Model Hamiltonian for the conductivity oscillations of magnetic multilayers

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The behavior of the electrical conductivity as a function of layer thickness of the superlattice systems Ni/Co, Ni/Cu, and Pd/Ag is studied. Experimentally an oscillatory dependence was found for the first two, while the latter exhibited a monotonous behavior. In our calculations we find that, in these superlattices, the current is carried by the sp-character electrons, which are quite insensitive to the interfaces. To interpret the experimentally observed resistivity oscillations we suggest a scattering mechanism of these carriers against d-character quantum well states that are present in only one of the superlattice materials, when the well state energy is close to E_F. [S0163-1829(97)02521-6]

Because of their intriguing properties magnetic multilayers have lately been in the focus of attention of both experimentalists and theoreticians. Among these properties the oscillatory exchange coupling between magnetic layers separated by nonmagnetic spacers,[1,2] and the giant magnetoresistance,[3] deserve special mention. Also, recent measurements of the resistivity and anisotropic magnetoresistance of Ni/Co multilayer systems[4,5] show a clear-cut oscillatory dependence on layer thickness.

On the other hand, the possibility of achieving the ballistic limit, in which the bound on the electrical conductance is not due to lattice imperfections, but rather to the presence of a constriction, has been intensively investigated over the years[6,7] since Landauer first suggested it[8] in 1957. Thus, in this context it is of special interest that the experiments of Ref. 4, which report the oscillatory dependence of the conductivity on layer thickness, were performed very near (if not at) the ballistic limit. Moreover, since the amplitude of the oscillations decreases when the number of repeated layers is reduced, there is good reason to attribute them to the superlattice structure of the systems. To test this hypothesis we previously performed[9] band structure calculations, and based on them we here put forward a theoretical model that accounts for the transport properties of these superlattices.

The conductivity of metallic systems depends only on the quantum states with eigenenergies close to the Fermi level E_F, and for superlattices these states shift as a function of the number of layers of each element. To study the dependence of the conductivity on the thickness of each superlattice material, we have considered fcc superlattices grown in the (111) direction, in particular Ni/Co, Ni/Cu, and Pd/Ag. In each of these three cases, only one of the pure elements of the pair has a d band that crosses the Fermi level in the Γ-L direction. It is therefore pertinent to pay special attention to the Γ-L direction of the Brillouin zone of these superlattices, where we expect folding effects to be important.

The electrical conductivity is usually calculated within the semiclassical approximation (Boltzmann equation).[10] It contains a contribution that is due exclusively to the band structure, which can be estimated assuming that the relaxation time τ is a constant, independent of wave number, band index, and number of layers of each element. This implies an average over the several scattering mechanisms that are present, such as defects, impurities, disorder, and interfaces. Several calculations of this type, which focus on the band contribution alone and assume both spin currents to be independent (two channel model), have recently been performed in the quest to understand the giant magnetoresistance phenomenon.[11–16] In this same context we have recently carried out[9] detailed calculations for Ni/Co superlattices of a variable number of layers, finding that the interfaces only reduce the minority spin conduction, which is mostly due to d electrons, when compared to the pure metals. However, as most of the current is carried by the majority spin bands, which are of sp character, the total resistivity of the system is barely affected by the interfaces. Moreover no conductivity oscillations of appreciable size, as a function of layer width, were obtained within these approximations.

But, an interesting property that does depend on the thickness of the Ni and Co layers was noticed as an outcome of the work of Ref. 9: quantum well states (QWS's) present in the Ni layers, of d character and minority spin, appear in the Brillouin zone along the Γ-L line and its vicinity. In addition, for some particular layer thicknesses they have energies very close to E_F. A similar result was recently reported for Cu/Co superlattices grown in the (100) direction.[17] We believe that the presence of these nondispersive states near E_F bears relation to the experimentally observed conductiv-

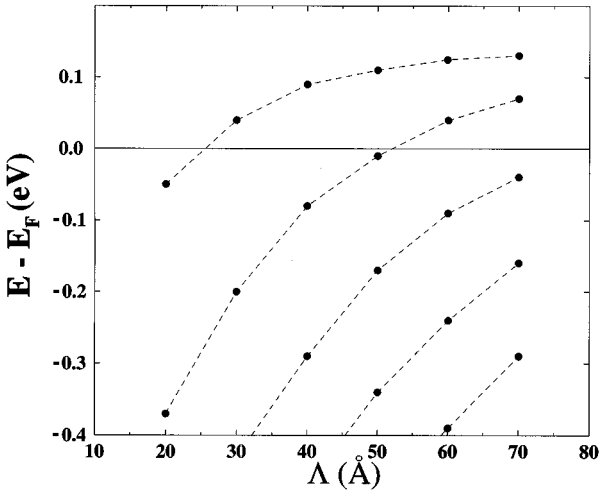

FIG. 1. Quantum well levels at Γ for the superlattices Co(0.6Λ)/Ni(0.4Λ) as a function of Λ. The dashed lines link QWS's with the same number of nodes.

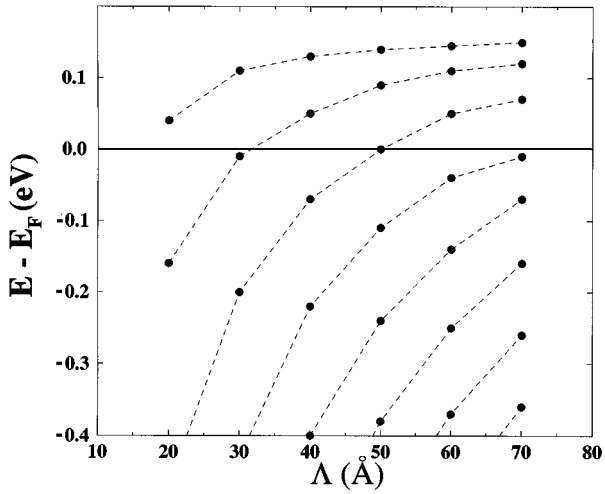

FIG. 2. Quantum well levels at Γ for the superlattices Ni(0.6Λ)/Cu(0.4Λ) as a function of Λ. The dashed lines link QWS's with the same number of nodes.

ity oscillations. Arguments of this type have been used[18] to explain the influence on the conductivity of different dilute transition metal impurities in Cu: when the impurity *d* level lies close to the Cu Fermi level, scattering of the conduction electrons is enhanced and hence an increase of the resistivity is to be expected.

In Fig. 5 of Ref. 9 we showed the calculated energy levels at the Γ point, for superlattices having nine Co layers (18 Å) and a variable number of Ni layers. This geometry was chosen to compare with the experiments of Ref. 4. We found a periodic approach towards E_F of the energy levels, with a periodicity of nearly four Ni layers, which is quite similar to that of the experimentally observed resistivity maxima. The figure resembles those due to the discretization of the perpendicular wave vector in a thin film, as for example shown in Refs. 19 and 20, and in fact this effect occurs both in slabs and superlattices.

Actually, to obtain the energy of these QWS's as a function of the number of layers of each material, it is not necessary to perform a fully self-consistent calculation, such as in Ref. 9. In fact they can be obtained, without a significant loss in precision, using a simple tight-binding method with the parameters of both bulk materials, but shifting the diagonal elements so as to align the two Fermi levels. The results shown in Figs. 1 and 2, for Ni/Co and Ni/Cu, respectively, were calculated within this approximation. In both cases there are QWS's of minority spin and *d* character, in the Ni layers, whose energies approach periodically the Fermi energy. Inspection of the wave vectors shows that the state of highest energy is nodeless, the second has one node, and so on. Experiments[5] show resistivity oscillations in these two cases, with approximately the same periodicity found through our calculations.

In the case of Pd and Ag there is a large interfacial misfit, since their lattice constants differ by 5%. Moreover, Pd becomes magnetic when strained to adopt the lattice constant of Ag. However, when we performed a full self-consistent calculation for superlattices with only a few layers of Pd, they turned out to be nonmagnetic. Consequently, we assumed that the magnetic splitting remains small for superlattices with additional Pd layers, and the Pd/Ag results we report here are nonmagnetic. Figure 3 shows the QWS for Pd/Ag which are of *d* character and located in the Pd layers. The main difference with the Ni/Co and Ni/Cu cases is that they all lie below E_F and approach monotonously this energy level as the thickness of Pd increases. Some states of *sp* character also appear close to E_F in this case, but they are not QWS's. The experiments reported in Ref. 5 show an increasing resistivity with Pd thickness, that saturates at about 25 Å (or 12 monolayers), with no oscillations.

Consequently, in the three pairs of elements we have examined in detail, Ni/Co, Ni/Cu, and Pd/Ag, there definitely is a correlation between the existence of QWS's close to E_F and the maxima of the measured resistivity.

How can we reconcile our previous nonoscillating conductivity results for the Ni/Co superlattices,[9] with the oscillations observed experimentally and the periodic appearance of QWS's near the Fermi energy? Evidently, we have to modify some of the hypothesis of the previous calculations

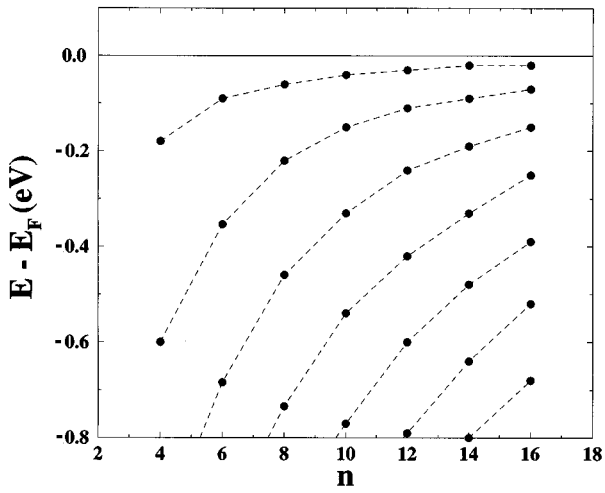

FIG. 3. Quantum well levels at Γ for the superlattices 9Ag/nPd as a function of n. The dashed lines link QWS's with the same number of nodes.

and consider the different scattering mechanisms separately. In particular, we may envision a spin-flip process that scatters an sp-character conduction electron of the majority band into a quantum well d-character minority state. Actually, spin relaxation effects in NiCu, which increase as the Ni concentration grows, have been observed experimentally by Hsu *et al.*[21] Moreover their measurements, in Co/NiCu multilayers, established that spin-flip scattering does occur in NiCu alloys.

As the d states are nondispersive they do not contribute to the conductivity; on the contrary, if their energies are close to E_F they should decrease the total conductivity. In the Boltzmann picture this implies that τ is not constant and that each scattering mechanism contributes separately to the resistivity with a different τ value. Similar mechanisms were already proposed by other authors. Among them we mention Suzuki and Taga,[22] who estimated the influence of d bound states on the minority conduction channel of Co-Cu superlattices, and Campbell and co-workers,[23,24] who studied the influence of spin-flip processes on the resistivity of Fe- and Ni-based alloys.

Following the above arguments we study an additional contribution to the resistivity due to s-d scattering (where the d states are QWS's). In the first two systems under consideration, Ni/Co and Ni/Cu, the scattering process involves a spin-flip, while for Pd/Ag this is not necessarily the case. In addition, we believe that the essence of the two channel model is preserved, since electrons are only rarely scattered by this mechanism, but one has to keep in mind that there is a small s-p-d hybridization of the genuine multilayer bands, and thus s-d scattering only constitutes a simplified notation for a more complex reality. We consider the following model Hamiltonian, which incorporates the interaction between a conduction s electron and a quantum well, mainly d state:

$$H=\sum_{\vec{k}} \varepsilon_{\vec{k}\uparrow}\, c^{\dagger}_{\vec{k}\uparrow}\, c_{\vec{k}\uparrow}+E_d d^{\dagger}_{\downarrow} d_{\downarrow} + \sum_{\vec{k}} \left(V_{k\uparrow d\downarrow}\, c^{\dagger}_{\vec{k}\uparrow}\, d_{\downarrow}+V^{*}_{d\downarrow k\uparrow} d^{\dagger}_{\downarrow} c_{\vec{k}\uparrow}\right), \tag{1}$$

where the first term is the quasifree electron Hamiltonian of the majority spin (mainly s-character) conduction band, the second term corresponds to the quantum well (mainly d-character) minority spin, and the third term is the interaction between the previous two.

In pure bulk magnetic metals the current is transported almost evenly by both spin channels. But, from Table II of our prior paper,[9] and assuming the system to be in the diffusive regime, we conclude that in superlattices the majority band is responsible for about 80% of the total conductivity σ. We also found that σ is only weakly anisotropic, as the effect of the interfaces is rather negligible on the majority carriers. Of course the genuine bands are hybridized and it is the small d character of the majority states that allows for a nonzero spin-orbit coupling with the QWS's, which are pure d minority states. For the time being we have assumed that a nondispersive state, with energy E_d, exists throughout the Brillouin zone. Realistic superlattice band structure calculations show that this is actually the case for a macroscopic fraction of k space. Thus, as already pointed out in Ref. 17, if E_d is close to the Fermi energy then ''a significant region of the superlattice Fermi surface has three-dimensional character.'' This provides a justification for the oversimplified hypothesis we put forward.

The Kubo formula for the conductivity is related to the diagonal element of the retarded Green's function $G^{\rm ret}_{kk}(\omega)$, as follows:[25]

$$\sigma=\frac{4e^2\hbar^3}{3m^2}\int\frac{d^3k k^2}{(2\pi)^3}\int\frac{d\omega}{(2\pi)}[\mathrm{Im}G^{\rm ret}_{kk}(\omega)]^2\delta(\omega-E_F). \tag{2}$$

The Hamiltonian of Eq. (1) can be solved exactly and the relevant Green's function reads

$$G^{\rm ret}_{kk}(\omega)=\frac{1}{\omega-\varepsilon_k+i0^+}\left(1+\frac{|V_{k\uparrow d\downarrow}|^2}{\omega-E_d+i\Gamma}\,\frac{1}{\omega-\varepsilon_k+i0^+}\right), \tag{3}$$

with

$$\Gamma=-\,\mathrm{Im}\sum_{\vec{k}}\frac{|V_{k\uparrow d\downarrow}|^2}{\omega-\varepsilon_k+i0^+}\approx\pi\mathcal{D}_s(E_F)\langle|V_{k\uparrow d\downarrow}|^2\rangle_{\rm FS}, \tag{4}$$

where $\mathcal{D}_s(E_F)$ is the density of states of the majority s band at the Fermi surface. The square of the interaction matrix element $\langle|V_{kd}|^2\rangle_{\rm FS}$, averaged over the Fermi surface, from now on will be denoted as V^2 and assumed to be constant.

However, the width of the unperturbed Green's function [i.e., the first term of Eq. (3)] is nonzero due to impurity scattering. This finite width, which we denote as γ and which simply replaces $0^+\rightarrow\gamma$ in Eq. (3), can be estimated from the experimental conductivity of pure Ni at low temperatures. In fact, the expression for σ can readily be evaluated (ignoring vertex corrections) in the $V=0$ limit, to yield the usual result $\sigma_0=e^2n_0\tau/m$, if $\tau=\hbar/2\gamma$. Here $n_0=p_F^3/3\pi^2\hbar^3$ is the total carrier density, p_F the Fermi momentum, e and m the electron charge and mass.

For $V\neq0$ the integrals in Eq. (2) can also be evaluated analytically, in the physically interesting range of $\gamma\gg\Gamma$, that is, when the s-d scattering is small compared to impurity scattering. Two new contributions to the conductivity σ appear, in addition to σ_0. They are proportional to V^2 and V^4, and can be written in the following way:

$$\frac{\sigma}{\sigma_0}=1-\frac{\Gamma}{\pi\gamma^2\mathcal{D}_s(E_F)}\,\frac{1-y^2}{(1+y^2)^3}\left[4-\frac{1}{\pi\gamma\mathcal{D}_s(E_F)}\,\frac{1-y^2}{1+y^2}\right], \tag{5}$$

where $y=(E_d-E_F)/\gamma$.

Of course, when $y=0$ the physically interesting maximal resistivity case is realized, while for $1>y\gg0$ the additional contributions to σ become negligible. Thus, oscillations of σ as a function of E_d-E_F, or equivalently as a consequence of variations of the multilayer thicknesses, are generated. Moreover, an order of magnitude estimate for the extra $(y=0)$ contribution to σ can be carried out quite straightforwardly. The conductivity of pure Ni, or pure Co, implies a value of $\gamma\sim0.1$ eV. On the other hand, the results of our full band-structure calculations for the superlattices yield $\mathcal{D}_s(E_F)\approx1$ state/(eV unit cell). Consequently, the experimentally observed conductivity reduction can be explained on the basis of a value of the QWS width $\Gamma\sim0.01$ eV, which seems quite reasonable. It is important to realize that in spite

of the fact that the impurity scattering width is much larger than the QWS width ($\gamma \gg \Gamma$), the reduction of σ due to the QWS can be quite significant. In fact, $\Gamma/\gamma = 0.1$ yields a 25% reduction of σ; for $\Gamma/\gamma = 0.2$ a 50% reduction is achieved. We believe this to be the main consequence that can be derived from our model Hamiltonian.

In conclusion, the behavior of the resistivity of these three superlattice systems, as a function of layer thickness, can be traced to the presence of quantum well states with energies close to E_F. Calculations of the conductivity using a constant relaxation time τ independent of wave number, band index, and the number of layers of each element, do not reflect the experimental results. Since in these superlattices the current is mainly carried by the sp-character majority spin electrons, we propose that the origin of the conductivity minima is due to the scattering of these electrons against the QWS. To explore the validity of this mechanism we have put forward a model Hamiltonian which, for reasonable values of the parameters, leads to results in good agreement with experiments.

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