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Ab initio studies of Ni–Cu–Ni trilayers: layer-projected densities of states and spin-resolved photoemission spectra

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Abstract. First-principles calculations have been performed in order to determine local magnetic moments, partial densities of states and spin-resolved photoemission spectra of Ni–Cu–Ni-trilayers. The tight-binding linearized muffin-tin orbitals (LMTO) method within the atomic sphere approximation has been used, and structural models have been built by translational repetitions of a face centred tetragonal-(001) Ni_4–Cu_N–Ni_4–ES_4 supercell ($N = 2, \ldots, 6$), where ES stands for an empty sphere and the four ES monolayers, separating the trilayers from one another, mimic the vacuum. The main finding of the present paper is that both densities of states as well as a Fermi energy (and thereby a work function) are to some extent sensitive to Cu-spacer thickness. In addition, a crossover from the antiparallel alignment of magnetizations of neighbouring Ni layers to the parallel configuration is computed, and it is confirmed that surface-Ni magnetic moments are enhanced in contrast to the Ni–Cu interface ones which are suppressed.

1. Introduction

Surface magnetism of transition metals has now been intensively studied for more than two decades, both experimentally [1–3] and theoretically [4–8]. Nickel, as a prototypical itinerant-electron ferromagnet, has received particular attention. Quite naturally, the first theoretical papers on magnetic properties of Ni were devoted to the most controversial topics such as: the dead layer question, the magnitude of the exchange splitting and the sign of photoelectron spin polarization (for an excellent review on surface magnetism see [9]). Studies of Ni in reduced geometry have evolved from pure Ni thin films, through Ni on noble-metal substrates, to Ni-based multilayers [10–13]. Unlike Co- and permalloy-based multilayers, multilayers with Ni have only been studied in a few experimental papers [10–13], and to the author's knowledge, in just one theoretical paper [14]. In particular, interlayer exchange coupling (IEC) [10, 12] and electronic transport properties [11, 13] have been investigated, whereas the theoretical paper [14] dealt with IEC and local magnetizations in infinite (no surface) Ni–Cu superlattices.

The aim of this paper is to test whether Cu-spacer thickness can influence surface properties of Ni-overlayers, especially their magnetic moments and spin-resolved photoemission spectra.

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2. Method of computation

The tight-binding linearized muffin-tin orbital method (TB LMTO) within the atomic sphere approximation (ASA) [15] has been used in its scalar relativistic version with the so-called 'combined corrections', accurate $\boldsymbol{k}$-space summation [16] and the exchange–correlation potential of von Barth and Hedin [17].

Structural models of Ni_4–Cu_N–Ni_4 trilayers with $N = 2, \ldots, 6$ have been constructed in the following manner. (i) On top of the fcc-(001) Ni–Cu–Ni, four monolayers of empty spheres (ES) have been placed in order to well isolate the trilayers from one another and mimic the vacuum at the Ni-layer surface. (ii) The Ni_4–Cu_N–Ni_4–ES_4 supercell has lateral dimensions of bulk Cu (with lattice constant $a_{Cu} = 3.615$ Å) and a tetragonal distortion in a perpendicular z-direction resulting from different interlayer spacings between non-equivalent monolayers. The spacings are equal to $R_{i,j} = \frac{1}{2}[\frac{1}{2}(a_i + a_j)^2 - a_{Cu}^2]^{1/2}$ where i and j run over Cu, Ni and ES, $a_{Ni} = 3.524$ Å, and a_{ES} has been arbitrarily put equal to a_{Cu}. (iii) The supercell is translationally repeated in all directions so as to make it possible to use the above mentioned computational TB-LMTO code (valid essentially for infinite crystalline structures).

3. Results and discussion

Numerical computations of the electronic band structure yield local magnetic moments of particular monolayers. As a test of the adopted method (with the empty sphere trick to imitate the vacuum) the calculated magnetic moments are compared with results of other authors for similar systems (identical systems are not available). For instance, the local magnetic moments (in μ_B) at Ni-monolayers, going from the surface down to the Ni–Cu interface, are distributed as follows: (0.68, 0.56, 0.60, 0.42), (0.68, 0.57, 0.61, 0.43), (0.69, 0.58, 0.61, 0.45), for Cu-spacer thicknesses of $N = 2, 4$ and 6, respectively. The agreement in local magnetic moments with other state-of-the-art calculations concerning Ni thin films and Ni–Cu bilayers [5–8] is fairly good (within an accuracy of a few per cent), and the well known observation that surface Ni-moments are enhanced whereas the interface ones are suppressed, is properly reproduced. Indirectly, it means that local densities of states are reliable, too. Indeed, it is easily seen from figure 1 that the local densities of surface- and subsurface-states show a typical behaviour, namely near the Fermi energy the former states are enhanced particularly down to around $E_F - 0.1$ Ryd, and their band is narrower than that corresponding to the latter states due to a partial dehybridization effect. In terms of the whole set of local orbital-resolved densities of states one can also compute photoemission spectra (PES). The method used here consists of weighting the DOS with appropriate atomic cross-sections ($c_{i,l}$) for photon scattering [19] and applying a convolution with a Lorentzian function (g) which accounts for finite experimental resolution, δ. Explicitly:

$$\mathrm{PES}(\sigma, i) = \sum_l c_{i,l} \int \widetilde{\mathrm{DOS}}(i, l, \sigma, E') g(E - E') \mathrm{d}E' \tag{1}$$

$$g(x) = \frac{1}{\pi} \frac{\delta}{\delta^2 + x^2} \tag{2}$$

where σ, i and l stand for the spin, the monolayer label and the orbital, respectively, and $\widetilde{\mathrm{DOS}}$ is equal to DOS except for $E > E_F$, where it is set to zero.

In figures 2 and 3, DOS of three outermost Ni monolayers are depicted. Figure 2 corresponds to a trilayer with $N = 4$ which is antiparallelly aligned, and figure 3 corresponds

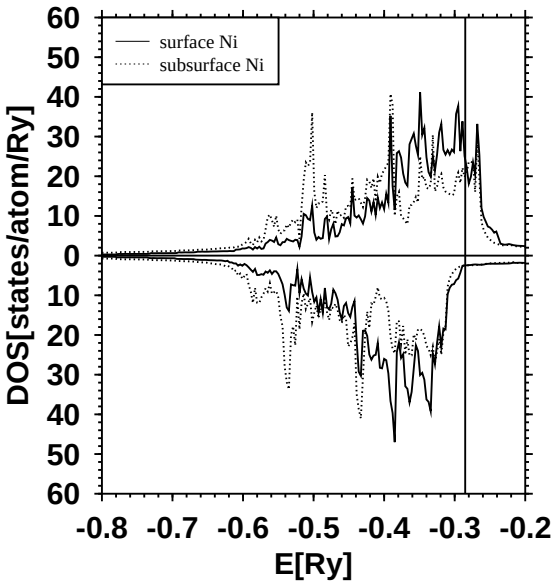

Figure 1. Surface (solid line) and subsurface (dotted line) densities of states for Ni_4–Cu_4–Ni_4–ES_4 with the antiparallel alignment of magnetizations of neighbouring Ni layers. The ESs are empty spheres which separate the Ni–Cu–Ni trilayers from one another and mimic the vacuum on the Ni-surface. The upper part of the figure corresponds to minority-spin electrons, whereas the lower part corresponds to majority-spin electrons (the vertical line denotes the Fermi energy).

to $N = 6$ (parallel configuration). These DOS have been used to compute photoemission spectra plotted with a dashed line in figures 2 and 3. Although the computed photoemission spectrum is simply a smoothed DOS, it deserves presentation since it is in principle directly comparable with an experiment and additionally it visualizes how the energy resolution filters out some details of the density of states. Contributions of as many as three surface monolayers (ML) have been taken into account in order to be consistent with the average escape depth estimated in [3].

Within the present approach a crossover from the antiparallel alignment to the parallel one has been found on the basis of total energy calculations (similar to that found in [18]) and shown to occur between $N = 5$ and 6 Cu-monolayers at roughly 10 Å, i.e. about $1\frac{1}{2}$ ML earlier than reported in [10]. The discrepancy may be due to different Ni-thicknesses in both cases, since it is known that magnetic slab thickness influences the phase of IEC oscillations without actually changing the period (see e.g. [20]). Incidentally, the thickness of the ES layer is set to four; this has not been changed because, judging from the tiny charges and magnetic moments of the inner ES monolayers, it separates the metallic trilayers well enough from each other. The $\boldsymbol{k}$-space sampling is quite dense and the number of irreducible $\boldsymbol{k}$-points is equal to 1320. Moreover, the Wigner–Seitz radii of Ni and Cu are independent of the Cu-spacer thickness, and only the ES radius is allowed to change in order to make the sum of all the atomic spheres equal to the volume of the system. It seems rather unlikely that another choice of the above mentioned 'hidden parameters', i.e. the Wigner–Seitz radii, $\boldsymbol{k}$-points sampling or even the ES-layer thickness, could change the crossover value, $N = 5, 6$, by more than one monolayer. A major effect which accompanies the crossover

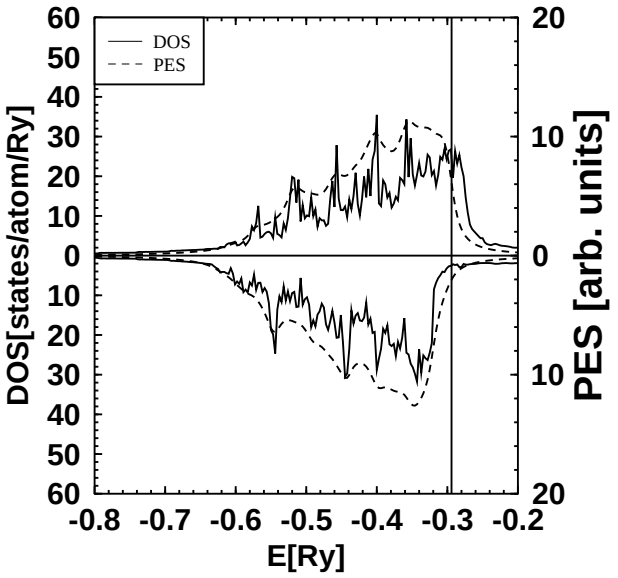

Figure 2. Surface densities of states (solid line) coming from the first 3 outermost Ni monolayers of the Ni_4–Cu_4–Ni_4–ES_4-trilayer in the antiparallel configuration. The dashed line corresponds to photoemission spectra (PES) obtained from the DOS's by weighting them with photon scattering cross-sections and applying a convolution with an energy-resolution Lorentzian function. The parameters used are: $\delta = 0.01$ Ryd (energy resolution) and $E_{ph} = 5.88$ Ryd $= 80$ eV (photon energy).

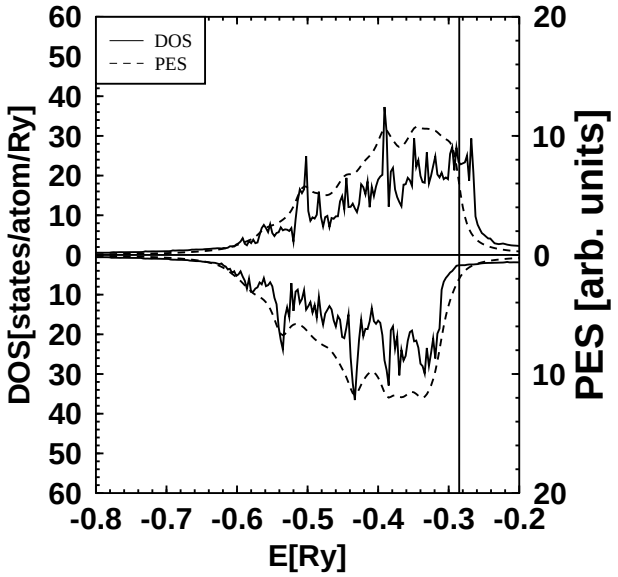

Figure 3. As in figure 2 but for the Ni_4–Cu_6–Ni_4–ES_4-trilayer in the parallel configuration.

is the change of the spin polarization due to the switching-over of the magnetizations of the Ni layers, and the second, less pronounced but noteworthy, effect is a change in the Fermi energy which must result in changing the work function. A slight modification of the majority-spin PES at around -0.35 Ryd is also visible (a flatter and broader peak for $N = 6$ than for $N = 4$). Moreover the DOS above E_F is also modified, and has a 0.01 Ryd wide mini-plateau for $N = 6$. The features above E_F might be measurable with high-resolution spin-resolved *inverse* photoemission [21]. Therefore it is suggested that it is necessary to look experimentally at the influence of the Cu-spacer thickness in Ni–Cu–Ni trilayers for predicted effects, i.e. the DOS (PES) features and the work function values.

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