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Interface magnetization: Cu films on Ni(100)

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Results of a self-consistent tight-binding calculation for one and two atomic layers of Cu on ferromagnetic Ni(100) show a Cu-induced moment reduction of about 0.33 and $0.13\mu_B$ per atom in the first and second Ni layers, respectively. The Cu d orbitals play almost no role. Even a single layer of Cu gives an almost ideal (surface-insensitive) interface behavior in the Ni, so spin-polarized interface states should be easily observable by photoemission.

Recently, there have been exciting advances in the understanding of thin magnetic film systems, both experimentally[1] and theoretically.[2] When a thin layer of Ni is deposited on a non-transition-metal substrate such as Cu or Al, the magnetic moment per Ni atom is reduced. Physically, it is of great interest to distinguish, as far as possible, the effect of the free film surface from that of the metal-metal interface. For computational reasons, however, calculations tend to focus on ultrathin films. This presents two problems in interpretation. First, the magnetic film is very thin, so the surface and interface effects cannot be separated for comparison. Second, the substrate itself is treated as a thin film, which in some cases could pose a problem. Moreover, for the Ni on Cu system, comparison with experiment is somewhat ambiguous; Ni deposited on a Cu surface may be expected to form clumps, which would drastically alter the magnetic behavior.

We therefore chose to calculate the magnetic and electronic structure of one and two monolayers of Cu on the (100) face of a *semi-infinite* ferromagnetic Ni crystal. The results are of great interest for several reasons. (1) The system is realistic, since Cu does form almost ideal layers on Ni. (2) Even for a *single* layer of Cu, the effect of the surface on the magnetic behavior of the Ni substrate is very small. For two layers of Cu, the interface is quite ideal, i.e., isolated from the surface. (3) Because only one or two Cu layers are needed, the interface can be directly probed experimentally, e.g., by photoemission. It should therefore be possible to observe interface states, reduced local-exchange splitting, etc.

We find a reduction in the Ni magnetic moment at the interface of about $0.46\mu_B$ per interface atom, mostly in the first layer. This is in excellent agreement with the experimental results of Bergmann,[1] who found that depositing a Pb-Bi alloy on a Ni film reduced the magnetic moment, apparently by $\sim 0.4\mu_B$. This reduction is caused by changes in the shape of the projected Ni local density of states (LDOS), and by a weaker local exchange interaction resulting from reduced local d character of the states at the Fermi energy (E_F). We find that the Cu d band does *not* play a crucial role. These effects are discussed in more detail below.

In order to avoid problems with a thin Ni substrate, we use a Green's-function method, which has been discussed elsewhere.[3] It permits us to consider a semi-infinite crystal, with an arbitrary finite number of layers treated self-consistently, by means of a standard tight-binding Hamiltonian.

For the Hamiltonian we chose the parametrized scheme of Slater and Koster[4] (also known as LCAO). The electron-electron interaction was treated in the generalized single-site model, which has been extensively discussed.[5] In this way

$$H = H_0 + H_{ee} , \tag{1}$$

$$H_{ee} = \sum_{i\sigma\sigma'} \sum_{\mu\nu\lambda\kappa} U_{\mu\nu\lambda\kappa} c^{\dagger}_{i\mu\sigma} c^{\dagger}_{i\nu\sigma'} c_{i\lambda\sigma'} c_{i\kappa\sigma} , \tag{2}$$

where $c^{\dagger}_{i\mu\sigma}$ creates, at site i, an electron in an orbital of symmetry μ and spin σ. The one-electron term H_0 is parametrized in terms of one- and two-center integrals, chosen so as to give the correct paramagnetic band structure. We include the $3d$, $4s$, and $4p$ levels. The interaction term H_{ee} is treated in the Hartree-Fock approximation. The ratios of the screened interaction parameters $U_{\mu\nu\lambda\kappa}$ were chosen from atomic data,[6] solid-state Auger measurements,[7] etc., and the overall magnitude was adjusted to give the correct[8] bulk magnetization, $0.616\mu_B$. Details of the calculation will be given elsewhere. For the Ni-Cu intersite matrix elements, we took the geometric mean of the respective Ni-Ni and Cu-Cu matrix elements. The intersite matrix elements for the two metals are quite similar, so this is an excellent approximation. We sampled 15 wave vectors in the irreducible eighth of the two-dimensional (square) surface Brillouin zone. The potential was converged in each case to 0.03 eV (2 mRy).

To test our Hamiltonian and method we first calcu-

lated the magnetic and electronic properties of the Ni bulk and ideal (100) surface. We found excellent agreement with the results of Wang and Callaway[9] and Wang and Freeman.[10] In principle, these fully self-consistent calculations, which use a local-spin-density approximation for the potential, should be more accurate than the parameterized tight-binding approach used here. However, the favorable comparison suggests that our method is fully adequate for this sort of problem. In addition, by permitting us to treat a semi-infinite crystal, the method avoids the spurious enhancement of the Friedel-like spin oscillations, which occurs in calculations for very thin films.

We then performed calculations for one and two layers of Cu on Ni(100). For the interface and adjacent layers of Ni, Table I gives the magnetization μ, the change Δn_d in the occupancy of the d orbitals at the site relative to the bulk, and gives $D_d(E_F)$, the projected d component of the LDOS at E_F for the layer. The excellent agreement between the two geometries shows that even for a monolayer of Cu, the Ni properties are a good approximation to the ideal interface. This means that, with regard to the Ni magnetic behavior, the "isolated" interface is surprisingly accessible experimentally. For example, it should be possible to observe the highly localized majority-spin interface state which splits off the top of the Ni d band at the corner ($\bar{M}$) of the surface Brillouin zone. Details of interface states will be reported elsewhere.

The Ni interface layer has its magnetic moment reduced to about half the bulk value. The loss of magnetic moment due to the interface is almost entirely confined to the first two Ni layers. Each layer is essentially charge neutral, and the local magnetic moment in the Cu is negligible ($< 0.02\mu_B$) even in the interface Cu layer.

There is a slight increase in the local d occupancy in the Ni interface layer, because narrowing of the Ni LDOS at the interface causes the local d band to pull below E_F and become more full. This effect is quite modest, however, because of the energy cost of transferring charge from the sp band to the d band. Charge transfer cannot explain the substantial reduction in magnetization at the interface.

Part of the explanation can be seen in Fig. 1, where we show the LDOS at various layers near the interface for two-layer Cu on Ni(100). The bulk Ni is also shown for comparison. At the interface, the Ni LDOS becomes rounded, and in particular it loses the sharp upper band-edge characteristic of the ideal fcc d band. This reduces the LDOS at E_F at the interface, as seen in Fig. 1 and Table I. For a rounded band there is less tendency for the magnetization to go to saturation (one spin-band completely full) than in a band with a sharp edge. In a rigid-band model such as the Stoner model, for a square band the magnetization of a ferromagnet always proceeds to saturation at $T=0$. For a band with a smooth tail, however, saturation is never achieved.

In addition, the states at E_F have reduced local d character due to hybridization with the Cu conduction band at the interface. The local effective ex-

TABLE I. Properties of Ni layers at Ni-Cu(100) interface—magnetic moment, d-band filling, and d LDOS at E_F. (The Ni interface layer is denoted by I.)

	μ (μ_B)	Δn_d	$D_d(E_F)$ (eV^{-1})
Cu 1–Ni			
I	0.30	0.09	1.33
$I-1$	0.50	0.00	1.61
$I-2$	0.60	0.00	1.63
Cu 2–Ni			
I	0.28	0.08	1.29
$I-1$	0.49	0.00	1.62
$I-2$	0.60	0.00	1.62
Ni bulk	0.62	0	1.60

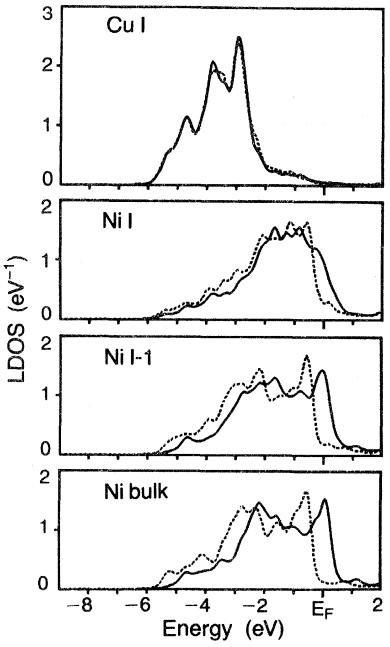

FIG. 1. Projected d component of LDOS at interface (I) and neighboring ($I-1$) layers of Ni-Cu(100) interface, from calculation for two-layers Cu on Ni(100). The Ni bulk is shown for comparison. Solid line—minority spin; dashed line—majority spin.

change potential felt by a state is roughly proportional to its local d character, and this local d character is reduced at the interface.

Some of the most interesting experiments[1] with thin Ni films have used free-electronlike substrates, and so it is important to know whether the d orbitals in Cu play an important role here. To examine this question we repeated the calculation above, but artificially removed the Cu d orbitals from the Hamiltonian. The resulting changes were remarkably small: no more than $0.04\mu_B$ for μ, and 0.02 for Δn_d, at any Ni site. For our purposes, the Cu d band can practically be regarded as an inert core level. This suggests that any difference between results for Ni on Cu, and on free-electronlike substrates, is due either to the lattice mismatch for substrates other than Cu, or more probably to the different hybridization strength between Ni d band and substrate conduction band.

In conclusion, we have found that there is a significant suppression of local magnetic moment at the Ni-Cu(100) interface. There is no appreciable penetration of the magnetization into the Cu. Rather, the Ni magnetization is reduced by $0.46\mu_B$ per interface atom, most of this in the interface layer. We attribute this reduction to the change in the shape of the Ni LDOS at the interface, and the consequent reduction in the LDOS at E_F; and to hybridization with the Cu conduction band, which reduces to d character of the states at E_F. For the Cu film on Ni geometry, a single layer of Cu gives a good interface, so the Ni interface behavior is surprisingly accessible to experiments such as photoemission. Since the Cu d band plays almost no role here, our results should apply equally to free-electronlike metals, except that the degree of hybridization between the Ni d band and substrate conduction band should vary from metal to metal.

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