Spin Density Waves in Thin Chromium Films

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The magnetic profile of $Fe/Cr_N/Fe$ bcc(001) films has been calculated by means of first principles density functional theory. It is shown that the magnetic profile of the chromium spacer can be expressed in terms of spin density waves (SDW). The dispersion and amplitude of the SDW are determined and the effects from the finite film thickness are observed and discussed. It is found that the SDW wave vectors are quantized and that for certain Cr thicknesses two SDWs with different wavelengths coexist. Connections to the magnetic interlayer coupling are discussed. [S0031-9007(99)09241-8]

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Chromium metal exhibits a great number of complex magnetic phenomena and as the archetype of a spin density wave (SDW) it has been intensively studied [1]. The SDW in bulk chromium is generally accepted as a typical example of how the electronic structure and the topology of the Fermi surface may influence magnetism in itinerant systems. However, the richness of the phenomena is even more manifested in the properties of Fe/Cr films where one finds effects like oscillatory magnetic interlayer coupling [2], giant magnetoresistance (GMR) [3], and noncollinear exchange coupling [4,5]. Thus it is of great importance to investigate the nature of the magnetic structure of chromium in layered systems, where several experiments have confirmed that a SDW is formed already for relatively thin Cr films [6–10].

The properties of Fe/Cr systems have recently also attracted a lot of theoretical attention, not least in connection to the magnetic interlayer exchange coupling (IXC) and GMR effects; see, e.g., Refs. [11–15]. However, hitherto theoretical first principles studies have not been very successful in reproducing the SDW character of the magnetism in Cr, due to computational difficulties with the large SDW unit cell size together with the weak energy dependence on its periodicity. In fact, most studies on the IXC of Fe/Cr/Fe were based on RKKY-like schemes, assuming the spacer layers being nonmagnetic [13,14]. For the same reason calculations for antiferromagnetic bulk chromium were for a long time limited to the commensurate state with two atoms per unit cell [16]. Only very recently, calculations were successfully performed for a realistic long wavelength bulk SDW [17].

In the present Letter we present self-consistent first principles electronic structure calculations of the layered resolved spin moments in $Fe/Cr_N/Fe$ bcc(001) films consisting of up to 52 Cr atomic layers. In this thickness range it is possible to safely observe a full period of the SDW, which in bulk chromium has about 21 monolayers (ML) between each node. Of major interest is the mechanism behind the formation of the SDW. In bulk Cr the SDW can be ascribed to nesting between parallel sheets of the paramagnetic Fermi surface [18], which gives rise to a peak in the **q**-dependent spin susceptibility at the nesting wave vector. The energy is lowered by forming the SDW due to the opening of a partial band gap at the Fermi energy. However, in a layered system where the perpendicular symmetry is broken it is not clear that the same mechanism is appropriate in describing the SDW stabilization. Of special interest is also the influence of the magnetic interfaces at the boundaries of the Cr film. These proximity effects originating from the ferromagnetic Fe films have recently attracted much attention. Especially the range of the proximity is of importance for the interfacial influence on the SDW.

The magnetic structure has been calculated self-consistently within the framework of density functional theory [19,20] in the local spin density approximation (LSDA) [21,22]. The calculational scheme is based on the linearized muffin-tin-orbital method [23] within a Green's function technique for surfaces and interfaces [24]. This approach has the advantage that it can handle semi-infinite systems with a broken perpendicular translational symmetry, i.e., it does not rely on a slab or supercell geometry. The interface systems investigated in the present work consist of chromium films embedded between two semi-infinite iron crystals. All interface calculations were done at the Cr bcc lattice constant, i.e., no relaxations were taken into account. A mesh of 36 special k-points was used in the irreducible part of the 2-dimensional Brillouin zone (2DBZ).

The calculated magnetic profile $M_N^\alpha(n)$ of the $Fe/Cr_N/Fe$ bcc(001) film oscillates with the layer position n within the chromium film with a period close to 2 ML as is seen in the example displayed in the inset of Fig. 1. This is very similar to what is found in bulk chromium and shows many characteristics of a SDW. We find that the Cr magnetic moments for an alignment $\alpha = (\mathrm{F}, \mathrm{AF})$ [ferromagnetic (F) or antiferromagnetic (AF)] of the Fe layers are generally well described by

$$M_N^\alpha(n) = \sum_i A_{N,i}^\alpha \sin(q_{N,i}^\alpha \pi n + \phi_{N,i}^\alpha), \qquad (1)$$

i.e., as a superposition of sinusoidal oscillations with wave vectors $q_{N,i}^\alpha$, amplitudes $A_{N,i}^\alpha$, and phases $\phi_{N,i}^\alpha$. Here and below we choose the propagation direction of the SDW to

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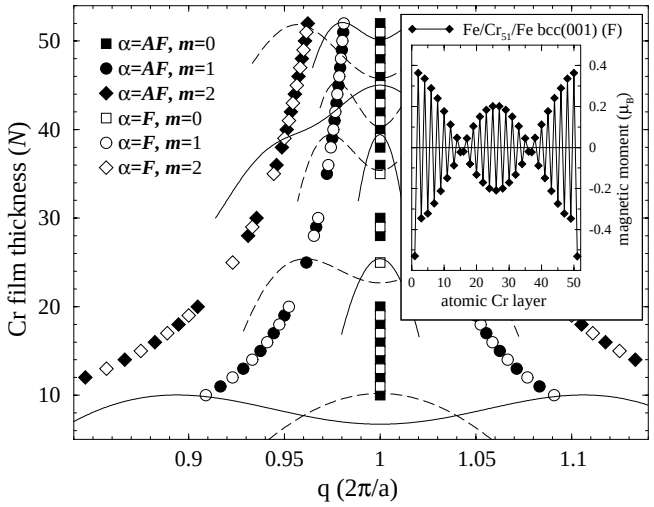

FIG. 1. The Fourier transformed magnetic moment distribution for different Cr film thicknesses for F (solid line) and AF (dashed line) alignment of the Fe layers. The scale is shifted as to align the maxima of the curves with the corresponding thickness. The SDW dispersion follows different branches (symbols) which are determined by the simple dispersion rules (see text). The inset shows the magnetic moment distribution for a 51 ML thick F film.

be perpendicular to the (001) plane so that $q^{\alpha}_{N,i}$ is the out of plane component in units of the cubic reciprocal lattice vector.

When fitted to the results of the full calculations, the dispersion, i.e., $q^{\alpha}_{N,i}$ as a function of chromium thickness N, is found to follow distinct branches as indicated in Fig. 1, which also shows some Fourier amplitudes of the magnetization profiles. Within each branch we find that the boundary conditions vary from F to AF for every second atomic layer thickness. The appearance of these branches is a direct consequence of the finite size of the chromium film. Generally, for a Cr film with thickness N the magnetic moment distribution can be expressed as a sine Fourier series [Eq. (1)] with N discrete wave vector components $q^{\alpha}_{N,i} = B^{\alpha}_i/(N+1)$. For a F (AF) magnetic alignment of the Fe layers the magnetic moment profile is even (odd) with respect to mirroring through the center of the Cr spacer layer, which leads to $B^{\alpha}_i = p^{\alpha}_i - 2\phi^{\alpha}_i/\pi$, where p^{α}_i is an odd (even) integer. The phase will usually depend on the boundary conditions at the Fe interfaces, but in order to allow for a commensurate AF SDW ϕ^{α}_i has to take the value $\pi/2$. Our fits always give a value close to this.

Thus, instead of a continuous spectrum as in the bulk, the dispersion of the SDW is quantized in different branches given by the expression $q^{\alpha}_{N,i} = 1 \pm m^{\alpha}_i/(N+1)$ in the vicinity of the commensurate AF ordering ($q = 1$), where m^{α}_i is an even integer in case of $\alpha =$ F and odd N or $\alpha =$ AF and even N, and corresponds to the number of nodes of the envelope function. This simple relation is found to give an almost perfect description of the results from the full calculations except for the single outermost Cr layers closest to the Fe interfaces. The maximal absolute error of the moment at any inner layer is smaller than $0.05\mu_B$ and the 2-norm of the fitting error $\|\Delta M^{\alpha}_N(n)\|_2/N < 0.01A^{\alpha}_N$. We may thus conclude that the magnetic profile of thin Cr films indeed can be interpreted in terms of SDW. Moreover, the range of proximity effects from the Fe interfaces is limited to the interface Cr atomic layer, i.e., only the moment of this Cr layer deviates substantially from the behavior described by the SDW of the Cr film. However, the SDW as a whole is very sensitive to the boundary conditions set up by the magnetic Fe layers, as will be discussed below.

In Fig. 2 the extracted amplitudes $A^{\alpha}_{N,i}$ are shown for the different film thicknesses N. In the present thickness range there exist three different branches of i which contribute to the SDW and which correspond to $m^{\alpha}_i = 0, 1, 2$. From these results several interesting features can be observed.

(i) The amplitude of the branch corresponding to a commensurate antiferromagnetic SDW ($m^{\alpha}_i = 0$), is almost constant until about 40 ML of chromium where it decays rapidly. This is in good agreement with experiments which find commensurate SDWs in very thin Cr films [7]. When the film thickness is below 30 ML the SDW amplitude of the magnetic ordering corresponding to $m^{\alpha} = 1$ is suppressed compared to its antiferromagnetic value, at 10 ML thickness almost by a factor of 2. In case of a single monolayer of Cr the moment actually vanishes in the AF case due to the symmetry. Altering the magnetic alignment and thereby changing the periodicity of the SDW may thus strongly suppress or enhance the magnetic amplitude.

(ii) When the amplitude of the $m^{\alpha}_i = 0$ branch decreases the branch $m^{\alpha}_i = 2$ increases in amplitude, and there is a

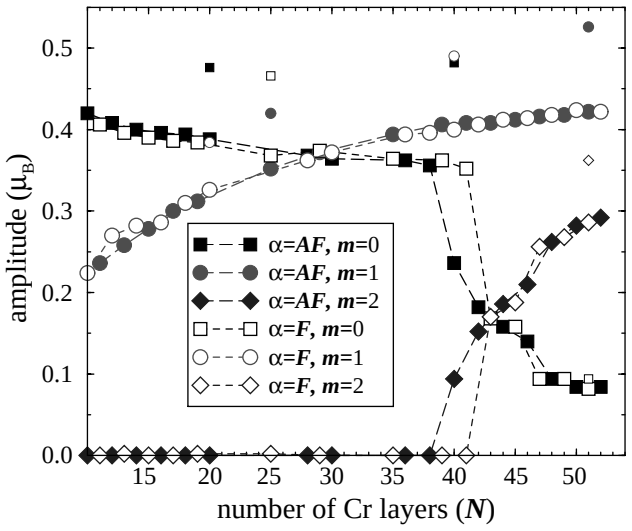

FIG. 2. The amplitudes of the individual branches obtained by fitting the calculated moment profiles. The smaller symbols indicate the corresponding amplitudes for calculations using the von Barth–Hedin form of LSDA functional, instead of the Vosko-Wilk-Nusair parametrization.

jump from one branch to another. For a finite thickness range around the branch jumps, two SDWs are found to coexist. Splitting of the SDW peaks have been observed in neutron scattering experiments for thin Cr layers in a Fe/Cr superlattice, [8] where it however was interpreted in terms of an interaction between SDW in neighboring Cr spacer layers rather than as due to the finite thickness of one Cr layer, as is clearly the reason in our present calculations. This splitting is illustrated by the Fourier amplitudes of the 45-ML-thick Cr F film in Fig. 1. In case of finite temperatures the relative distribution between different harmonics may change and even multiple SDWs may coexist.

(iii) The shift in wave vector $\Delta q_N^\alpha = 2/(N+1)$ at the branch jump is twice the difference in the q vector between F and AF alignment, i.e., $|q^{\mathrm{F}} - q^{\mathrm{AF}}| = 1/(N+1)$. This is again a direct consequence of the finite set of possible wave vectors.

(iv) Because of the restriction of possible wave vectors, the dispersion of the SDW is determined by the symmetry of the boundary condition rather than by the nesting of the spacer material, in contrast to bulk chromium. However, in the limit of thick films Cr wants to have a SDW with a wave vector as close as possible to the bulk value $q_0 = 0.95$ (in units of the reciprocal lattice vector). This leads to branch jumps each 20th ML, i.e., the asymptotic periodicity of the branch jumps is determined by the nesting of the spacer material. A similar effect has been observed within a model calculation [25].

(v) In Fig. 2 one can see that the amplitude is not fully converged with the film thicknesses used in the calculations. Except for the commensurate SDW, the amplitudes are monotonically increasing. This, in combination with branch jumps, makes it hard to directly compare with the experimental bulk value of $0.6\mu_{\mathrm{B}}$. An extrapolation of the amplitudes to thick films seems to lead to a too small value. However, it is found that the amplitude is a very sensitive quantity. This can be observed by changes in the lattice constant and by the dependence on the specific LSDA functional used in the calculations. The latter is demonstrated by comparing two different LSDA parametrizations [21] and [22]. The amplitude is found to increase by about 20% with the von Barth–Hedin instead of the Vosko-Wilk-Nusair functional form as shown in Fig. 2. A similar effect is found when the lattice constant is increased by 1%.

(vi) With the presence of a Cr SDW the RKKY-like theories [26] for the IXC assuming a nonmagnetic Cr layer are not valid, [15] and the short wavelength oscillation is instead due to the SDW. In fact, as we will notice below, the SDW introduces gaps at the nesting parts of the nonmagnetic bulk Fermi surface, so the RKKY and SDW pictures for the short wavelength oscillations are mutually exclusive. Since it is not the scope of the present study, our calculations are not brought to the accuracy needed to resolve the energy difference between the F and AF alignment of the Fe layers, i.e., to obtain the IXC energy. However, the SDW contribution to the magnetic interlayer coupling can be estimated in the limit of thick films. In this case the q_N^α value of the Cr film will deviate only slightly from the bulk value q_0 and then the SDW energy per atom E_N^α should be quadratic in $q_N^\alpha - q_0$. This gives a SDW contribution to the magnetic interlayer coupling proportional to $(q_N^{\mathrm{AF}} - q_0)^2 - (q_N^{\mathrm{F}} - q_0)^2$. Inserting the simple dispersion relation from above, $q_N^\alpha = 1 \pm m^\alpha/(N+1)$, and including branch jumps in order to minimize the energy, it is found that the total SDW contribution to the interlayer exchange coupling J can be written as

$$J = N(E_N^{\mathrm{AF}} - E_N^{\mathrm{F}}) \propto (-1)^{N+1} \frac{NF(N,\delta_0)}{(N+1)^2}, \quad (2)$$

where $\delta_0 = 1 - q_0$ is the incommensurability of the bulk SDW, and $F(N,\delta_0)$ is a periodic function of N with the period $2/\delta_0$. In the interval $0 < N + 1 < 2/\delta_0$,

$$F(N,\delta_0) = 2|1 - (N+1)\delta_0| - 1. \quad (3)$$

The first factor in Eq. (2) gives rise to the 2 ML oscillation while $F(N,\delta_0)$ contributes with a, due to branch jumps, sawtooth shaped function, with a node each $1/\delta_0$ atomic layer. Thus the SDW contribution to the IXC oscillates with a short period of 2 ML superimposed by a long period of 20 ML between each node or phase slip, and with an amplitude inversely proportional to the thickness of the film. This periodicity is also in full agreement with experimental findings [6]. In conclusion, it is the strong influence of the Fe interfaces on the Cr SDW as a whole which mediates the long range IXC. However, this simple estimate of the IXC assumes only one SDW for each alignment. For thicker films the boundary effects can be diminished by allowing for a distribution of wave vectors in the SDW, in order to better adjust to the proximity effects from the Fe interfaces. Moreover, the estimate of the coupling energy does not include the influence of a thickness dependent amplitude of the magnetization.

(vii) As mentioned above, the phase of the SDW is found to be almost constant, $\phi_{N,i}^\alpha \approx \pi/2$, which means that the SDW adjusts itself as to maximize the interface Cr moment. This is in striking contrast with experiments where the SDW prefers to have a node at the Fe interface [8]. This disagreement is very likely due to the imperfect interfaces in the experimental samples. This leads to frustrations of the Cr interface atoms, which prefer an antiparallel alignment to the Fe moments, which is minimized by the interface node of the SDW. However, for the perfect interface case the energy is instead optimized by an interface SDW belly due to the enhanced Cr moments at the Fe interface. For very thin Cr films, where experiments observe a commensurate SDW and hence a SDW belly at the interface, another mechanism is responsible to escape the interfacial frustration [7,27]. The SDW is found to develop a noncollinear spiral form.

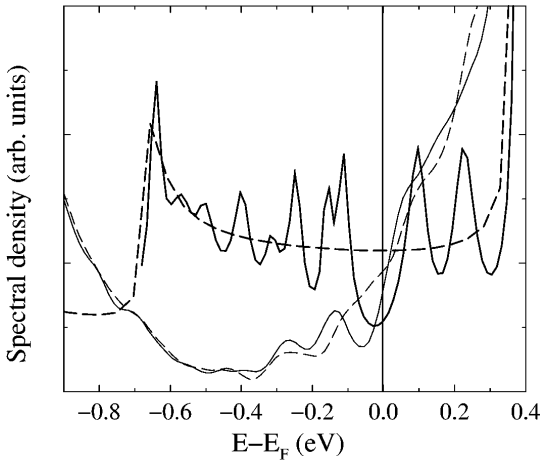

FIG. 3. The $D(\mathbf{k}_\parallel, E)$ calculated at $\mathbf{k}_\parallel = (0, 0.25)$ for nonmagnetic bulk Cr (dashed line) and for spin polarized Cr embedded in AF $Fe/Cr_{50}/Fe$ bcc(001) (solid line). The thin lines show the corresponding DOS.

Since we have already noticed that the Cr SDW does not have a wave vector that is equal to the nesting wave vector q_0, it is of interest to see whether the calculated layer SDWs are stabilized by a similar mechanism as the Fermi nesting in bulk. For instance, do pseudogaps form at the Fermi level E_F? In Fig. 3 the spectral function $D^\sigma(\mathbf{k}_\parallel, E) = \pi^{-1} \operatorname{Im} \operatorname{Tr} G^\sigma(\mathbf{k}_\parallel, E)$, where $G^\sigma(\mathbf{k}_\parallel, E)$ is the Green's function of spin σ, in-plane wave vector $\mathbf{k}_\parallel$, and energy E, is shown as a function of E for both a film with $N = 50$ and for nonmagnetic Cr bulk. The in-plane wave vector $\mathbf{k}_\parallel = (0, 0.25)$ (in units of the reciprocal lattice vector) is chosen as to be in the middle of the nesting bulk Cr Fermi sheets. The spectral function of the film oscillates around the one in bulk, with peaks arising from the quantum well confinement. However, around the Fermi energy there is a larger distance between these peaks with a deep intervening valley. Hence it is clear that the formation of the SDW does produce a partial gap also in the finite film case.

This gap is formed over a substantial region of the 2DBZ, as can be seen in the total density of states (DOS) in Fig. 3. The total DOS was calculated by integrating the $\mathbf{k}_\parallel$-resolved spectral function over the 2DBZ and summing over spins. The gap in the DOS is however not perfectly aligned at the Fermi energy. This is due to a combination of two effects. First, as can be seen for the bulk DOS there is a rapidly rising "background," which gives an impression of a down-shift of the gap. Second, due to the discreteness of possible wave vectors, a perfect nesting cannot be expected.

In this Letter we have calculated the SDW of thin Cr films embedded in bcc Fe (001) and have shown that the SDW dispersions are to a great extent influenced by the finite film thickness. Especially, only a discrete set of ordering wave vectors is possible for each thickness. For some thicknesses there are two coexisting SDWs of different wave vectors. With a simple expression for the SDW contribution to the interlayer exchange coupling, we note that the jumps between different dispersion branches well explain the observed phase slips in the coupling energy as a function of thickness.

We believe that this work should stimulate some new experimental work, such as, for example, a search for a systematic shift in the neutron spectra when an AF aligned sample is aligned ferromagnetically in a magnetic field, as predicted by the present Letter, or to give confirmations of the simple quantized dispersion rules for the SDW wave vectors.

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