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Spin-resolved photoemission and band-mapping in epitaxial fcc Fe–Co alloys on Cu(100)

M. Zharnikov, A. Dittschar, W. Kuch, C.M. Schneider, J. Kirschner

MPI für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

Abstract

The electronic properties of fcc Fe–Co/Cu(100) were investigated by spin-resolved photoelectron spectroscopy. At a fixed concentration a dispersion of the majority and minority bands with Δ_5-symmetry towards higher binding energy along the Δ-axis of the Brillouin zone could be followed upon increasing photon energy. Upon variation of the concentration the spectra for the alloy films reveal a gradual shift of the Δ_5 minority spin band through the Fermi level and a change of the sign of the spin polarization at the Fermi level in full agreement with the expected tendency from Co to Fe. Such a behavior agrees with both a simple mixture of the Fe and Co electronic properties in a randomly mixed alloy film, and a chemical separation (clustering) of both constituents.

Keywords: Spin-resolved photoelectron spectroscopy; Thin films – epitaxial; Film growth

Epitaxial growth of ultrathin films allows the stabilization of structural phases which are unstable in the bulk. Recently we have succeeded in stabilizating fcc Fe–Co alloys (stable in the bulk only above 1260 K) at room temperature over a wide range of compositions by epitaxial growth on Cu(100) [1]. We have investigated the growth mode of these alloy films as well as their structural and magnetic properties [1]. In this paper we address the electronic properties of this system. We take advantage of the fact that the electronic properties of both Fe and Co ultrathin films on Cu(100) have been rather extensively studied (see e.g. Refs. [2–8]); these data are used as a starting point to analyze the electronic properties of the alloy films.

The Fe_xCo_{1-x} films were epitaxially deposited in a UHV chamber by coevaporation from two separate sources onto the Cu(100) single crystal kept at room temperature; the resulting composition of the films was determined with an accuracy of 5%. As it was previously reported, Fe–Co alloy films grow pseudomorphically on Cu(100). The films assume a tetragonally distorted fcc structure (fct). At low Fe concentration this structure is tetragonally compressed, whereas in Fe-rich films a strong tetragonal expansion takes place. In the intermediate region of Fe concentrations (25–65% of Fe) a coexistence of both compressed and expanded fct phases is found.

The films are ferromagnetic with an in-plane easy axis of magnetization (only for very small thicknesses and large Fe concentrations the easy axis of magnetization is found normal to the surface [9]). Strong in-plane anisotropy is not observed. The dependence of the saturation magnetization (measured by using magneto-optical Kerr effect) on composition, namely a linear increase with the Fe content up to 95% Fe implies a stabilization of strong ferromagnetism (completely filled majority spin band) in the alloy films [10] which should reveal itself in their electronic properties.

These properties were studied by means of spin-resolved photoelectron spectroscopy using circularly polarized synchrotron radiation from the 6.5 m normal-incidence monochromator beamline at the BESSY storage ring. The spin- and momentum-resolved spectrometer described earlier [11] was used. The measurements have been carried out at normal light incidence and normal emission in order to eliminate magnetic dichroism effects and make contact to previous investigations of Fe and Co along a high symmetry line. The samples were magnetized along the [100] in-plane direction. The films of 5 ML thickness were investigated; these films are believed [3,5,7] to be thick enough to develop a fully three-dimensional bulk band structure.

In Figs. 1–3 sets of spin-resolved spectra (left columns) for 5 ML alloy films are depicted together with the corresponding spin polarization distributions (right columns). In Figs. 1 and 2 alloy composition is varied at fixed photon energy; at $h\nu = 14$ eV (Fig. 1) and $h\nu = 21$ eV (Fig. 2) different regions of the Brillouin zone are probed. In Fig. 3 alloy composition is fixed and photon energy is varied.
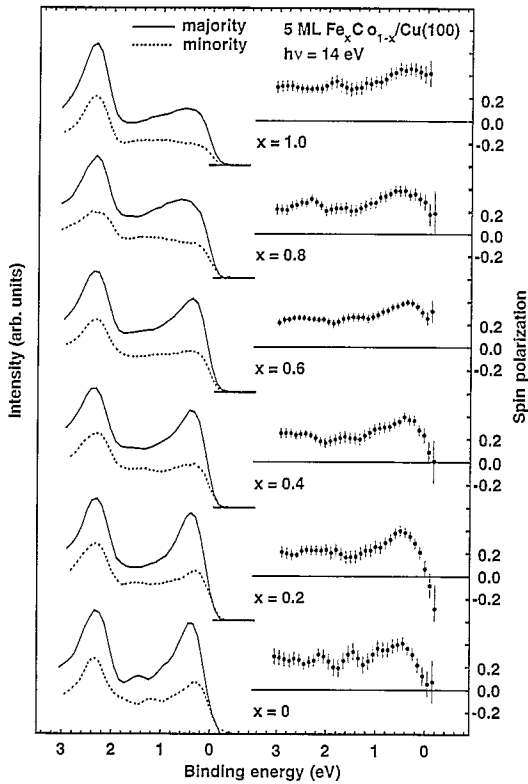
Fig. 1. Spin-resolved photoemission spectra (left column) and the corresponding in-plane spin polarization distributions (right column) obtained from 5 ML Fe_xCo_{1-x}/Cu(100) films (3 ML at $x = 1$) at a photon energy of 14 eV. The spectra for the majority and minority electrons are depicted using solid and dashed lines, respectively.

Fig. 2. Same as Fig. 1 but for 5 ML Fe_xCo_{1-x}/Cu(100) films (3 ML at $x = 1$) at a photon energy of 21 eV.
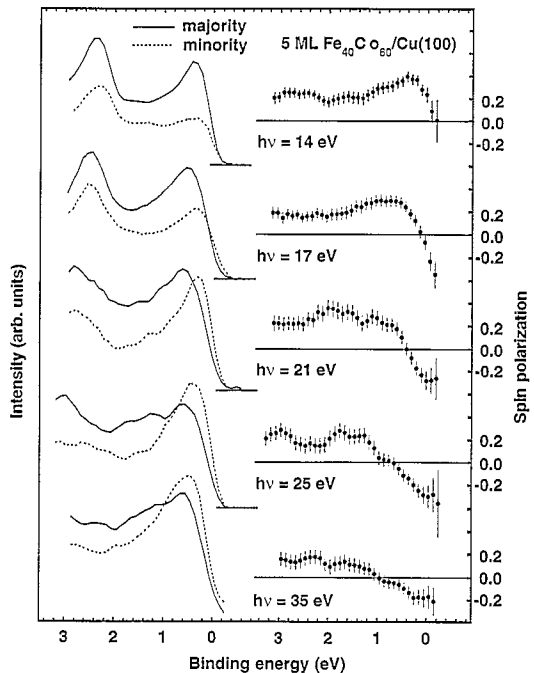
Fig. 3. Same as Fig. 1 but for 5 ML $Fe_{40}Co_{60}$/Cu(100) film at different photon energies.

The spin polarizations in Figs. 1–3 were calculated by normalizing the difference between the majority and minority contributions to their sum. The spectra in Figs. 1–3 have two main features in common: a threshold-like structure just below the Fermi level and a peak centered around a binding energy of 2.4 eV. By comparison with spectra of clean copper, the former structure is found to be mainly due to cobalt and iron; the peak at ∼ 2.4 eV is dominated by contributions from copper and therefore lies outside the scope of this paper.

The analysis of the data is complicated by two circumstances. First, the symmetry of the system is reduced from 4mm to $\overline{2}mm$ because of the in-plane orientation of the magnetization. Therefore, the electronic states cannot be classified by the irreducible representations Δ_6 and Δ_7 of the double group associated with 4mm; Δ_6 and Δ_7 split into four bands of different symmetry in the ferromagnets. Instead, there are only two one-dimensional representations γ_6 and γ_7, which should result in very extended hybridization of the electronic states of different spatial symmetry. The original dispersion of the bands will there-
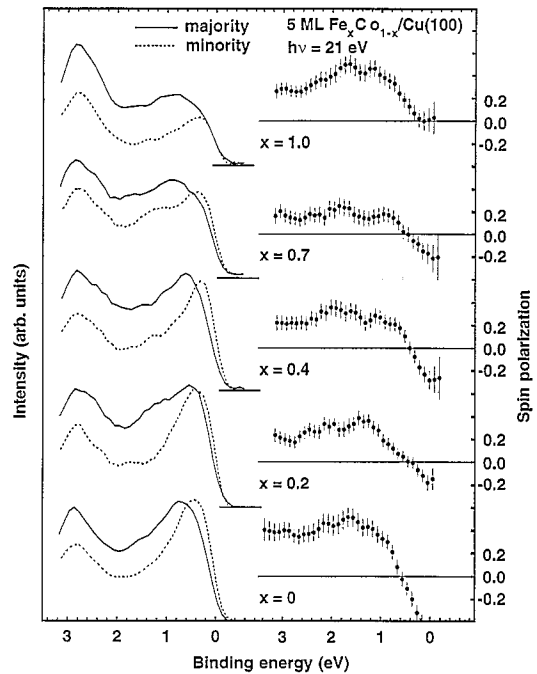
fore be distorted. Because of an avoided crossing of the bands with the same symmetry, hybridization gaps will appear. In the regions of such gaps strong mixing of spatial symmetries between different bands will take place because of the spin–orbit interaction.

The second circumstance complicating our analysis is the lack of band structure calculations for fcc Fe–Co alloys. Therefore only calculations for the pure fcc Fe [2,8,12] and Co [5,8] can be used to interpret the experimental results. It is well known that fcc Fe assumes several magnetic phases: an antiferromagnetic phase (isotropic fcc structure) and two ferromagnetic phases with high and low spin [12–14]. The ferromagnetic solutions only exist at an enlarged value of the atomic volume. This is obviously the case for the Fe-rich Fe–Co alloy films, because of a strongly tetragonally expanded fcc structure. Taking into consideration the composition dependence of the magnetization (see above) one will rather expect the high spin behavior in this composition region. Therefore, the band structure calculations for the high spin phase of fcc Fe [12] are taken to analyze photoemission data. These calculations together with analogous calculations for fcc Co [5] are depicted in Fig. 4.

Our experimental geometry restricts the possible initial states by selection rules to bands of Δ_5 spatial symmetry (solid lines), whereas the final states are represented by a band of Δ_1 spatial symmetry (the sections of the final state band shifted by a fixed photon energy are indicated by thin dashed lines in Fig. 4) [3]. The Δ_5-bands in Co are characterized by a smaller value of the exchange splitting and are located lower with respect to the Fermi level as compared to Fe. Whereas the difference between the $\Delta_5^\uparrow$-bands in Fe and Co is relatively small, this is not the case for the $\Delta_5^\downarrow$-bands in these systems. The $\Delta_5^\downarrow$-band in Fe intersects the Fermi level near the Γ point, whereas for Co such a crossing happens in the middle of the Brillouin zone. Assuming that the band structure of the Fe–Co alloys is placed in between those for Fe and Co with respect to the energetic positions and the exchange splitting of the bands, one can expect a strong change of the minority contribution in the experimental spin-resolved spectra upon variation of the alloy composition. In fact, one can easily follow such a change in Figs. 1 and 2. The spectra reveal a gradual shift of the minority band above the Fermi level with increasing Fe concentration (the character of polarization at the Fermi level also changes), in full agreement with the tendency expected. For $h\nu = 14$ eV (Fig. 1) the $\Delta_5^\downarrow$-band should lie above the Fermi level for both Co and Fe (see Fig. 4); the minority contributions at this energy are therefore relatively small and the spin polarization at the Fermi level is positive. At $h\nu = 21$ eV (Fig. 2) the middle of the Brillouin zone is probed; the Fermi level crossing of the $\Delta_5^\downarrow$-band can then be observed, especially if a hybridization between this band and a band of Δ_1 spatial symmetry occurs near E_F (the position of the Δ_1-band should change upon variation of the alloy compo-
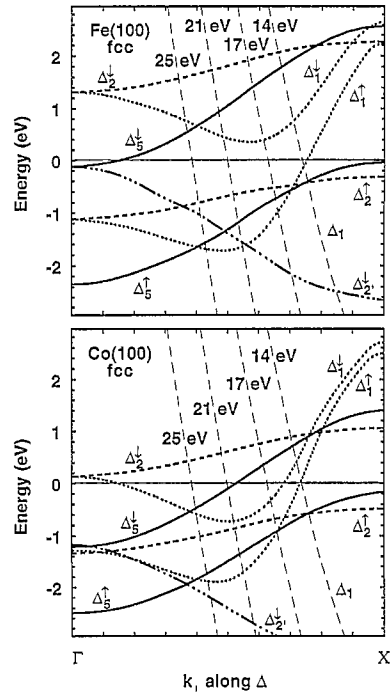

Fig. 4. Semirelativistic band structure calculations for the high-spin phase of the fcc Fe [12] and full relativistic band structure calculations for fcc Co [5]. Bands hybridization in the case of Co has been excluded to simplify the comparison with the band structure of Fe. The spatial symmetry of the individual bands and their prevailed spin character are marked. The thin dashed lines are sections of the final state band, shifted by the photon energy indicated at the respective line. The crossings of these sections with the initial state bands of the Δ_5 spatial symmetry provide the binding energies of the initial states at a given photon energy.

sition). Such a crossing can be inferred at Fe concentrations of 40%, because of the observed narrowing of the peak just below the Fermi level in the minority spectra for this composition. The negative sign of the spin polarization at this composition and a dispersion of the minority peak with decreasing Fe concentration below 40% support this conclusion (at Fe concentrations above 40% only a growth of the minority peak is observed).

As to the $\Delta_5^\uparrow$-band, it should in principle be less strongly dependent on the alloy composition than the $\Delta_5^\downarrow$-band, because of the small difference between the $\Delta_5^\uparrow$-bands in Fe and Co. Nevertheless, some pronounced changes of the majority contribution with the composition can be expected just because of hybridization effects. The bands of Δ_1, Δ_2 and $\Delta_{2'}$ spatial symmetry hybridize strongly with the $\Delta_5^\uparrow$-band, the regions of the Brillouin zone where the hybridizations occur and the strength of this effect being strongly dependent on the alloy composition. This partly explains the changes in the majority spin spectra upon variation of the composition in Fig. 1 and Fig. 2. A small gradual upward shift of the $\Delta_5^\uparrow$-band with

increasing Fe concentration (which is not very pronounced in the spin-resolved spectra) can clearly be traced in the spin polarization at $h\nu = 14$ eV (Fig. 1) as a shift of the positive peak at binding energies of 0.3–0.5 eV.

The electronic properties of the Fe–Co alloy film at a fixed composition are qualitatively similar to those of pure Fe and Co. This can clearly be seen in Fig. 3 where the photoemission data for 5 ML $Fe_{40}Co_{60}$/Cu(100) film for different photon energies are depicted. The spectrum at $h\nu = 14$ eV shows a prominent majority peak near the Fermi energy. With increasing photon energy the majority peak disperses away from E_F and becomes broader, and a minority peak evolves just below the Fermi energy and disperses with increasing energy above 21 eV. This behavior can be correlated to the dispersion of the bands with Δ_5 spatial symmetry towards higher binding energies.

At $h\nu = 35$ eV we do not probe a specific region of the Brillouin zone but get some information on the wave vector integrated density of states along the Δ-axis in k-space [4]. The peak at ~ 2.4 eV in the minority spectrum at $h\nu = 35$ eV in Fig. 3 and the strong peak at ~ 0.5 eV in the corresponding minority spectrum can therefore be related to the majority and minority Δ_5-states near the Γ point. It provides a value of about 1.9 eV for the exchange splitting in this specific region of the Brillouin zone which correlates rather well with the value of the magnetic moment of 2.06 μ_B obtained for $Fe_{40}Co_{60}$ by a simple weighted sum of the magnetic moments of Co and high spin Fe (the effective Stoner parameter can be assumed as 1 eV/μ_B [10,15]).

In conclusion, we have shown that the electronic properties of the fcc Fe–Co alloys on Cu(100) are in between of the analogous properties of pure fcc Co and the high-spin phase of fcc Fe. This is implied by both the composition dependence of the minority photoemission spectra and the value of the exchange splitting.

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