Magnetism in Cr Thin Films on Fe(100)

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The spin polarization of secondary electrons from a Cr film on Fe(100), measured with scanning electron microscopy with polarization analysis, oscillates as a function of Cr thickness with a period near two atomic layers, consistent with incommensurate spin-density-wave antiferromagnetism in the Cr. The position of a phase slip due to incommensurability varies reversibly by 14 layers over the temperature range of 310 to 550 K. The Cr surface magnetic moment persists well above the Néel temperature of bulk Cr.

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Chromium, with its incommensurate spin-density wave (SDW), exhibits a great richness of phenomena and has therefore been the subject of intense study for many years [1]. Cr is an itinerant antiferromagnet with a bulk Néel temperature $T_N = 311$ K. The SDW is characterized by a wave vector **Q** which is determined by the nesting properties of the Cr Fermi surface. Recently, interest in the behavior of thin films of Cr has been high because Fe films separated by thin Cr layers were the first structures for which antiferromagnetic exchange coupling [2] of the Fe layers, giant spin-valve magnetoresistance [3,4], and oscillations in the exchange coupling were observed [5]. The exchange coupling of Fe layers, through epitaxial films of Cr with smooth interfaces, has been found [6,7] to undergo numerous oscillations from ferromagnetic to antiferromagnetic coupling as a function of Cr thickness with a period determined by the SDW wave vector **Q**.

We find that the surface of a thin Cr film on an Fe(100) substrate has a moment which persists to well above the bulk Néel temperature. The direction of the surface moment changes with each additional Cr layer added to the film, showing that the Cr is antiferromagnetically ordered. The antiferromagnetic order is slightly incommensurate with the lattice consistent with SDW antiferromagnetism where the SDW polarization axis is fixed by the magnetization of the Fe substrate. The incommensurability differs considerably from that found in bulk Cr and, moreover, varies strongly and reversibly with temperature in the measured range from T_N to approximately 1.8 times T_N.

In this experiment, a Cr film of linearly increasing thickness is evaporated as a very shallow wedge (wedge angle of order 0.001°) onto a single-crystal Fe whisker substrate *in situ* in a scanning electron microscope equipped with an Auger capability. Reflection high-energy electron diffraction (RHEED) was used to determine the optimum conditions for growth of the Cr film. Typically, the film was evaporated at 6–10 monolayers (ML) per minute with the Fe substrate at a temperature from 250 to 350 °C. Scanning electron microscopy with polarization analysis (SEMPA) measures the spin polarization of secondary electrons generated in the SEM thereby giving a direct measure of the net spin density or magnetization near the specimen surface. Details of the experimental setup have been reported previously [6].

In discussing the behavior observed at the surface of a Cr film with increasing thickness, it is useful to contrast the magnetic properties of the Cr against what one observes in an Fe layer deposited on top of the Cr wedge. The magnetization of a 1-nm Fe film on the Cr wedge which is grown on a Fe(100) whisker substrate to form an Fe/Cr/Fe sandwich structure is shown in Fig. 1. In this SEMPA image, white (black) corresponds to magnetization to the right (left). The Fe substrate has a domain wall running along its length which provides a useful way to verify the zero of the magnetization, i.e., the magnetization near the domain wall on one side of it is equal and opposite to that just on the other side of the wall. The short-period oscillation in the exchange coupling, which causes the magnetization to change each layer, is superposed on a long-period coupling such as has been observed for rough Cr interlayers [5,6]. In the

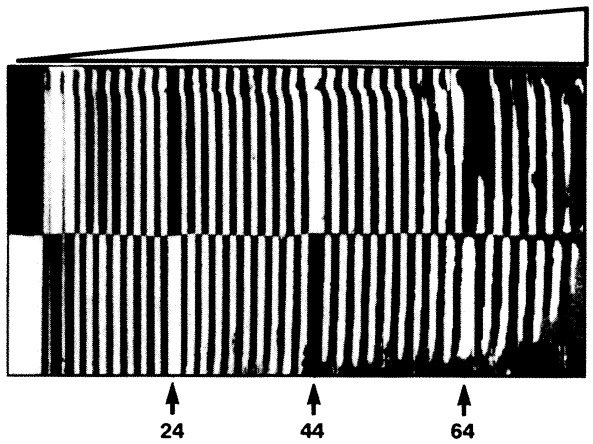

FIG. 1. SEMPA image of the magnetization in an Fe layer covering a varying thickness Cr film (wedge) grown on a Fe(100) single-crystal whisker substrate. The arrows mark the Cr interlayer thickness, in atom layers, where phase slips in the magnetization oscillations occur due to the incommensurability of the SDW. The actual area imaged is approximately 0.1 mm high × 1 mm long.

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thinner parts of the wedge, the short period oscillations dominate only after the first antiferromagnetic transition at 5 layers and initially are not symmetric about zero which leads to the black and white stripes of slightly different width in the image. Coupling is observed through over 75 layers corresponding to a Cr thickness of over 10 nm. There is a change in phase or "phase slip" apparent between layers 24 and 25, 44 and 45, and 64 and 65. Thus, just below the phase slip at 24 layers, the Fe overlayer is coupled ferromagnetically to the substrate when the number of Cr layers is even, and just above the phase slip at 25 layers it is coupled ferromagnetically when the number of Cr layers is odd. The phase slips are due to the fact that the wave vector **Q** which governs the coupling is incommensurate with the lattice wave vector.

Measurements of the bare Cr wedge, before the top layer of Fe was deposited, are shown in Figs. 2(a)–2(c). The thickness of the Cr film deposited on the ferromagnetic Fe substrate is accurately determined by the RHEED intensity oscillations shown in Fig. 2(a). The RHEED is measured under conditions where, by kinematic diffraction arguments, a maximum in the intensity corresponds to a completed Cr layer. The RHEED oscillations were used to determine the thickness scale in the figure; the interval spacings on the scale are slightly different owing to small variations in the Cr evaporation rate [8]. The amplitude of the RHEED intensity oscillations decreases at larger thickness indicating increased roughness and disorder.

The measured spin polarization for the Cr wedge is shown in Fig. 2(b). The polarization of 29% from the Fe substrate at the beginning of the Cr wedge decreases rapidly with Cr coverage until the polarization is dominated by the oscillations in the Cr polarization which have a maximum peak to peak value of 4.2% at 25 ML. The $1/e$ "sampling depth," or average attenuation length l, in Cr for the electrons measured by SEMPA is found to be $l=0.55\pm0.04$ nm by fitting an exponential to the polarization decrease as electrons from the Fe are attenuated by the Cr. The Cr polarization, after subtracting the exponential decrease in Fe polarization, is shown with the scale magnified by a factor of 4 in Fig. 2(c).

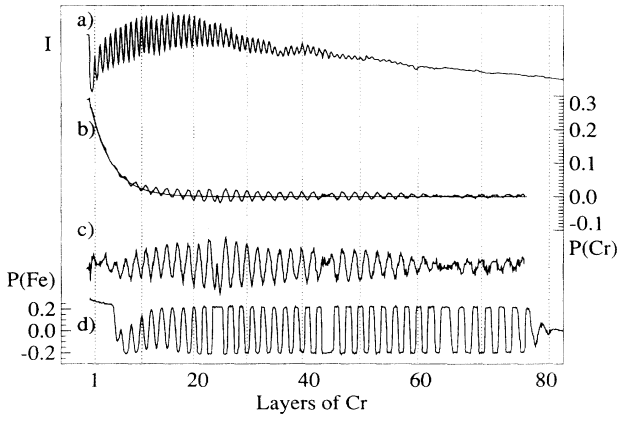

FIG. 2. (a) RHEED intensity oscillations determine the thickness of the Cr wedge deposited on the Fe whisker. (b) The spin polarization P(Cr) of secondary electrons emitted from the Cr wedge. (c) Data of (b) after subtracting the exponential shown and multiplying by 4. (d) The spin polarization P(Fe) from a layer of Fe (giving the magnetization image of Fig. 1) deposited on the Cr wedge of (a)–(c).

For purposes of comparison, we show in Fig. 2(d) the profile of the electron-spin polarization P(Fe) from the Fe overlayer which gave the magnetization image of Fig. 1. Comparing P(Cr) and P(Fe) in Figs. 2(c) and 2(d), we note that P(Fe) of the overlayer is opposite to that of P(Cr). This is consistent with spin-polarized photoemission measurements, which found that the Cr interface layer couples antiferromagnetically to Fe [9], if we assume that no change occurs in the Cr polarization direction with the addition of the Fe overlayer.

If Cr were a commensurate antiferromagnet, we would expect alternating planes of aligned spins for layer stacking in the [001] direction. Since the measured spin polarization is dominated by the surface layer, because of attenuation of electrons coming from deeper layers, P(Cr) is expected to change sign with each additional Cr layer, as observed [10] in Fig. 2. Since the first Cr layer couples antiferromagnetically to the Fe substrate, if P of the substrate is positive, one then expects for a commensurate antiferromagnet negative P(Cr) when the number of Cr layers is odd and positive P(Cr) when it is even. However, from Fig. 2(c) we see that the measured P(Cr) is negative for $n=4$, contrary to expectations. We conclude that there is a "defect" in the antiferromagnetic ordering between 1 and 4 layers giving a phase change in P(Cr). The presence of such a defect has been considered in calculations of the coupling in Fe/Cr/Fe [11].

While the polarization of secondary electrons from the bare Cr surface reverses approximately every layer, there are periodic changes in phase of P(Cr) at 24-25, 44-45, and 64-65 layers, the same places as in the Fe magnetization oscillations. A phase slip in the oscillation of the Cr surface moment occurs when an additional node of the SDW is introduced in the Cr film changing the antiferromagnetic stacking order by 1 layer. From neutron scattering measurements of bulk Cr, it is known that the Cr magnetic moments along a transverse SDW are in a lattice direction perpendicular to the **Q** vector [1]. The Cr moment in the SDW varies as $\mu=\mu_b\cos(Qz)$ where for bulk Cr at zero temperature $\mu_b=0.62\mu_B$. The period is given by $2\pi/Q$, where $Q=(\pi/d)(1-\delta)$, d is the layer spacing, and δ is a measure of the incommensurability [1]. The number of layers between phase slips is $1/\delta$. In our experiment, the coupling of the Cr to the Fe substrate both fixes the SDW wave vector **Q** in the [001] direction perpendicular to the Fe whisker surface and determines the polarization of the SDW along an axis given by the substrate magnetization.

If the phase of a SDW in the Cr film is pinned at the

Cr/Fe interface, one might expect to observe, with changing Cr thickness, a sinusoidal modulation of the amplitude of P(Cr) with a period identifiable with the phase slips. To first order, P(Cr) is approximately constant suggesting that the Cr surface has a well-defined magnetic moment. The amplitude of P(Cr) appears more closely tied to the film roughness as indicated by its correlation with the amplitude of the RHEED intensity oscillations. Moreover, the positions of the phase slips do not change when a layer of Fe is evaporated onto the Cr film, which is consistent with a ferromagnetic Cr surface layer providing a boundary condition similar to that of an Fe layer. We attribute the more abrupt reversals of P(Fe) in Fig. 2 compared to P(Cr) to the large *intralayer* coupling in the thicker Fe ferromagnetic layer.

All of the Cr measurements discussed so far were made just above room temperature near T_N for bulk Cr. As the temperature was varied over the measured range up to $1.8T_N$, the Cr thickness at which phase slips occurred varied reproducibly and reversibly. We plot the change in position with temperature of the phase slip nominally located at 24-25 layers in Fig. 3. Its position shifts 14 layers between 310 and 550 K. [P(Cr) for thicknesses greater than 40 layers was not measured as a function of temperature.] The phase of the oscillations for less than 24 layers was not observed to change with temperature. We also show in Fig. 3 the temperature dependence of $1/\delta$, the number of layers between phase slips, for bulk Cr from neutron scattering [12]. The 20 layers between phase slips measured for Cr/Fe(100) near T_N is fewer than in bulk Cr even at 0 K and the change on increasing the temperature is stronger. These differences are presumably due to the fact that the Cr lattice constant is 0.6% less than Fe leading to a strained Cr layer with a body-centered tetragonal structure and to the variation of strain with temperature owing to the different thermal expansion coefficients of Fe and Cr. The amplitude of the oscillations in P(Cr) changed less than 20% on heating to $1.8T_N$.

The magnetic behavior of Cr/Fe(100), exhibiting an incommensurate SDW within the film, even at temperatures well above the bulk Cr Néel temperature, is quite remarkable. This effect is due to the presence of the Fe substrate which influences the Cr thin film much differently than a nonmagnetic substrate or a semi-infinite Cr crystal would. Although thermal fluctuations destroy SDW antiferromagnetism in bulk Cr above T_N, the Fe substrate establishes a SDW in the Cr film for some distance from the interface. There are two closely related ways to view this response in the Cr. Even above the Néel temperature, the magnetic susceptibility is still enhanced at a wave vector **Q** where there is strong Fermi-surface nesting [13], so the Cr responds antiferromagnetically to the presence of the Fe. Alternatively, if the Cr is viewed as paramagnetic, RKKY-like oscillations would be established. These will be quite similar to the antiferromagnetic order because both derive from the same strong Fermi-surface nesting. A theoretical investigation will be required for a complete explanation of the underlying causes of the unusual behavior that we have observed.

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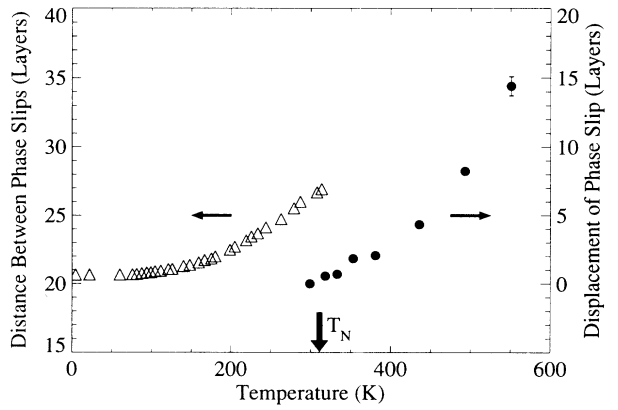

FIG. 3. The temperature dependence of the number of layers between phase slips $1/\delta$ for bulk Cr (from Ref. [12]) and the change in the position, in layers, of the phase slip in Cr/Fe(100) with temperature. The solid circles are larger than the error bars except at the highest temperature. The Néel temperature of bulk Cr is marked by the arrow.

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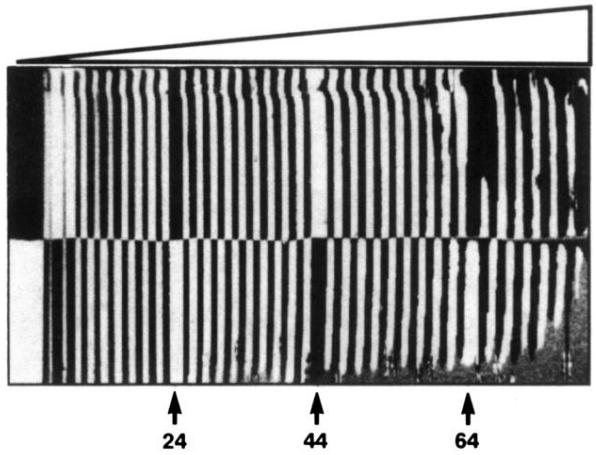

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