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Short-period oscillations in photoemission from thin films of Cr(100)

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Angle-resolved photoemission (PE) study of thin films of Cr grown on Fe(100) reveals thickness-dependent short-period oscillations of the PE intensity close to the Fermi energy at $\mathbf{k}_{\parallel} \sim 0$. The oscillations are assigned to quantum-well states (QWS) caused by the nesting between the Fermi-surface sheets around the Γ and the X points in the Brillouin zone of antiferromagnetic Cr. The experimental data are confirmed by density-functional calculations applying a screened Korringa-Kohn-Rostoker Green's function method. The period of the experimentally observed QWS oscillations amounts to about 2.6 monolayers and is larger than the fundamental 2-monolayer period of antiferromagnetic coupling in Cr.

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Thickness-dependent oscillations of magnetic coupling in multilayer structures depend both on the electronic structure of the spacer materials and on conditions at the interfaces.[1–3] It was found that in case of nonmagnetic spacers both the Rudermann-Kittel-Kasuya-Yoshida[4,5] and polarized quantum-well states[6–8] (QWS) models can describe certain aspects of the experimental findings.

The situation is more complicated for magnetic spacers, e.g., Cr films in Fe/Cr/Fe(100). This intensely investigated system shows giant magnetoresistance and spin-valve effects and is applied in magnetic sensor technology.[9] Below the bulk Néel temperature, T_N=311 K, Cr reveals antiferromagnetic (AF) order [often referred to as the commensurate spin-density wave (SDW)] that is modulated by an incommensurate SDW along the ⟨100⟩ directions.[10–12] The ferromagnetic or AF type of coupling between Fe layers in Fe/Cr/Fe(100) varies with the thickness of Cr spacer following a short period [2 monolayers (ML)], whereas the strength of the coupling is modulated by a long period of ~11 ML, about half a wavelength of the incommensurate SDW.[13–16]

In their study, Schilfgaarde and Harrison[17] suggested that the incommensurate long-period modulation stems from aliasing of the short-period oscillations due to a slight mismatch between the nesting vector **k** spanning the Fermi-surface (FS) sheets around the Γ and the H points in the Brillouin zone (BZ) of paramagnetic Cr and the period of the reciprocal lattice in the ⟨100⟩ directions. This model, however, is not supported by the experimental results obtained by scanning electron microscopy with polarization analysis.[15] On the other hand, it was shown that the long-period oscillations can be related to QWS corresponding to nesting conditions with smaller **k** vectors spanning other sheets of the Fermi surface.[18,19]

So far, quantum-well states related to the short-period oscillations were only observed in one specific direction off normal to the surface of thin films of antiferromagnetic Cr (*sc* lattice of CsCl-type) grown on Fe(100).[19] Previous spectroscopic attempts to monitor the normal-emission QWS with a correspondingly large **k** vector did not yield decisive results. It was not clear by now to which extent these short-period QWS with $\mathbf{k}_{\parallel}=0$ can actually form in the system considered.

In this paper we report on thin layers of antiferromagnetic Cr epitaxially grown on a Fe(100) buffer film, studied with angle-resolved photoemission (PE) and analyzed by means of density functional calculations. Short-period oscillations of spin-integrated normal-emission PE intensity close to the Fermi energy (E_F) originating from QWS have been observed upon thickness variation. The period of these oscillations is larger than the period of the previously observed off-normal QWS.[19] It can only be explained by nesting features of the Fermi surface of antiferromagnetic Cr that are not present in the unfolded FS of nonmagnetic Cr. In contrast, both the AF ordering and the off-normal QWS have their origin in the same nesting regions that are already obvious in the Fermi surface of nonmagnetic Cr.[10] It is concluded that QWS with $\mathbf{k}_{\parallel}$ close to zero provide only a minor contribution to the short-period magnetic coupling of the Fe layers in Fe/Cr/Fe(100). The main contribution stems from the AF ordering of neighboring (100) layers of Cr that leads to strong modulations of the layer- and spin-resolved electron density of states at E_F.

The regime of the short-period oscillations was studied for Cr films of thicknesses up to 10 ML, where the long-period modulation, which might complicate the analysis, is expected to be suppressed.[20] The samples were prepared using layer-by-layer thermal deposition of Cr onto a 60-Å-thick Fe(100) film grown on a W(100) substrate followed by slight annealing at 400 K. The thicknesses of the deposited layers were monitored with quartz microbalances with an accuracy of 20%. The crystalline order of the samples was checked by low-energy electron diffraction. In order to distinguish between contributions into the normal-emission PE signal from the Cr- and the Fe-derived valence states, the spectra were acquired directly below the $3p$-$3d$ excitation threshold of Fe (the Fano antiresonance, $h\nu$=54 eV), where the Fe $3d$ PE intensity is strongly suppressed. The experiments were performed at the Russian-German beamline at BESSY II (Ref. 21) using an electron-energy analyzer CLAM4 with an angular resolution of 1°. The overall system energy resolution

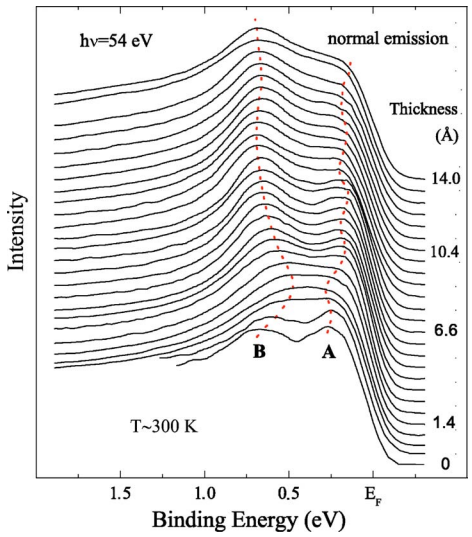

FIG. 1. (Color online) Normal-emission PE spectra of Cr/Fe/W(100) taken at various Cr thicknesses. The dashed lines are guides to the eye to follow the BE changes of the features, which were obtained by a least-squares fit of the spectra.

was set to 130 meV (full width at half maximum). All measurements were carried out at room temperature well below T_N at the surface of Cr (Ref. 22) with a base pressure in the low 10^{-9}-Pa range.

Angle-resolved PE spectra acquired from the Cr/Fe/W structure in normal-emission geometry are shown in Fig. 1. There are basically two features (A and B), which vary their binding energies (BE) with Cr coverage. Since structure A is located close to the Fermi energy, it causes variations of the PE intensity at E_F. These binding energy and corresponding intensity variations have a period of ~2.6 ML (Fig. 2), which is larger than the short period of the magnetic coupling oscillations in Fe/Cr/Fe(100). To study the origin of

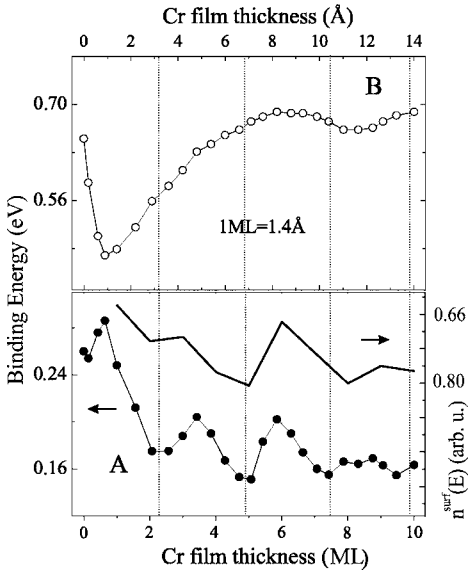

FIG. 2. Binding energies of features A and B as function of Cr deposition. Dotted vertical lines mark a 2.6-ML period of oscillations. The variation of the surface local density of states at the Fermi energy ($\mathbf{k}_\parallel \sim 0$) is shown in the lower panel.

feature A, normal-emission PE experiments with photon-energy tuning in the range from 40 to 90 eV were performed for 5-ML Cr coverage. Since no dispersion of feature A was monitored in this way, a two-dimensional (2D) character of this structure has to be concluded. Feature A is, however, not a surface state, because it shows oscillating behavior with Cr thickness. The binding energy of peak B reveals a nonmonotonic behavior on the Cr thickness as well. For very low Cr coverages it first drops and then increases again unless the thickness amounts to 6 ML. At 8 ML it shows another minimum.

To understand the above thickness-dependent variations observed in PE experiments performed without spin resolution, the electronic structure of the system was calculated self-consistently in the framework of the local spin-density approximation (Vosko-Wilk-Nusair parametrization) using a screened Korringa-Kohn-Rostoker Green's function method.[23] The Cr-covered surface was modeled by a free *bcc*-like Fe slab of 10 ML thickness covered on both sides with n layers of Cr (n from 1 to 10). Cr is put on both sides of the Fe slab in order to benefit from inversion symmetry. This is a purely technical point that is quite usual in slab calculations. We have checked that it does not influence the results by convergence tests on the Fe layer thickness. The experimental lattice constant for Cr was taken without accounting for surface relaxation. For the potentials the atomic sphere approximation was used; nevertheless, the charge density was expanded in spherical harmonics up to an angular momentum of $l_{\max}=6$. This ensures a proper treatment of the charge relaxation at the surface. To elucidate the origin of the PE spectra the spin-dependent local density of states (LDOS) of the surface Cr layer was calculated using the site diagonal part of the Green's function. Due to the 2D periodicity of the system the in-plane wave vector $\mathbf{k}_\parallel$ is a good quantum number. Real space properties are obtained by a Fourier transformation and integration over the 2D surface Brillouin zone. The obtained LDOS was broadened with a Gaussian of 130-meV width according to the finite experimental energy resolution.

Results for the spin-dependent LDOS of the topmost Cr layer for thicknesses of 1 and 2 ML are presented in Fig. 3. The interface coupling of Cr with Fe is antiferromagnetic, and also the successive Cr layers carry moments of opposite sign. In the following the terms spin up and spin down will be used with respect to the Fe, where spin up is defined as the majority spin. If Fe(100) is covered by 1 ML of Cr, this layer carries a large spin moment of $-3.22\mu_B$ per atom. The Cr $3d$-spin-up band is situated above E_F and thus only marginally hybridized with the Fe $3d$-spin-up band (not shown), that is, almost completely occupied. As a result, the Cr $3d$-spin-up band is quite narrow and has a steep slope just above E_F, stabilizing the large moment. In turn, the moment yields a large spin split of the Cr bands and the Fermi level is placed close to a peak of the almost occupied spin-down band (feature C). The peak gives rise to a high value of the spin-down LDOS at the Fermi energy. If Fe(100) is covered by 2 ML of Cr, the unoccupied $3d$-spin-down band of the surface layer is broader than the unoccupied Cr $3d$-spin-up band in the previous case, since now a more pronounced hybridization with the subsurface Cr $3d$ states takes place.

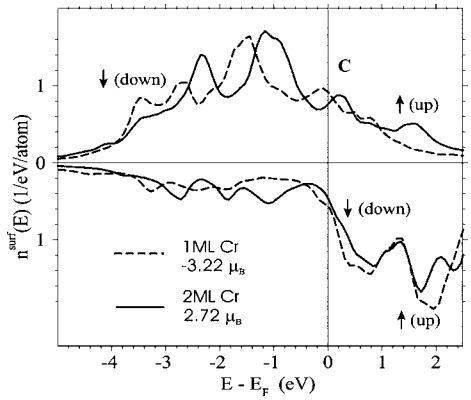

FIG. 3. Spin-dependent LDOS at the topmost Cr layer for 1-ML (dashed) and 2-ML (solid) Cr coverages The spin directions are defined with respect to the Fe substrate (arrows). The upper (lower) panel shows the local majority (minority) spin projections.

The result is a less steep slope above E_F leading to a reduced moment of $2.72\mu_B$, a related smaller spin split, and a shift of the local majority peak *C* above the Fermi level. Hence, the total (spin-up plus spin-down) LDOS at E_F becomes smaller as shown in the left inset in Fig. 4 (dashed).

Further increase of the Cr thickness causes a stabilization of the surface magnetic moment at an absolute value of $2.47\mu_B$. This is the consequence of the saturation of the discussed hybridization phenomena. The obtained value is in good agreement with previous calculations.[24,25] Correspondingly the BE of peak *C* does not change anymore and separate constant-amplitude oscillations of the spin-up and the spin-down LDOS with a period of exactly 2 ML are monitored (not shown), as a result of AF arrangement of the successive monolayers. These modulations are due to the nesting conditions of the Fermi surface of nonmagnetic Cr that cause the AF order of bulk Cr and also the off-normal QWS measured by Li *et al.*[19] They are further responsible for the 2-ML short-period oscillations of the interlayer coupling between adjacent Fe layers in Fe/Cr/Fe structures. Note that the total surface LDOS at E_F varies only marginally at Cr coverages higher than 4 ML (left inset in Fig. 4).

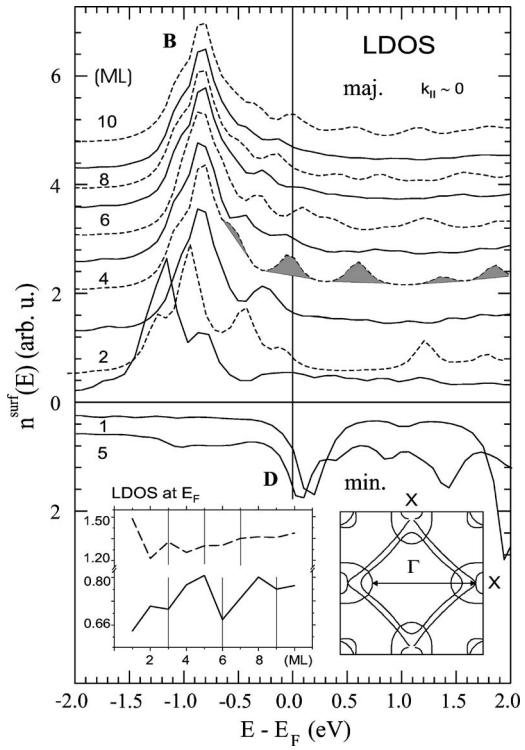

FIG. 4. Spin-majority surface LDOS ($\mathbf{k}_\parallel \sim 0$) for 1 to 10 ML of Cr on Fe/W(100). The solid and dashed lines are used for odd and even numbers of ML. QWS for 4-ML coverage are shaded. The spin-minority surface LDOS for 1 and 5 ML of Cr are shown in the bottom. The left inset represents oscillations of the **k**-integrated (dashed) and **k**-resolved (solid) surface LDOS at E_F. The right inset demonstrates a calculated FS cut within the BZ of AF Cr. The solid line with arrows denotes a nesting vector ($\mathbf{k}_\parallel \sim 0$).

Therefore these spin-resolved **k**-integrated LDOS oscillations cannot explain the behavior of our spin-integrated experimental spectra close to the Fermi energy (feature *A*). In contrast, the angle-resolved PE data taken in the normal-emission geometry should be compared to results of LDOS calculations restricted to the **k** region close to $\mathbf{k}_\parallel = 0$. To account for the finite angular resolution of 1°, the integration of the LDOS was restricted to about 0.4% of the surface Brillouin zone. The corresponding **k**-resolved spin-majority LDOS results for the topmost Cr layers in the cases of 1- to 10-ML Cr systems are shown in Fig. 4 (top). The related spin-minority LDOS for 1 and 5 ML of Cr is presented in the bottom of this figure. To analyze the character of the electron eigenstates, probability amplitudes of the states in the center of the Brillouin zone were calculated. For the states close to the Fermi level we find a strong confinement effect; that means all states are localized either inside the Fe or the Cr layer. Two types of states mainly concentrated in the Cr layer can be distinguished: surface and quantum-well states. In contrast to the surface state, which is always located close to E_F (feature *D* in the spin-minority LDOS), the QWS (shaded for 4-ML coverage) change their binding energies considerably with increasing Cr coverage. Unlike the simpler *sp*-derived QWS, which move monotonic in energy as a function of film thickness due to the paraboliclike dispersion of the corresponding bulk bands,[6,7,26] the observed *d* originating QWS change their binding energies not always monotonic. The important observation is, however, that the period of Fermi-energy crossings by the QWS, which is displayed in the spin-integrated **k**-resolved LDOS as variations of the intensity at E_F (left inset in Fig. 4, solid line), is very close to the period observed in our PE experiment. This correspondence is made explicit in Fig. 2 (lower panel), where the inverted LDOS data are compared with the experiment. The Fermi-energy LDOS oscillations obtained upon integration over 0.4% of the surface Brillouin zone can be related to Fermi-surface nesting conditions. The Fermi surface of antiferromagnetic Cr calculated in the present study (Fig. 4, right inset) differs from the Fermi surface of nonmagnetic Cr usually considered in the literature[10,17,18] not only in a trivial folding but also in a slight distortion of the sheets around the *X* point. This causes an additional nesting, depicted in Fig. 4, with a longer real-space period of 2.8 ML. Nesting with such a period is not present in nonmagnetic Cr. In the explicit slab calculation we find $\mathbf{k}_\parallel$-resolved LDOS oscillations of about this period.

We assign the PE intensity oscillations of peak *A* in our data to the QWS behavior. The obtained theoretical 3-ML period of oscillations (Fig. 4, left inset) lies within the error bars of the experimental period. Nonmonotonic maximum-to-minimum intensity variations of the Fermi-energy signal can be explained by contribution of the surface state *D* that moves continuously toward E_F with Cr coverage (see the 1-ML and 5-ML minority LDOS in Fig. 4). Our study is a clear observation of short-period QWS with $\mathbf{k}_{\parallel} \sim 0$ in $\langle 100 \rangle$ directions in thin films of Cr. It is anticipated that due to the larger period of oscillations these QWS do not significantly contribute to the 2-ML antiferromagnetic coupling in Fe/Cr/Fe systems.

Feature *B* in Fig. 1 can be assigned to the main LDOS peak (Fig. 4), which is located at about 1.2 eV BE for 1 ML of Cr on Fe(100). As obtained from our calculations, the binding energy of this peak follows changes of the magnetic moment of Cr. With decrease of the latter, peak *B* shifts toward the Fermi energy. For the 2-ML system, it is found at 0.90 eV. For coverages of more than 3 ML the position of peak *B* at 0.80 eV does not change anymore. So the energy location of feature *B* can be used as a measure of the magnetic moment of the surface Cr layer. The strong decrease of the BE of this feature at very low Cr coverages seen in Fig. 1 can be explained by intermixing at room temperature of Fe and Cr at the Cr/Fe(100) interface observed also by other experimental techniques.[27] The magnetic moment at the intermixed interface is strongly reduced, since neighboring Fe and Cr atoms tend to align their magnetic moments antiparallel to each other. Upon further Cr deposition, the relative concentration of Fe in the surface region decreases and the surface magnetic moment increases. The BE minimum of peak *B* observed at about 8 ML coverage might be associated with a constitution of the incommensurate SDW state, which is expected to appear in this coverage range.[20]

To summarize, we have found quantum-well states in thin antiferromagnetic Cr films on Fe(100) causing oscillations of normal-emission PE intensity and related projected density of states with a period significantly larger than the common antiferromagnetic 2-monolayer period.

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