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Exchange splitting and ferromagnetic order in a monolayer film of cobalt on Cu(111)

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The analysis of exchange splitting in a monolayer and a double layer of Co on Cu(111), which was given recently by Miranda *et al.* on the basis of their photoemission spectra of such films, is completed by combination with former measurements of the spontaneous magnetization in these films. The result is strong evidence for the intra-atomic origin of exchange splitting in Co, which therefore cannot be taken as a direct measure of long-range-order spontaneous magnetization.

In a recent paper, induced by the interest in ferromagnetic order in monolayer films, Miranda *et al.*[1] published angle-resolved photoemission spectra (PES) for one and two monolayers of Co on Cu(111); the analysis of magnetism in these films given by Miranda *et al.* is incomplete in neglecting what is known on the spontaneous magnetization in these films. The aim of the present Comment is to complete this analysis, including both photoemission and magnetometric data.

Ferromagnetic order in monolayer magnetic films and in single-crystal films consisting of only a few atomic layers has been analyzed for many years. A decrease of spontaneous magnetization and Curie temperature with decreasing thickness was established in good agreement between theory[2,3] and magnetometric measurements with epitaxial films of 48 at.% Ni–52 at.% Fe(111) (Ref. 4) and Co (Refs. 5 and 6), both prepared on Cu(111) in UHV and coated by Cu (for a review refer to Ref. 7). In particular, a monolayer of Co [fcc(111) or hcp(0001)] in a Cu matrix turned out to be ferromagnetic with T_c (monolayer) = 0.30 T_c (bulk) and a linear dependence of the spontaneous magnetization on temperature. For the case of Ni-Fe films the good agreement of the experimental magnetization with the theory of the bare size effect,[2,3] which neglects any electronic interaction with the matrix, was taken as an indication for weak interaction between the magnetic *d* band in the magnetic film and the electron system of the Cu matrix.[7] It is reasonable that this conclusion, which was taken from the Ni-Fe experiments, applies also to the Co films.

Just this conclusion was drawn independently by Miranda *et al.* from the analysis of their photoelectron spectra, resulting in "observation that the interaction between Co and Cu is rather weak, indicating that Co on top of Cu behaves like a quasi-two-dimensional transition metal." This weak interaction being established from both types of experiment, we can, for the discussion of the magnetic properties of the films, neglect the difference that the films for magnetometry were coated by Cu whereas the films for PES were uncoated.

The most interesting feature of the photoelectron energy distribution of the monolayer and the doublelayer Co on Cu is the appearance of a characteristic three-peak structure, known from the bulk Co(0001) surface.[8] The analysis of this three-peak structure in terms of exchange splitting ΔE_{ex} results in $\Delta E_{ex} = 0.7$ eV for both the monolayer and the double layer, for the point $\bar{\Gamma}$ of the two-dimensional Brillouin zone. For comparison, the bulk Co(0001) surface, at Γ, is characterized by $\Delta E_{ex} = 1.0$ eV [mean value for the upper *d* band (0.85 eV) and the lower *d* band (1.2 eV)].

An important problem for the photoemission analysis of magnetism in surfaces and in thin films concerns a possible relation between exchange splitting $\Delta E_{ex}(D;T)$ and spontaneous magnetization $J_s(D;T)$, both as a function of the number of atomic layers in a film D and of the temperature T. Rigid-band considerations suggest proportionality of ΔE_{ex} and J_s. Regarding the dependence on temperature, this suggestion was contradicted, for the case of bulk Ni(111), by Eastman *et al.*,[9] who showed, by angle-resolved photoemission, that ΔE_{ex} persists above T_c, with $\Delta E_{ex}(1.3\ T_c) = 0.6\ \Delta E_{ex}(0\ \mathrm{K})$. This indicates a localized intra-atomic origin of ΔE_{ex}, which therefore cannot be taken as a measure of ferromagnetic long-range order for the case of Ni. This behavior was demonstrated independently for the case of Ni by appearance potential spectroscopy[10] and two-electron-capture spectroscopy.[11] It is in accordance with modern theories of magnetism in 3*d* metals.[12,13]

The same conclusion can be drawn for the case of Co from the present experiments with the monolayer and the double layer. This can be seen as follows: The photoemission spectra were taken at room temperature[14]; they should be compared with the mag-

netization at the same temperature. At 300 K, the spontaneous magnetization of the monolayer and the double layer, in comparison with the bulk magnetization at 0 K, $J_s(\infty; 0)$, has decreased to $J_s(1; 300\ \mathrm{K}) = 0.28\ J_s(\infty; 0)$ and $J_s(2; 300\ \mathrm{K}) = 0.7 J_s\ (\infty; 0)$, respectively, as taken from magnetometry.[6,7]

Contrarily, the photoemission analysis (M) results in $\Delta E_{ex}(1; 300\ \mathrm{K}) = \Delta E_{ex}(2; 300\ \mathrm{K}) = 0.7 \Delta E_{ex}(\infty; 300\ \mathrm{K})$. This means that the exchange splitting ΔE_{ex} is the same for the monolayer and for the double layer, whereas the spontaneous magnetization J_s differs by more than a factor of 2. The conclusion is that again the exchange splitting, determined from photoelectron spectroscopy, cannot be taken as a measure of spontaneous magnetization. Therefore the photoemission from the Co monolayer and double layer provides further experimental evidence for the intra-atomic nature of exchange splitting in $3d$ metals, in particular, for cobalt.

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