Jolla, California, 92093-0319
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The resistivity and anisotropic magnetoresistance of Ni/Co multilayers exhibit an oscillatory dependence on Co or Ni layer thickness, with an approximate period of 20 Å. The oscillations disappear as the number of bilayers is reduced, clearly proving that this is a superlattice effect. This is the first time that superlattice effects are observed in the transport properties of metallic multilayers.

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The transport properties of magnetic multilayers have recently attracted much attention, especially since the discovery of the giant magnetoresistance effect in Fe/Cr multilayers [1]. In these and other magnetic/nonmagnetic multilayers, the coupling between magnetic layers oscillates between ferromagnetic and antiferromagnetic as a function of the nonmagnetic layer thickness [2]. This oscillatory coupling produces oscillations in the magnitude of the magnetoresistance, which is a maximum (minimum) for antiferromagnetically (ferromagnetically) coupled magnetic layers. It was recently shown that the coupling [3] and the magnetoresistance [4] also oscillate as functions of the magnetic layer thickness in Fe/Cr and Co/Cu multilayers.

On the other hand, the same mechanism thought to be responsible for the giant magnetoresistance effect, i.e., the spin dependent scattering at the interfaces, is detrimental for the observation of superlattice effects, as interfacial scattering may destroy long range coherence in the electronic wave functions. In fact, in most metallic multilayers grown to date the conduction electrons' mean free path is limited by the layer thicknesses [5,6], which implies the absence of extended electronic wave functions throughout the superlattice. As a consequence, the resistivity increases smoothly with decreasing layer thickness, saturating for very thin layers [7–11]. A simple model of parallel decoupled double layers [10] explains this behavior.

In this Letter, we report on the dependence of magnetotransport properties of Ni/Co multilayers on the Ni and Co thicknesses. Their resistivity and anisotropic magnetoresistance (AMR) are found to oscillate as functions of the Co and Ni thicknesses. These oscillations are due to a genuine superlattice effect because they disappear as the number of bilayers is reduced. To the best of our knowledge, this is the first observation of superlattice effects in the transport properties of metallic multilayers.

The $(Ni_aCo_b)_N$ multilayers (with a and b the Ni and Co layer thicknesses in Å, respectively, and N the total number of bilayers) were grown by molecular beam epitaxy (MBE) on $Al_2O_3(11\bar{2}0)$ substrates. Co and Ni were deposited using two independent electron guns with computer controlled pneumatic shutters. The evaporation rates were ~0.1 Å/s for Ni and ~0.05 Å/s for Co. The base pressure in the ultrahigh vacuum (UHV) chamber was 2×10^{-10} torr, and did not exceed 5×10^{-9} torr during growth.

A 50–80 Å thick Co layer was grown at 350 °C on $Al_2O_3(11\bar{2}0)$. This buffer layer grows epitaxially, in the fcc structure, with Co(111) parallel to $Al_2O_3(11\bar{2}0)$ [12]. Four different in-plane domains are present, as shown by the low energy electron diffraction (LEED) pattern in Fig. 1(a) for an approximately 80 Å Co layer. The Ni/Co multilayers, approximately 1000 Å thick, were grown at 150 °C on these buffer layers. The LEED pattern after completion of the growth [as shown in Fig. 1(b) for a $(Ni_{42}Co_{18})_{16}$ multilayer] is similar to that of the buffer layer, indicating that the epitaxial relationship and crystallinity hold throughout the whole multilayer.

Figure 2 shows typical x-ray diffraction spectra, for both low and high angle. These spectra indicate that for all the multilayers, Ni and Co grow in the fcc structure, with the (111) direction normal to the surface [13]. The crystalline coherence length, calculated from the width of the main superlattice peaks (≈750 Å), is similar to the total film thickness. Together with the LEED data, this indicates that the multilayers are *quasisingle* crystalline (single crystals in the growth direction but with four different in-plane domains). Because of the low scattering contrast between Ni and Co, a quantitative structural refinement [14] of these spectra is difficult, since the obtained parameters are not unique. Nevertheless, the observation of up to the fourth order satellite peak in

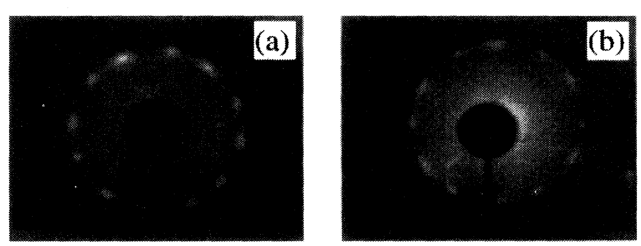

FIG. 1. (a) LEED pattern after depositing 80 Å of Co on $Al_2O_3(11\bar{2}0)$ at 350 °C. The primary beam energy is E_p = 177 eV. (b) LEED pattern after completion of a $(Ni_{42}Co_{18})_{16}$ multilayer. E_p = 169 eV.

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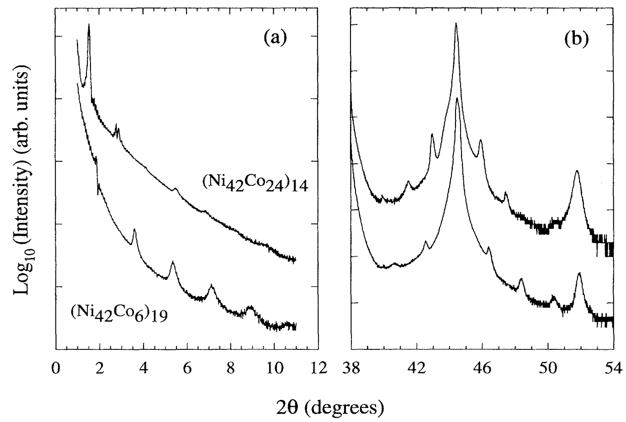

FIG. 2. Low (a) and high (b) angle x-ray diffraction spectra for two Ni/Co multilayers. The peaks at $2\theta \sim 44.5°$ are the central Bragg peaks of the superlattices, corresponding to a weighted average of Ni(111) and Co(111). The tail at the left side of the high angle spectrum is due to the $(11\bar{2}0)$ reflection of the sapphire substrate, and the peak at $\sim 51.6°$ to a small quantity (<1%) of (100) oriented grains. The other peaks shown are satellite peaks due to the superlattice periodicity.

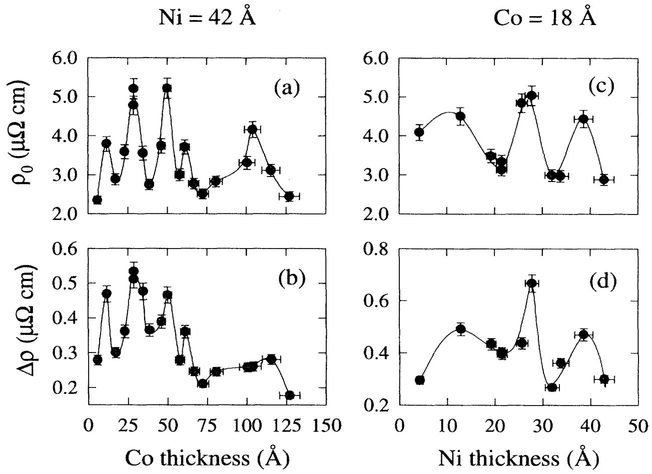

FIG. 3. (a) Average resistivity and (b) anisotropic magnetoresistivity for a series of Ni/Co multilayers with $b = 42$ Å as a function of Ni thickness. (c) Average resistivity and (d) anisotropic magnetoresistivity for a series of Ni/Co multilayers with $a = 18$ Å as a function of Co thickness. The solid lines are a guide to the eye.

some of the multilayers, despite this low contrast, implies an approximately square wave composition modulation. Using Auger spectra taken during growth, we estimate an upper limit of 4–5 Å for the interdiffused region thickness at the interfaces [13]. It is important to point out that there was no correlation between the resistivity oscillations described below and any structural parameter that we could measure.

The four point magnetoresistance was measured at a temperature $T = 4.2$ K in fields up to 30 kOe on photolithographically patterned samples. The magnetoresistance was measured with the magnetic field in the plane of the multilayer and parallel or perpendicular to the current. The anisotropic magnetoresistance $\Delta\rho$ is defined as $\Delta\rho = \rho_{\parallel} - \rho_{\perp}$, where $\rho_{\parallel}$ and $\rho_{\perp}$ are the saturation resistivities with the magnetic field applied parallel and perpendicular to the current, respectively. The average resistivity ρ_0 is defined as $\rho_0 = (\rho_{\parallel} + \rho_{\perp})/2$.

The magnetotransport properties of Ni/Co multilayers [15] are qualitatively similar to those of most ferromagnetic materials [16]; i.e., the parallel magnetoresistance is positive, while the perpendicular magnetoresistance is negative. In contrast, in multilayers that exhibit giant magnetoresistance, both quantities are negative and of the same order of magnitude. In addition, Ni/Co multilayers show large magnetoresistances with low saturation fields, which makes them possible candidates for use as magnetoresistive recording sensors [15].

Figures 3(a) and 3(b) show the resistivity and the anisotropic magnetoresistance as a function of the Co thickness for a series of multilayers with fixed Ni thickness $a = 42$ Å. Both quantities display large oscillations, well outside the measurement uncertainty, with a ~20 Å period. Moreover, for two independently grown samples, prepared in separate experimental runs, ρ_0 was reproduced within 10%, and $\Delta\rho$ within 3%. Since the data were obtained in random order, the systematic variations in ρ_0 and $\Delta\rho$ imply that this is a real physical effect, and not just random scatter due to uncontrolled variations of structural parameters. It is important to emphasize that the structure of the multilayers was thoroughly characterized, as mentioned above, with *in situ* electron diffraction and Auger spectroscopy and *ex situ* x-ray diffraction [13], and no correlation was found between any structural parameter and the variations in the resistivity. Figures 3(c) and 3(d) show a similar behavior for a series with fixed Co thickness $b = 18$ Å. Note that although Fig. 3 shows ρ_0 as a function of layer thickness, oscillations are also observed in $\rho_{\parallel}$ or $\rho_{\perp}$ because the magnitude of the oscillations (~200%) is much larger than the AMR (~10%).

For very thin layers (i.e., a fixed Co thickness $b =$ 6 Å), we only found one strong peak in ρ_0 for $a \sim$ 10 Å and a weak shoulder at $a \sim 33$ Å. In this case, the multilayers behave more like an ordinary NiCo alloy with respect to the transport properties. This agrees with expectations based on the structural studies mentioned above, which indicate an interdiffused region smaller than 5 Å. It is known [17–20] that Ni_xCo_{1-x} alloys show a maximum in $\Delta\rho$ when $x \sim 0.7$, which is approximately equal to the Ni concentration in a Ni_9Co_6 multilayer (~0.6). When the Ni thickness increases, layers of pure Ni begin to grow between the interdiffused regions, thus decreasing the resistivity and the anisotropic magnetoresistance.

The low resistivity of the samples is further evidence of their high crystallinity and low interface scattering.

Assuming that the conduction electrons are free electron-like, their mean free path, calculated from the Sommerfeld approximation, ranges between 100 and 200 Å [21]. Thus, in almost every sample displayed in Fig. 3(c), the mean free path is larger than the modulation wavelength of the multilayer, indicating that the interface scattering is very small. In fact, the mean free path seems to be limited by the in-plane grain size, which ranges between 100 and 200 Å, as obtained from the width of the in-plane (220) peak using grazing angle x-ray diffraction [13]. Since these samples show long range structural coherence, it should be possible to observe superlattice effects in their electronic properties. This would explain why, unlike most metallic multilayers reported until now, the resistivity of our samples does not monotonically increase with decreasing layer thicknesses.

One possible explanation of the ρ_0 oscillations relies on the zone folding of the electronic energy bands along the (111) direction due to the superlattice periodicity. The resulting superlattice bands will have energy gaps at the superlattice Brillouin zone (BZ) boundary [22,23]. If the Fermi level of the sample lies within a superlattice energy gap, the resistivity along the (111) direction will increase. In Ni the Fermi level crosses the $\Lambda_{3\downarrow}$ band at some point of the BZ along the (111) direction [24]. If the band structure of the superlattice is similar to that of its constituents, resistivity changes would be observed when $\Lambda = a + b$ causes a superlattice energy gap to appear at the Fermi level. However, because of differences in the Ni and Co band structures, the position of the resistivity maxima and minima will also depend on the relative concentrations of the two materials. In the case of the Ni 42 Å and Co 18 Å series, the periodicities are ~20 and ~12–16 Å, respectively. The slight periodicity difference may be a result of the different relative Co to Ni concentration ratios. However, the opening of minigaps is along the growth direction, whereas we measure the resistivity along the layers. This may not pose a problem if strong spin-orbit effects are present [25].

Another possibility is the creation of localized states due to interface disorder which slightly breaks the superlattice symmetry, turning the superlattice structure into a quasiperiodic system. A similar effect has been studied theoretically in crystals with incommesurate periodic potentials [26]. The energies of these localized states are functions of the number of Ni and Co atoms in a superlattice period. If these states lie near the Fermi level, the transport properties could change significantly.

In both models described above, the ρ_0 oscillations result from a superlattice effect. In order to test whether this is indeed the case, we grew other $a = 42$ Å series of samples with different number of bilayers N. Figure 4 shows ρ_0 for $15 < b < 60$ Å samples with $N = 2$ together with the data plotted in Fig. 3 ($N > 10$). Clearly the oscillatory behavior disappears as the number of bilayers is reduced to $N = 2$. Similar data for $N = 7$ showed

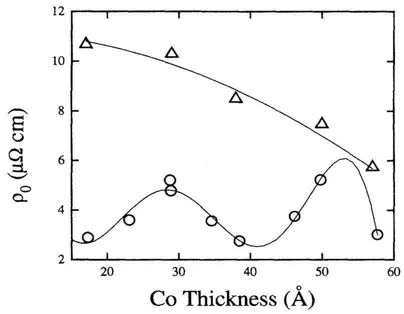

FIG. 4. Average resistivity as a function of Co thickness in Ni/Co multilayers with $b = 42$ Å with different number of bilayers. △: $N = 2$; ○: data from Fig. 3 ($N > 10$). The lines are polynomial fits to the data.

oscillations. This proves that the phenomenon is a superlattice effect, and not due to a single layer mechanism such as quantum confinement. The increase in ρ_0 with decreasing N is probably due to a relative increase in surface scattering due to the reduced *total* film thickness.

Finally, the possibility that these oscillations are magnetic in origin is currently being investigated.

A cursory inspection of Fig. 3 shows that, to a first approximation, the anisotropic magnetoresistance and the resistivity are related. A plot of $\Delta\rho$ vs ρ_0 (Fig. 5) reveals that both quantities are roughly proportional. A linear least squares fit to the data points gives $\Delta\rho/\rho_0 \approx 0.10$. Within the two subband model originally developed for magnetic alloys [27], the low-temperature anisotropic magnetoresistance can be written as

$$\frac{\Delta\rho}{\rho_0} = \gamma\left(\frac{\rho_{0\downarrow}}{\rho_{0\uparrow}} - 1\right), \qquad (1)$$

where $\gamma \approx 0.01$ is a constant dependent on the amount of spin-orbit coupling and $\rho_{0\downarrow}$ ($\rho_\uparrow$) is the resistivity of the subband with spin antiparallel (parallel) to the magnetization direction. Note also that the least squares fit straight line extrapolates through the origin within experimental error, in good agreement with expectations based on Eq. (1). Thus, within this model, $\alpha = \rho_\downarrow/\rho_\uparrow \sim 10$, and the dependence of $\Delta\rho$ on the Ni or Co thicknesses

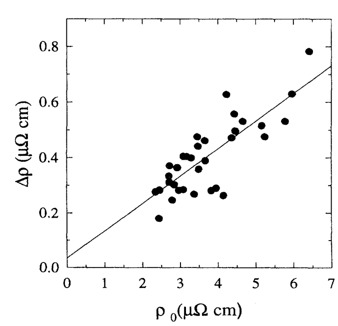

FIG. 5. Relationship between the anisotropic magnetoresistivity $\Delta\rho$ and the average resistivity ρ_0 for all Ni/Co multilayers. The straight line is a linear fit to the data.

is just a consequence of the dependence of ρ_0 (for Ni/Co alloys, values for α ranging from 13 to 29 have been reported [27]).

Previously, superlattice effects in metallic multilayers have only been observed in structural (the appearance of superlattice peaks in x-ray diffraction) [28] and magnetic (the collective behavior of magnons in magnetic/nonmagnetic superlattices) properties [29]. In the first case, only structural coherence in the perpendicular direction is required. In the second case, the superlattice effect is associated with the dipolar coupling between the two magnetic layers, and so structural coherence at the atomic level is not required, only long-range magnetic order at the hundreds of angstroms length scale. The *transport* superlattice properties, however, require that the electron mean free path must be longer than a few bilayers so that the electronic wave functions extend over several superlattice periods [30]. Therefore, not only is structural coherence in the range of several multilayer periods required, but spin scattering at the interfaces, which could limit the extent of the electronic wave function, must also be small. We believe this is the reason electronic superlattice effects were not previously observed in metallic multilayers.

In summary, we have discovered systematic oscillations in the transport properties of Ni/Co multilayers as a function of the Co or Ni thickness. As the number of bilayers is reduced, the effect tends to disappear, which is further proof that the oscillations are due to the superlattice structure. This is the first time that superlattice effects have been observed in the transport properties of metallic multilayers.

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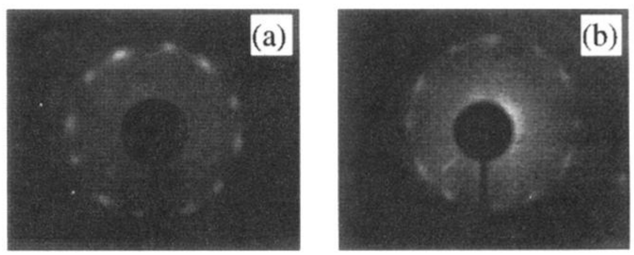

FIG. 1. (a) LEED pattern after depositing 80 Å of Co on $Al_2O_3(11\bar{2}0)$ at 350 °C. The primary beam energy is E_p = 177 eV. (b) LEED pattern after completion of a $(Ni_{42}Co_{18})_{16}$ multilayer. E_p = 169 eV.