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Ferromagnetic order in ultrathin Rh layers on Fe(100)

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The electronic structure and magnetic properties of ultrathin Rh layers on Fe(100) have been studied by spin- and angle-resolved photoelectron spectroscopy. Spin-resolved valence-band and core-level spectra show that Rh monolayers are ferromagnetically ordered at room temperature with their spin moments predominantly coupled parallel to the Fe ones. This is in agreement with the results of our FLAPW calculation for the ground state of the system. Overlayers thicker than 3 ML show no spin polarization at room temperature. Our results indicate that the main origin of the development of Rh local moments and their long-range ordering within the overlayer is the direct overlayer-substrate hybridization rather than the dimensionality of the system.

Current *ab initio* calculations[1–3] point out that monolayers of the 4*d* metals Ru ($1.7\mu_B$) and Rh($1.1\mu_B$) as well as the 5*d* metal Ir ($0.9\mu_B$) possess a ferromagnetic ground state with large magnetic moments when epitaxially grown on Ag(100) or Au(100) surfaces. In those cases the magnetism is stablized due to the low dimensionality of these systems. These materials provide the combination of magnetism and large spin-orbit interaction and one expects new results on the magnetocrystalline anisotropies and thermodynamical properties of the magnetic films. Those theoretical predictions need, however, still to be experimentally confirmed.

Growing 4*d* or 5*d* metals on magnetic substrates adds another possibility of obtaining magnetic 4*d* and 5*d* films. In this case magnetism is induced due to *d*-*d* hybridization between the overlayer and substrate. Until now among the possible 4*d* and 5*d* metals only Pd (Refs. 4–8) and Pt (Ref. 9) have been studied as overlayers on magnetic substrates or as multilayers. The choice of these overlayers has been derived from their large magnetic susceptibilities as bulk materials. As shown recently for the case of Pd / Fe(100) (Ref. 10) the strong hybridization between Pd and Fe leads to a new material with properties quite different from bulk Pd. This spurred our systematic theoretical study of Ru, Rh, and Pd monolayers on Fe(100) by *ab initio* calculations using the full-potential linear augmented-plane wave (FLAPW) method[11] to determine the overlayer system with the most pronounced magnetic properties. The calculations show that among the three systems studied Rh has the largest magnetic moment with $0.82\mu_B$ while the moments of Ru and Pd monolayers on Fe are $0.49\mu_B$ and $0.29\mu_B$, respectively. Thus the magnetic moment of Rh in Fe(100) is nearly as large as on Ag(100) and considerably larger than that of dilute Rh in Fe ($0.60\mu_B$).[12]

Along with our FLAPW study we present experimental results probing the electronic and magnetic structure of ultrathin Rh films evaporated on Fe(100). We have applied spin-resolved and spin-integrated photoelectron spectroscopy, Auger electron spectroscopy, and low-energy electron diffraction (LEED) to investigate the growth mode and the crystallographic, electronic, and magnetic structure of the Rh overlayers. We give direct evidence that Rh monolayers deposited on the Fe(100) surface are ferromagnetically ordered.

The overlayers were produced *in situ* by e^--beam evaporation calibrated by a quarz microbalance thickness monitor. The base pressure in the UHV system during the experiments was $(1-2)\times 10^{-10}$ mbar. It could be kept below 3×10^{10} mbar during Rh evaporation. All experiments described and the evaporations were made at room temperature. The spin-resolved photoelectron spectra were recorded with a high-transmission 90° spherical analyzer in front of a 100-keV Mott scatterer. Excitation source was the TGM5 wiggler-undulator beamline[13] at the BESSY storage ring providing intense linearly polarized synchrotron radiation. During the measurement the sample was remanently magnetized along an in-plane [010] direction of the Fe substrate.

Our LEED study of the system revealed nonepitaxial overlayer growth. Starting from the sharp-diffraction spots on a low intensity background from the clean and annealed Fe substrate, the spots rapidly became diffuse along with a strong increase of the background intensity during deposition of the Rh overlayers. At 3-ML Rh coverage, the LEED spots were no longer visible at primary kinetic energies below 100 eV. The atomic size mismatch between Rh and Fe is 7.7%, i.e., presumably somewhat too large to allow epitaxial growth of the overlayer. The growth mode perpendicular to the surface has been probed by core-level photoelectron and Auger electron spectroscopy. Upon Rh deposition the Fe 3*p* level was continuously attenuated and it vanished nearly completely at about 5-ML overlayer thickness. With increasing thickness neither the position nor the line shape of the Fe 3*p* peak was affected. Our analysis of the data shows that the Rh layers on Fe(100) grow in the layer-by-layer mode or very close to it. In particular, significant contributions of either island growth or alloy formation are not compatible with our results.

Information about the electronic structure of the system is obtained by energy-distribution curves (EDC's) from the *valence-band* region of the system shown in Fig. 1 as a function of overlayer thickness. The spectra are taken at 45-eV photon energy with normal light incidence and normal electron emission. Starting from the clean Fe a relatively fast change of the valence-band emission is observed upon depositing the Rh overlayer. Along with a strong attenuation of the Fe peak close to the Fermi level (E_F) a new structure develops at around 2-eV binding energy, the latter being due to mainly Rh 4*d* derived emission. Going from 1–5 ML the peak maximum of the Rh 4*d* states shifts from 1.8 to 2.6 eV binding energy representing the evolution of the Rh valence band with the overlayer thickness. Above 5 ML thickness no further changes of the electronic structure could be detected.

In Fig. 2 we present spin-resolved energy-distribution curves recorded under the same conditions as the EDC's in Fig. 1. The spectrum for the clean substrate is similar to the one from the Γ point of the Brillouin zone of bcc Fe which has been interpreted earlier.[14] It is determined by strong minority spin emission close to E_F and two majority spin peaks at higher binding energy as indicated in the figure. With increasing Rh deposition first a strong attenuation of the minority spin peak is found already at 0.25 ML thickness, which might be due either to elastic scattering of strongly forward directed photoelectrons when passing the unordered Rh overlayer or to suppression of a surface resonance at the $\bar{\Gamma}$ point of the surface Brillouin zone of the Fe(100) surface.[15] This attenuation is followed by the development of new structures with opposite spin character. For 1 ML thickness the main majority and minority spin features due to Rh 4*d* emission are located at 2.5 and 1.8 eV binding energy, respectively. Thus, the spin splitting of these peaks is 0.7±0.2 eV.

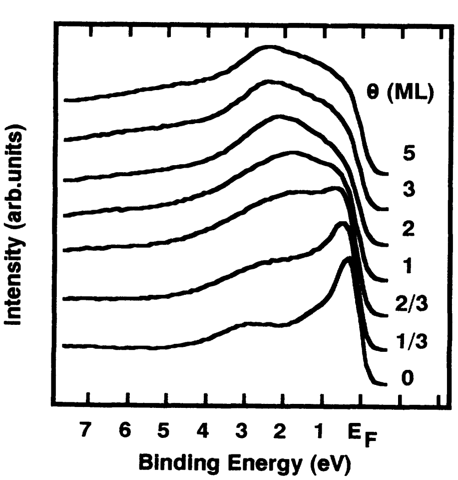

FIG. 1. Spin-summed EDC's for Rh/Fe(100) at $h\nu = 45$ eV. Rh overlayer thickness as indicated.

The spin polarization of the Rh 4*d* states might be better illustrated in the difference curves in Fig. 3. We have substracted a spectrum of 0.5 ML Rh/Fe(100) from one of 1.3 ML simultaneously for both spin channels. The so determined contribution of the overlayer to the total valence-band emission of 1.3-ML Rh/Fe(100) is strongly spin polarized and contains different spin split features. It is obvious that the Rh sites carry long-range-ordered magnetic moments.

Our results are further supported by spin-resolved *core-level* spectroscopy data from the Rh 4*p* and the Fe 3*p* states. The spectrum in Fig. 4, taken at 120-eV photon energy, has been recorded from a sample of 1.2± 0.2-ML Rh/Fe(100). In spin-summed photoelectron spectroscopy the Fe 3*p* peak is located at 52.8±0.2 eV and the main peak of the Rh 4*p* emission appears at 5.5±0.2 eV lower binding energy. The Fe 3*p* emission is dramatically spin dependent similar to the clean metal case described by Carbone and Kisker[16] and by Sinkovic *et al.*[17] The spin-down peak is more pronounced, it has the smaller linewidth, and lower binding energy than the spin-up one. The measured spin splitting is 0.5±0.1 eV,

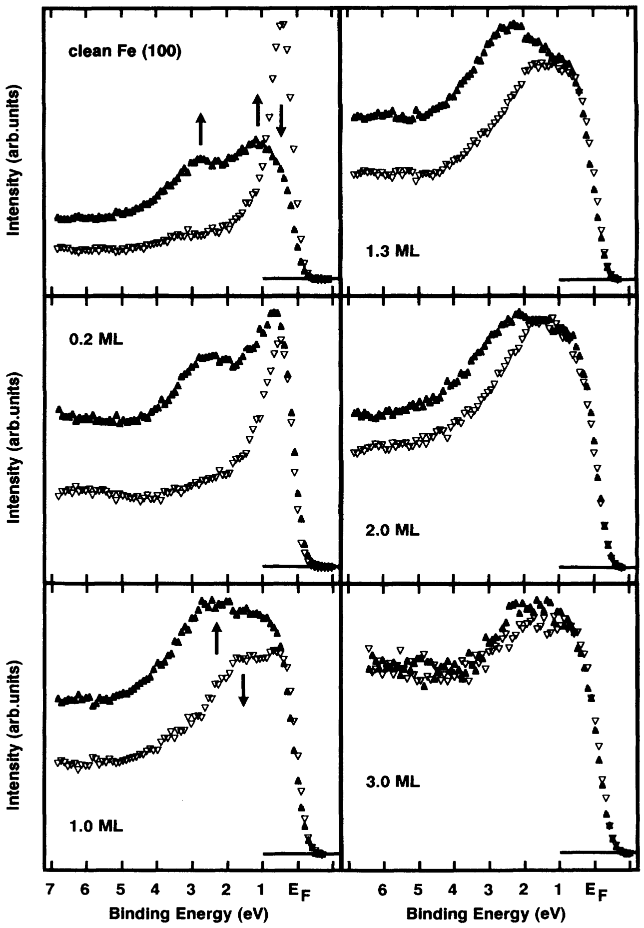

FIG. 2. Spin-resolved EDC's for Rh overlayers of various thicknesses (as indicated) on Fe(100). △, majority spin; ▽, minority spin. $h\nu = 45$ eV, normal emission.

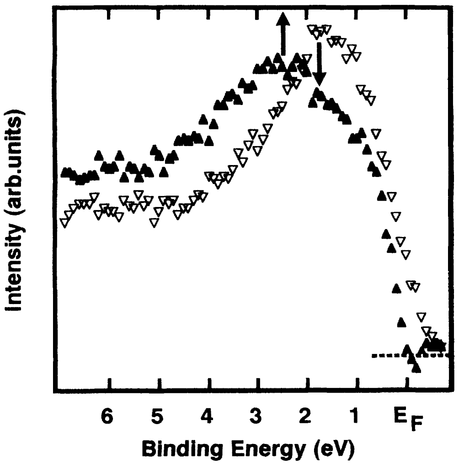

FIG. 3. Spin-dependent difference curve: 1.3–0.5-ML Rh/Fe(100) (see text).

very similar to the results from clean Fe. The important point here is that next to the Fe $3p$ the Rh $4p$ emission also shows a signficant dependence on the photoelectron spin. The line shapes and peak positions of the Rh $4p$ level are different for the two spin characters, with the same tendencies as in the Fe $3p$ emission. This indicates the ferromagnetic order within the overlayer and the parallel coupling of the Rh and the Fe moments.

An important aspect of this work is the driving mechanism for the magnetic order of the Rh overlayer. The theoretically determined magnetic moment in the Rh overlayer is so large that two distinct reasons for its occurrence might be assumed. One is the reduced dimensionality of the system which causes a strongly reduced Rh-Rh coordination compared to the bulk so that the average Rh-Rh hybridization is small. This leads to a band narrowing and, in special cases, to an increased density of states at E_F. Thus, the system might tend to approach or even exceed the limit of the Stoner criterion for magnetic order. This idea is similar to what is expected in certain bulk samples for lattice expansion. Another possible origin for magnetic order in these layers is the direct hybridization between the overlayer and the exchange-split valence states of a magnetic substrate.

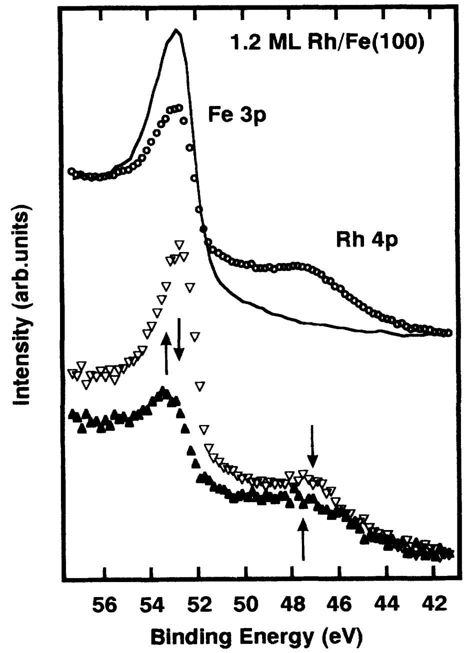

FIG. 4. Spin-resolved Fe $3p$ and Rh $4p$ core levels from 1.2±0.2 ML Rh/Fe(100) at $h\nu=120$ eV. ——, clean Fe reference; ○, spin summed; △, majority spin; ▽, minority spin (spin-resolved data after substraction of a constant background).

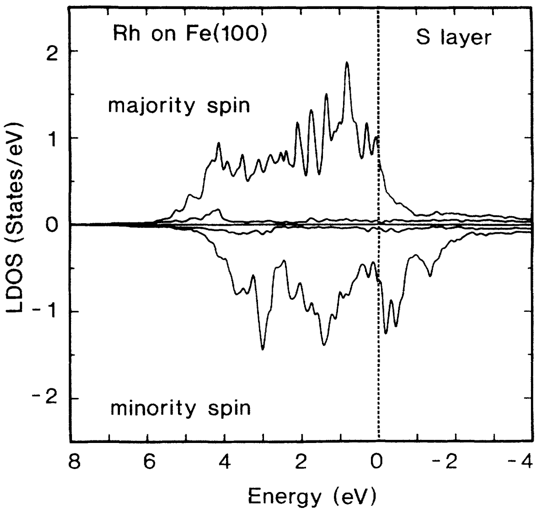

FIG. 5. Local density of states (LDOS) including small $s+p+f$ contributions for 1-ML Rh/Fe(100).

To investigate this point we show in Fig. 5 the results of our FLAPW calculation of the system. In the figure the calculated layer projected density of states (LDOS) of an ordered Rh monolayer on Fe(100) is drawn. In agreement with our experimental results the theory predicts the existence of a ferromagnetic Rh overlayer on Fe(100). The magnetic moment per Rh site is found to be $0.82\mu_B$. In the calculation we find a rather strong hybridization between the Rh overlayer and the Fe substrate. This is clear when comparing our data to those for a Rh layer on Ag(100).[3] There the Rh LDOS is significantly narrower than in this case, where a total bandwidth of about 4–5 eV is found. The average exchange splitting of the main features in our calculation is of the order of 1 eV, close to the value of the spin splitting of the main Rh $4d$ peaks in our spin-resolved EDC's (0.7±0.2 eV). A more detailed comparison between the experimental and the theoretical results is, however, not possible. The calculation cannot model the unordered overlayer and it does not contain the symmetry of the bands (selection rules) as well as the excitation cross sections of the different states.

Strong spin-dependent electronic interactions as in this case can induce magnetic moments into the overlayer. The measurement of a relatively large spin polarization also for 2-ML overlayer thickness at room temperature is an indication for the hybridization as the main origin of the magnetic order. The dimensionality of the system should not play the main role because otherwise one would expect the ferromagnetic order of the system to be more strongly reduced after evaporation of the second layer.[11] Also the fact that Rh monolayers on Au(100) (Refs. 18 and 19) have not been found to order ferromagnetically at room temperature supports the importance of the *d-d* hybridization in this case. Thus, we are able to derive a picture in which the ferromagnetic order of the Rh monolayer is mainly determined by the direct hybridization between overlayer and substrate. The interface Rh layer is ferromagnetic and presumably remains ferromagnetic when further layers are deposited. However, the influence of the substrate decays rapidly with the distance and the emission from further layers has little or no polarization.

In conclusion, we have shown that Rh monolayers grown layer by layer but not epitaxially on Fe(100) are ferromagnetic. The Rh atoms carry a magnetic moment which is coupled parallel to the magnetic moments of the Fe substrate. The evidence for the ferromagnetic order has been given by spin-resolved valence band and core-level photoelectron spectroscopy. The valence-band spectra are in reasonable qualitative agreement with our results of a FLAPW calculation for 1 ordered ML Rh/Fe(100). We have shown that the magnetic order of the overlayer is mainly driven by the hybridization of the Rh 4*d* states with the 3*d* states of the substrate.

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