Thin ferromagnetic films on nonmagnetic metallic substrates: A model calculation

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We report a model calculation of the electronic and magnetic properties of a single layer of ferromagnetic material deposited on a nonmagnetic metallic substrate. A tight-binding Hamiltonian with a Hubbard-type interaction term at the surface is used. We obtain the surface Green's function by means of the transfer-matrix method and use the generalized Friedel sum rule to treat charge-transfer effects self-consistently. We find the surface magnetization to be dependent on surface-bulk coupling. We also find that majority states are mostly localized at the surface, while minority states are mainly delocalized. The relevance of our results to experimental data on Ni, Co, and Fe overlayers is discussed.

I. INTRODUCTION

The properties of magnetic thin films have attracted considerable attention in recent years. In particular, several experimental studies[1,2] have been carried out in Fe, Co, and Ni films deposited on nonmagnetic substrates. This, in turn, has stimulated several theoretical investigations of specific systems[3,4] as well as model calculations.[5] Here we report results from a model calculation for a semi-infinite paramagnetic metal on top of which a single atomic layer of an itinerant ferromagnetic metal has been deposited.

The model is characterized by a single-level tight-binding Hamiltonian with hopping between nearest-neighbors only. In addition, a Hubbard-type interaction term is included to describe the magnetic surface. Although the simplicity of our model precludes direct quantitative comparison with experimental results, it does provide a qualitative understanding of the main physical features of the systems under consideration.

This paper is organized as follows: In Sec. II we write down the model Hamiltonian and outline the method of solution. In Sec. III we present numerical results obtained with parameters appropriate to Ni, Co, and Fe on a typical itinerant paramagnet. Finally, Sec. IV contains a brief discussion of the implications of the results and their relation to analogous work.

II. MODEL AND SOLUTION

Our model describes a semi-infinite paramagnetic metal on top of which a single-layer ferromagnetic film has been deposited. We adopt a simplified system in which atoms occupy the sites of a simple cubic structure of lattice constant a. A single electron state per atom and spin direction is assumed and only nearest-neighbor interactions are considered.

The planes parallel to the surface are labeled by the index l, with $l=0$ denoting the ferromagnetic surface layer. An arbitrary lattice point is defined by

$$\vec{R}=\vec{R}_{||}+l\vec{a}\ , \tag{2.1}$$

where $\vec{R}_{||}$ is a lattice vector in the surface plane and $\vec{a}$ is a vector which is orthogonal to the surface and connects adjacent layers.

We express our model Hamiltonian as follows:

$$H=H_0+H_{0,1}+\sum_{l=1}^{\infty}H_l+\sum_{l=1}^{\infty}H_{l,l+1}\ , \tag{2.2}$$

where

$$H_0=\epsilon_0\sum_{\vec{R}_{||},\sigma}a_\sigma^\dagger(\vec{R}_{||},0)a_\sigma(\vec{R}_{||},0)+\frac{t_s}{2}\sum_{\vec{R}_{||},\sigma}\sum_{\vec{\delta}}[a_\sigma^\dagger(\vec{R}_{||},0)a_\sigma(\vec{R}_{||}+\vec{\delta},0)+\text{H.c.}]$$
$$+U\sum_{\vec{R}_{||}}a_\uparrow^\dagger(\vec{R}_{||},0)a_\uparrow(R_{||},0)a_\downarrow^\dagger(\vec{R}_{||},0)a_\downarrow(\vec{R}_{||},0)\ , \tag{2.3a}$$

$$H_{0,1}=\tfrac{1}{2}t_{SB}\sum_{\vec{R}_{||},\sigma}[a_\sigma^\dagger(\vec{R}_{||},0)a_\sigma(\vec{R}_{||},1)+\text{H.c.}]\ , \tag{2.3b}$$

$$H_l=\tfrac{1}{2}t_B\sum_{\vec{R}_{||},\sigma}\sum_{\vec{\delta}}[a_\sigma^\dagger(\vec{R}_{||},l)a_\sigma(\vec{R}_{||}+\vec{\delta},l)+\text{H.c.}]\ , \tag{2.3c}$$

$$H_{l,l+1}=\tfrac{1}{2}t_B\sum_{\vec{R},\sigma}[a_\sigma^\dagger(\vec{R}_{||},l)a_\sigma(\vec{R}_{||},l+1)+\text{H.c.}]\ , \tag{2.3d}$$

where the vector $\vec{\delta}$ connects nearest neighbors within the same layer, and $a_\sigma^\dagger(\vec{R}_{||},l)$ $[a_\sigma(\vec{R}_{||},l)]$ is the creation [destruction] operator for an electron state at site $\vec{R}$ with spin σ.

The parameters of this Hamiltonian are the surface-level position ϵ_0, the hopping matrix elements t_S,t_{SB}, and the Hubbard interaction parameter U. The position of all bulk levels is taken to be the same and chosen, without loss of generality, to be zero. Energies will be measured in units of t_b (i.e., $t_B=1$).

To complete the specification of the model two additional parameters are required: the bulk occupation per atom, n_B, and the nominal surface occupation per atom (i.e., the occupation of the ferromagnetic material in its bulk form), n_S.

In order to take advantage of the invariance of the Hamiltonian under lattice translations parallel to the surface we introduce the mixed Bloch-Wannier representation

$$b_\sigma^\dagger(\vec{k},l)\equiv\frac{1}{\sqrt{N}}\sum_{\vec{R}_{||}}e^{-i\vec{k}\cdot\vec{R}_{||}}a_\sigma^\dagger(\vec{R}_{||},l)\ , \tag{2.4}$$

where $\vec{k}$ is a vector in the two-dimensional Brillouin zone, and N is the number of atoms per layer parallel to the surface. The interaction term in H_0 is treated in the Hartree-Fock approximation. The resulting effective Hamiltonian H_{eff} can then be expressed as follows:

$$H_{\text{eff}}=\sum_{\vec{k}}H_{\vec{k}}\ , \tag{2.5}$$

where Eq. (2.4) has been used and

$$H_{\vec{k}}=\epsilon_{0\uparrow}(\vec{k})b_\uparrow^\dagger(\vec{k},0)b_\uparrow(\vec{k},0)+\epsilon_{0\downarrow}(\vec{k})b_\downarrow^\dagger(\vec{k},0)b_\downarrow(\vec{k},0)+\tfrac{1}{2}t_{SB}\sum_\sigma[b_\sigma^\dagger(\vec{k},0)b_\sigma(\vec{k},1)+\text{H.c.}]$$
$$+\epsilon_B(\vec{k})\sum_l\sum_\sigma b_\sigma^\dagger(\vec{k},l)b_\sigma(\vec{k},l)+\tfrac{1}{2}t_B\sum_l\sum_\sigma[b_\sigma^\dagger(\vec{k},l)b_\sigma(\vec{k},l+1)+\text{H.c.}]\ , \tag{2.6a}$$

$$\epsilon_B(\vec{k})=2t_B(\cos k_x a+\cos k_y a)\ , \tag{2.6b}$$

$$\epsilon_{0\uparrow}(\vec{k})=\epsilon_0-\tfrac{1}{2}Um_0+2t_S(\cos k_x a+\cos k_y a)\ , \tag{2.6c}$$

$$\epsilon_{0\downarrow}(\vec{k})=\epsilon_0+\tfrac{1}{2}Um_0+2t_S(\cos k_x a+\cos k_y a)\ , \tag{2.6d}$$

and

$$m_0=\frac{1}{N}\sum_{\vec{k}}\langle b_\uparrow^\dagger(\vec{k},0)b_\uparrow(\vec{k},0)-b_\downarrow^\dagger(\vec{k},0)b_\downarrow(\vec{k},0)\rangle\ . \tag{2.6e}$$

Ground-state expectation values have been denoted as $\langle\cdots\rangle$.

Thus, the original Hamiltonian has been recast into a sum of H_k's, each of which is isomorphic to the Hamiltonian of a semi-infinite linear chain. These chains are coupled through the self-consistency relations (2.6) for the magnetization m_0.

These linear-chain Hamiltonians are conveniently treated by means of the transfer-matrix method[6] which allows us to obtain analytic expressions for the double-time Green's function

$$G_{ll'}^\sigma(\vec{k},t)\equiv\langle\langle b_\sigma(\vec{k},l;t)\,|\,b_\sigma^\dagger(\vec{k},l';0)\rangle\rangle\ , \tag{2.7}$$

which contains all relevant physical information. (Zubarev's notation[7] has been used.) In particular, the surface-layer magnetization m_0 is given by

$$m_0=-\frac{1}{\pi N}\text{Im}\sum_{\vec{k}}\int_{-\infty}^{\epsilon_F}[G_{00}^\uparrow(\vec{k},\omega)-G_{00}^\downarrow(\vec{k},\omega)]d\omega\ , \tag{2.8}$$

where ϵ_F is the Fermi energy and $G_{ll'}^\sigma(\vec{k},\omega)$ is the Fourier transform of $G_{ll'}^\sigma(\vec{k},t)$. Moreover, $G_{00}^\sigma(\vec{k},\omega)$ is

$$G_{00}^\sigma(\vec{k},\omega)=(\omega-\epsilon_{0,\sigma}-t_{SB}\{[\omega-\epsilon_B(\vec{k})]^2-t_B^2\}^{-1/2})^{-1}\ . \tag{2.9}$$

Our model allows for charge transfer between adjacent layers. Total charge conservation imposes an additional constraint which we handle by fixing the position of the surface level ϵ_0 in such a way as to assure charge neutrality of the whole system. To this end we use the generalized form of Friedel's sum rule,[8]

$$\Delta N = \frac{1}{\pi N} \sum_{\vec{k}} \eta(\epsilon_F, \vec{k}) , \tag{2.10}$$

where ΔN is the difference between nominal bulk and surface occupations and η is the phase shift

$$\eta(\epsilon_F, \vec{k}) = -\arg\det(I - VG_0) . \tag{2.11}$$

Here I is the identity matrix, G_0 is the Green's function of the infinite paramagnetic bulk, and V is the difference between the Hamiltonian of Eq. (2.2) and the one which describes the infinite paramagnet.

The actual calculation was implemented according to the following scheme.

(1) The nominal-bulk occupation n_B is chosen to be 1.18 electrons per atom which is an appropriate value[5] for amorphous Pb_3Bi. The surface-hopping parameter t_S is taken to be $t_S = \frac{1}{3}$, which is a representative value for d-band ferromagnets.

(2) The surface-nominal occupation n_S and the surface-bulk hopping t_{SB} are taken as variable parameters. Once these are chosen we obtain the magnetization m_0 as a function of U by means of the following procedure: (a) An initial value of $\Delta\epsilon \equiv Um_0$ is inserted in Eq. (2.6). (b) Equations (2.10) and (2.11) are solved for ϵ_0. (c) The magnetization m_0 is evaluated by means of Eq. (2.6e). (d) The Hubbard parameter U is now given by $\Delta\epsilon/m_0$.

Steps (b) and (c) require the evaluation of integrals over a two-dimensional Brillouin zone. These are reduced to one-dimensional integrals weighted by the density of states of a square lattice and then computed numerically.

On the basis of the numerical relation between U and m_0, calculated as outlined above, we obtain the results given in Sec. III.

III. RESULTS

In order to compare with experimental results, we now select the values of the parameters n_S and U so as to describe Ni, Co, and Fe thin films deposited on a paramagnetic metal.

The values of n_S are chosen to fit the fractional d-band occupations. The interaction parameter U is taken to reproduce the magnitude of the bulk magnetic moments, properly scaled to account for d-band degeneracy.

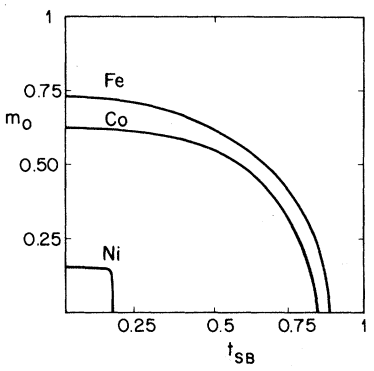

FIG. 1. Surface magnetization per atom as a function of surface-bulk coupling. The parameters are chosen to represent Ni, Co, and Fe.

Figure 1 displays the surface magnetization m_0 as a function of the surface-bulk coupling t_{SB}. These results indicate that m_0 is a decreasing function of t_{SB} up to a critical value, at which the film becomes paramagnetic. For nickel films this critical value turns out to be particularly small, and the transition to paramagnetic behavior is rather abrupt. This might provide an explanation for the disagreement between experimental results which do find[2] magnetic moments in very thin films and those[1] which do not. In the limit $t_{SB} \to 0$ the computed value of m_0 is in fair agreement with *ab initio* calculation[9] for isolated thin nickel films.

Cox *et al.*[5] have reported a similar calculation for a periodic slab geometry. Our results differ from theirs in two ways: First, our critical t_{SB} values are significantly smaller and second, we find a qualitative difference between Ni, on the one hand, and Co and Fe on the other.

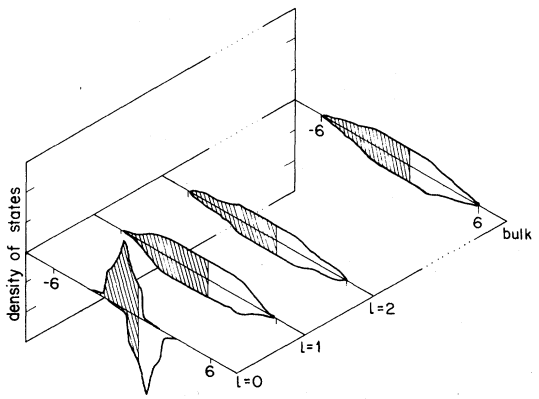

FIG. 2. Local density of states for the first three layers and for a bulk layer (in arbitrary units).

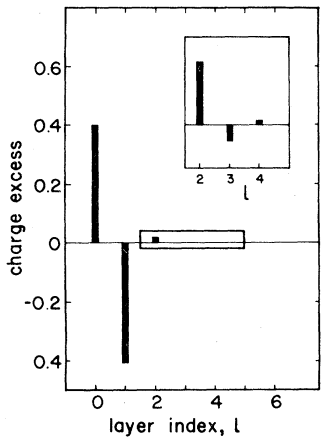

FIG. 3. Layer occupation per atom minus bulk occupation as a function of layer index. The vertical scale is magnified ten times in the inset.

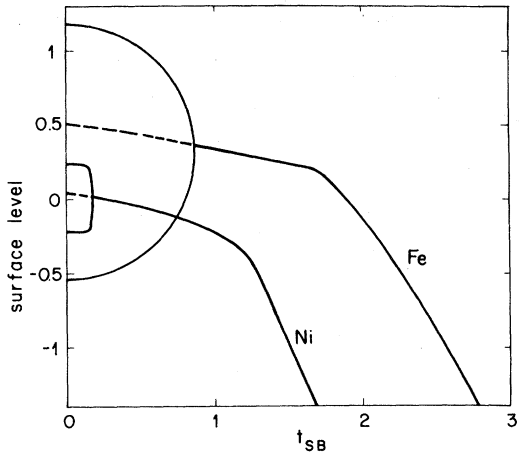

FIG. 5. Centers of surface bands as a function of surface-bulk coupling. In the magnetic regime the upper (lower) lines corresponds to the minority (majority) bands. The dashed lines correspond to a paramagnetic surface. The parameters are chosen to represent Ni and Fe as shown.

The local density of states for an iron film and for both spin directions is shown in Fig. 2. The surface layer has a marked two-dimensional character. Moreover, the majority states turn out to be mostly localized on the surface. In the $l=2$ layer, the density of states already closely resembles that of the bulk. This indicates that the surface perturbation does not extend beyond the first few layers, a fact that is also borne out by the way in which both the charge and the magnetization propagate into the bulk, as displayed in Figs. 3 and 4, respectively.

Finally, in Fig. 5 we show the position, relative to the center of the bulk band, of the center of gravity of the surface states for both spin directions as a function of t_{SB}. We notice that the broken line, which extrapolates the paramagnetic results into the magnetic region, does not coincide with the center of gravity of the spin-up and spin-down states. Moreover, it is a slowly varying function of t_{SB}.

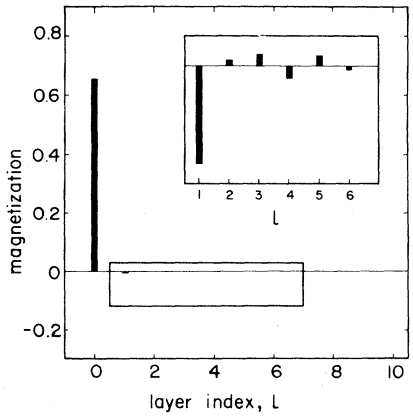

FIG. 4. Magnetization per atom as a function of layer index. The vertical scale is magnified 50 times in the inset.

IV. SUMMARY AND CONCLUSIONS

We have presented a model for the behavior of itinerant ferromagnetic thin films on paramagnetic metals. A half-space geometry has been considered, thus avoiding the uncertainties related to the use of periodic slabs. In addition, the Friedel sum rule has been employed to treat charge transfer between film and substrate in a rigorous way.

Our results differ in some aspects from those of Cox *et al.*[5] We attribute this to the different ways in which charge transfer is treated and, possibly to spurious effects related to interslab coupling. In addition, we do not assume that the surface level is independent of t_{SB}.

We find that the magnetization is confined to the magnetic film, except for small Friedel-type oscillations which decay slowly into the bulk. We also find that the majority states are mostly localized on the film.

The variation of m_0 with t_{SB} is qualitatively different for Ni, on the one hand, and for Fe and Co on the other. The m_0 vs t_{SB} curve for Ni is steplike in form, indicating that a small change in film-substrate coupling might turn the film from mag-

netic to nonmagnetic. This is in agreement with the results of Tersoff and Falicov[10] who studied a Ni-Cu system and found the magnetization of an Ni layer on Cu(111) to be significantly smaller than that of Ni deposited on Cu(100). In conclusion, the model presented is simple enough to investigate a wide range of systems, and at the same time it seems to explain the essential physics of the problem.

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