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LETTER TO THE EDITOR

Magnetic phase diagram of Fe/Cr multilayers and wedges

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Abstract. Based on a simple model with antiferromagnetic interactions at the Cr–Fe interfaces, we construct the magnetic phase diagram for the Cr spacer within Fe/Cr multilayers and wedges. The spin-density wave is found to undergo transitions between incommensurate (I) phases with a decreasing number of nodes n as the temperature increases for a fixed number N of Cr monolayers. In the limit $N \to \infty$, the incommensurate to commensurate (C) transition temperature from $n = 1$ to $n = 0$ nodes is independent of the Cr–Fe coupling strength but sensitively depends on the strain and doping. These results agree with recent neutron-scattering studies that the SDW inside Fe/Cr multilayers may be either C or I depending on the temperature and Cr thickness.

Although Fe/Cr multilayers have been extensively studied since the discovery of giant magnetoresistance in 1988 [1], the magnetic phase diagram of Fe/Cr multilayers has only recently [2, 3] been investigated using neutron scattering. Those studies reveal that the spin-density wave (SDW) [4] within the Cr spacer may be either commensurate (C) or incommensurate (I) with the bcc lattice. The C phase is stabilized when the number of monolayers (ML) N inside the Cr spacer is less than 30, or when the temperature exceeds the Néel temperature 310 K of pure Cr. By contrast, SEMPA measurements [5] on Fe/Cr/Fe wedges indicate that the I phase is stable for $N > 23$ MLs, and up to at least 550 K. Because the lattice strain induced by the Fe whisker on the bottom of the wedge should *suppress* the Néel temperature of Cr, the discrepancy between the measured IC transition temperature T_{IC} in wedges and multilayers is particularly puzzling. Based on a simple model with antiferromagnetic interactions at the Cr–Fe interfaces, we construct the magnetic phase diagram for the Cr spacer in Fe/Cr multilayers and wedges. Our model explains the different IC transition temperatures of strained and unstrained Cr films and reveals how T_{IC} can be controlled through doping and pressure.

Since the magnetic form factor of Cr is strongly peaked at the atomic sites [4], the SDW within the Cr spacer may be approximated by the form

$$\boldsymbol{S}(z) = \hat{m}\,\alpha_s\, g(-1)^{2z/a}\,\cos\left(\frac{2\pi}{a}\,\delta' z - \theta\right) \tag{1}$$

where α_s is a constant, θ is a phase and $g(T)$ is the order parameter. For bulk Cr at low temperatures, $\alpha_s g = 0.6\ \mu_B$. The hole Fermi surface of pure Cr is slightly larger than its electron Fermi surface so the nesting wavevectors $Q_\pm = (2\pi/a)(1 \pm \delta)$ differ from $2\pi/a$ by the mismatch [4] $\delta \approx 0.05$. The SDW ordering wavevectors $Q'_\pm = (2\pi/a)(1 \pm \delta')$ are obtained by minimizing the nesting free energy [6, 7] $\Delta F(g, \delta')$ with respect to g and δ'. When $\delta' = 0$, the SDW of equation (1) is C with the bcc lattice. For bulk Cr alloys, $0 \leqslant \delta' < \delta$ and the SDW ordering wavevectors are always closer to commensuration than

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the nesting wavevectors. Nodes of the I SDW are separated by $1/\delta'$ MLs, where each ML has a thickness of $a/2$. For pure Cr just below its Néel temperature, $1/\delta' \approx 27$.

Doping or lattice strain affects the bulk free energy of Cr alloys through the energy mismatch $z_0 = 4\pi\delta v_F/\sqrt{3}a$ (v_F is the Fermi velocity) between the electron and hole Fermi surfaces. If $T_N^* \approx 80$ meV is the Néel temperature of a perfectly nested alloy with $\delta = 0$ and $z_0 = 0$, then the bulk free energy can be written [7]

$$\Delta F(g, \delta', T, z_0) = \rho_{eh}\, g^2 \ln\left(\frac{T}{T_N^*}\right) + \rho_{eh} \sum_{n=0}^{\infty} \left\{ g^2 \frac{1}{n+1/2} - T \int_{-\infty}^{+\infty} \mathrm{d}\varepsilon \ \ln\left| 1 - g^2 \frac{2\mathrm{i}\omega_n - z_0 + 2\varepsilon}{(\mathrm{i}\omega_n - \varepsilon)[(\mathrm{i}\omega_n - z_0/2 + \varepsilon)^2 - (z_0\delta'/2\delta)^2]} \right| \right\} \qquad (2)$$

where $\omega_n = (2n+1)\pi T$ are the Matsubara frequencies and ρ_{eh} is the density-of-states of the nested portions of the Fermi surface. While doping with V increases z_0, doping with Mn lowers the mismatch between the Fermi surfaces. When the Mn concentration exceeds about 0.3%, the mismatch is sufficiently small to stabilize the C SDW phase with $\delta' = 0$ at T_N.

Due to the lattice mismatch between Cr and Fe, the lattice constant of Cr inside the Fe/Cr/Fe wedge is about 0.6% smaller [5] than in bulk. It is well known [4] that pressure (or strain) has the same effect on the Cr SDW as increasing the mismatch z_0 between its nested Fermi surfaces [8]. Additional evidence for this effect is provided by the enhanced incommensurability of the SDW within the wedge: the measured node-to-node distance of 20 MLs at 300 K is smaller than the 27 MLs expected for bulk Cr. We estimate the energy mismatch for the wedge to be $z_0 = 6.4T_N^*$, which is substantially larger than the mismatch $z_0 \approx 5T_N^*$ for pure Cr.

At a microscopically smooth interface, the Cr–Fe interaction is expected to be antiferromagnetic with the form $A\boldsymbol{S}_{Fe}^{I,II} \cdot \boldsymbol{S}(z)$ at interfaces I ($z = a/2$) and II ($z = Na/2$). Such an antiferromagnetic interaction has been observed in Fe/Cr multilayers [9] and was obtained in first-principles calculations [10]. For simplicity, we assume that the Fe moments are either F or AF aligned with $\boldsymbol{S}_{Fe}^{I} = \boldsymbol{S}_{Fe}^{II}$ or $\boldsymbol{S}_{Fe}^{I} = -\boldsymbol{S}_{Fe}^{II}$, both parallel to the interface. The SDW will then be transversely polarized with respect to the ordering wavevectors along the z axis. While these assumptions seem well satisfied in the wedges measured by NIST, recent measurements on Fe/Cr multilayers [2] indicate that interfacial disorder may produce a non-collinear coupling [11] between adjacent Fe moments. This in turn generates a spiral modulation of the Cr moment in the C SDW phase. We shall return to this possibility below.

With antiferromagnetic interactions at the interfaces, the free energy of the multilayer or wedge may be written as [12]

$$E = A\boldsymbol{S}_{Fe}^{I} \cdot \boldsymbol{S}(a/2) + A\boldsymbol{S}_{Fe}^{II} \cdot \boldsymbol{S}(Na/2) + \Delta F\, a^3\, (N-1)/2 \qquad (3)$$

where $A > 0$ is the antiferromagnetic coupling constant at an interface with area a^2. This assumes that the SDW is rigid with order parameters g and δ' independent of z. While the oscillations in the magnetic couplings were studied in a previous work [12], here we tackle the much more arduous task of constructing the general magnetic phase diagram of Fe/Cr multilayers and wedges.

Since the interfacial energies always induce some SDW ordering with $g > 0$ no matter how high the temperature, the paramagnetic state is unstable within this model. In a more realistic albeit far more complex model, the SDW amplitude $g(z)$ would vanish inside the spacer at a sufficiently high temperature despite some residual ordering near the interfaces at $z = a/2$ and $z = Na/2$. Even in the magnetically ordered state, the SDW order parameters

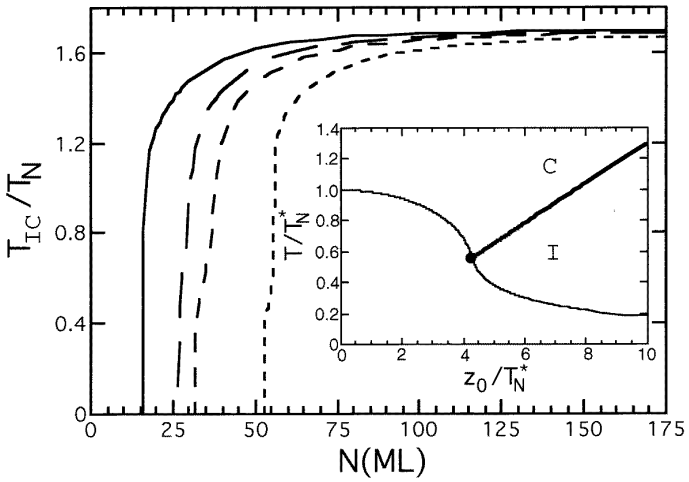

Figure 1. The IC transition temperature normalized by the bulk T_N versus N for $z_0 = 5T_N^*$ and $\gamma = 1$ (solid), 3 (long dash), 4 (medium dash) and 6 (short dash). In the inset is plotted the $N \to \infty$ limit of T_{IC}/T_N^* (thick solid) and the paramagnetic phase boundary T_N/T_N^* (thin solid) versus z_0/T_N^*. The solid circle denotes the bulk triple point.

may be enhanced near the interfaces. However, due to the small coherence length (4.3 Å) of an electron–hole pair in Cr, the SDW is altered only within two or three monolayers from the interface. This has been confirmed by recent first-principles calculations [10]. So the IC phase boundary evaluated from this model should be qualitatively accurate.

Because the nesting free energy ΔF is proportional to $\rho_{eh} T_N^{*2}$, the total free energy E depends only on the dimensionless constant $\gamma = A\,\alpha_s\,S_{\mathrm{Fe}}/(V/N)\rho_{eh}T_N^*$ [13]. For fixed γ, E is minimized with respect to the SDW parameters g and δ' [14]. Note that the number n of SDW nodes inside the Cr spacer is approximately given by $(N-1)\delta'$. In the absence of any external magnetic fields, E is also minimized with respect to the orientation of the Fe magnetic moments. When the bulk contribution $\Delta F(N-1)$ is sufficiently small compared with the interface energies, the C SDW has a lower free energy than the I SDW [12]. So the I phase becomes unstable at small spacer thicknesses N or at high temperatures.

The resulting IC transition temperature $T_{IC}(N)$ for Fe/Cr multilayers with $z_0 = 5T_N^*$ is plotted versus N in figure 1. With increasing γ, the interfacial energies dominate and the C phase is stabilized over a wider range of N and T. When $\gamma = 4$, the C phase is stable at all temperatures for $N \leqslant 31$ MLs, as found in sputtered Fe/Cr multilayers. With decreasing γ, this critical thickness tends to zero.

For large N, the IC transition temperature plotted in figure 1 is remarkably independent of the Cr–Fe coupling constant γ. In the limit of large N, the SDW order parameter g vanishes at the IC phase boundary like γ/N. After expanding the bulk free energy ΔF in powers of g^2 up to order g^4, we find that $T_{IC}(N \to \infty)$ is implicitly given by

$$\sum_{n=0}^{\infty} \mathrm{Re}\left(\frac{1}{{X_n}^3}\right) = 0 \tag{4}$$

where $X_n = n + 1/2 + \mathrm{i}z_0/8\pi T_{IC}(\infty)$. Consequently, $T_{IC}(\infty)$ depends only on the energy mismatch z_0 and is independent of γ. This expression is satisfied by the thick solid line in the inset to figure 1. The thin solid curve is the bulk paramagnetic phase boundary

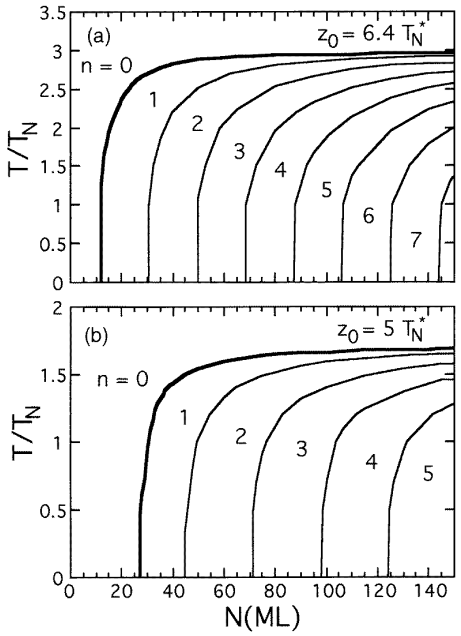

Figure 2. Phase diagram of Fe/Cr multilayers and wedges for $\gamma = 3$ and (a) $z_0 = 6.4T_N^*$ and $T_N = 0.282T_N^*$ or (b) $z_0 = 5T_N^*$ and $T_N = 0.384T_N^*$. The thick solid curve denotes the IC transition while the thin solid curves separate different I phases with n nodes.

T_N evaluated with $\gamma = 0$ and the filled circle is the bulk triple point [7] where the paramagnetic, C and I phases meet. Recall that the energy mismatch z_0 increases with V doping, lattice strain or pressure. Precisely at the triple point, $T_{IC}(\infty) = T_N$ and the two transitions coincide. In contrast to the behaviour of the bulk Néel temperature, however, the IC transition temperature increases with the energy mismatch z_0. So the lattice strain present in Fe/Cr/Fe wedges pushes T_{IC} far above both the bulk Néel temperature and the IC transition temperature of relatively strain-free sputtered Fe/Cr multilayers. Consequently, applying pressure to multilayers or wedges should enhance the incommensurability of the SDW and increase the IC transition temperature.

The magnetic phase diagrams of strained ($z_0 = 6.4T_N^*$) and unstrained ($z_0 = 5T_N^*$) Fe/Cr trilayers are plotted in figures 2(a) and (b). Both figures use $\gamma = 3$ for the coupling strength at the interface. The thick solid curves denote the IC transition while the thinner curves denote the transitions between I phases with different number (n) of nodes. At a fixed temperature, phase slips occur whenever a solid curve is crossed. Away from a phase slip, the magnetic coupling alternates between F and AF with increasing thickness N. On either side of a phase slip, the magnetic coupling (AF or F) is the same. If the phase slips occur between thicknesses N_i and $N_i - 1$, then the distance between phase slips is given by $s_i = N_{i+1} - N_i$. While the SDW is C before the first phase slip at N_1, the I SDW has $n = i$ nodes for $N_i \leqslant N \leqslant N_{i+1} - 1$. The distance s_1 between the first two phase slips is always the smallest. For large Cr spacers, the bulk free energy $\Delta F(N - 1)$ dominates the interfacial energies. So below the bulk Néel temperature, $s_i \to 1/\delta'$ as $i \to \infty$. In other words, the distance between phase slips approaches the distance between nodes of the bulk SDW. Above the bulk Néel temperature and close to the IC phase boundary, the phase slip

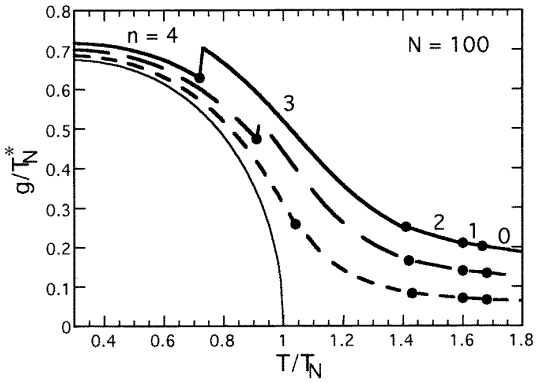

Figure 3. The SDW order parameter g/T_N^* versus the normalized temperature T/T_N for $z_0 = 5T_N^*$, $N = 100$ and $\gamma = 3$ (thick solid), 2 (long dash), 1 (short dash) and 0 (thin solid). The I–I and IC transitions are denoted by filled circles.

distances s_i become more disparate with the higher s_i diverging most rapidly as $T \to T_{IC}$.

In figure 3, we plot the dependence of the SDW amplitude g on temperature for $z_0 = 5T_N^*$, $N = 100$, and four values of the coupling strength γ. Transitions between different I phases and between the I and C phases are denoted by the filled circles. For example, when $\gamma = 1$ the I SDW undergoes transitions from a state with $n = 4$ nodes to one with $n = 3$ nodes, then $n = 2$ nodes, and then $n = 1$ node, before transforming into a C SDW with $n = 0$ nodes as the temperature passes $1.05T_N$, $1.44T_N$, $1.61T_N$ and $1.68T_N$, respectively. While the IC transition temperature for $N = 100$ is fairly insensitive to γ, the SDW amplitude quickly approaches its bulk value as $\gamma \to 0$.

Measurements by the NIST group [5] on Fe/Cr/Fe wedges closely follow the scenario depicted in figure 2(a). In terms of the Néel temperature $T_N = 0.384T_N^* \approx 310$ K of unstressed Cr, the IC transition of the stressed film is given by $1.98T_N \approx 615$ K. Although the NIST measurements only go up to about 550 K, this value is close to the IC transition temperature which can be extrapolated from the SEMPA data. The NIST group observed a very uniform pattern of phase slips with the same s_i depending only on temperature. The values of $\gamma = 3$ and $z_0 = 6.4T_N^*$ used in figure 2(a) were chosen to give the smallest possible variation of s_i and a bulk value of $1/\delta' = 19$ at $T_N = 227$ K, slightly smaller than the observed phase slip distance of 20 MLs at 300 K.

For $T = 300$ K, the first predicted phase slip at $N_1 = 13$ MLs in figure 2(a) occurs earlier than the first observed [5] phase slip at 24 MLs in an Fe/Cr/Fe wedge. Within our model, the distance N_1 to the first phase slip is always less than the limit $s_{i\gg1} \approx 20$ MLs and only approaches this value as $\gamma \to \infty$. Accounting for the intermixing of Fe and Cr within the first 5 MLs of the wedge [15], an initial phase slip at $5 + 19 = 24$ MLs can be obtained using a somewhat larger coupling constant of $\gamma \approx 6$. As expected, this value is larger than the coupling strength of $\gamma \approx 4$ in sputtered multilayers. The intermixing within the first few monolayers is also necessary to explain the reversal [5] of the expected F and AF couplings. Probably due to their restricted temperature range and the finite size of their wedge with $N < 80$ MLs, the NIST group did not observe the phase slip pattern to deviate from uniformity at high temperatures. Doping the Cr spacer with a small concentration of Mn impurities ($x < 0.3\%$) could lower the IC transition temperature below 550 K and allow this behaviour to be observed.

So far, neutron-scattering measurements on sputtered multilayers have not followed the same pattern as the NIST data. Although figure 2(b) for $z_0 = 5T_N^*$ predicts an IC transition temperature of about $1.7T_N \approx 530$ K, the measured IC transition temperature of 300 K is far smaller. Both Schreyer's measurements on Fe/Cr multilayers [2] and Fullerton's recent measurements on CrMn/Cr multilayers [16] indicate that the C and I phases coexist between 200 and 300 K. Neither of these groups find any sign of the I–I phase transitions predicted by figure 2(b) and clearly indicated by the NIST measurements. This behaviour may be attributed to thickness fluctuations or atomic steps at the interfaces. Such fluctuations may place the SDW nodes near the interfaces [3] or may favour a spiral C phase, such as observed by Schreyer *et al* [2]. Those possibilities are currently under investigation.

To summarize, we have evaluated the phase diagram of Fe/Cr trilayers by assuming that the magnetic interactions at the interfaces are antiferromagnetic. Our results are in good agreement with measurements on Fe/Cr wedges, where interfacial disorder is minimized and the scenario of I–I transitions for a fixed thickness N is supported.

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