Helical and Incommensurate Spin-Density Waves in Fe/Cr Multilayers with Interfacial Steps

R. S. Fishman

Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6032

(Received 28 May 1998)

Although absent in bulk transition metals, a noncollinear, helical (H) spin-density wave (SDW) is stabilized by steps at the interfaces in Fe/Cr multilayers. Using the random-phase approximation, we evaluate the phase boundary between the H SDW and the collinear, incommensurate (I) SDW found in bulk Cr. In agreement with neutron-scattering results, the I-to-H transition temperature T_{IH} is always lower than the bulk Néel temperature T_N and the nodes of the I SDW lie near the Fe-Cr interfaces. While a H SDW with a single $\pm\pi/2$ twist has lower free energy than a I SDW above T_N, H SDW's with larger twists are stable between T_{IH} and T_N. [S0031-9007(98)07739-4]

PACS numbers: 75.30.Fv, 75.30.Ds, 75.70.Cn

Despite its absence in bulk transition metals, recent neutron-scattering measurements [1,2] on Fe/Cr multilayers suggest that a helical (H) spin-density wave (SDW) appears inside the Cr spacer at high temperatures or small Cr thicknesses N. By contrast, extensive measurements on bulk Cr alloys [3] only reveal collinear, incommensurate (I), or commensurate (C) SDW's. Although predicted [4] to be stabilized by the steps at Fe-Cr interfaces, the precise conditions required to support a H SDW have been unknown. This Letter uses a simple model to compare the free energies of the C, H, and I SDW's in an Fe/Cr trilayer with interfacial steps.

Early measurements [5] on Fe/Cr multilayers indicated that the magnetic coupling between adjacent Fe layers survives up to about 500 K, far above the bulk Cr Néel temperature of $T_N \approx 310$ K. Since then, the role of the SDW in Fe/Cr multilayers has been intensely debated [6]. Only recently have neutron-scattering measurements [1,7] confirmed the presence of a SDW in Fe/Cr multilayers. Measurements by Schreyer *et al.* [1] strongly suggest that a noncollinear, H SDW produces the observed [8] 90° or biquadratic coupling [9] between adjacent Fe moments for thicknesses below 30 monolayers (ML's) or temperatures above T_N. The biquadratic coupling gradually disappears for larger thicknesses or smaller temperatures, as the H SDW is replaced by an I SDW with nodes close to the Fe-Cr interfaces [7]. Schreyer *et al.* [1] find that the H and I SDW phases coexist in a region of thicknesses above 30 ML's and for temperatures between 200 and 300 K.

Based on a tight-binding approximation, Stoeffler and Gautier [10] first argued that a H SDW would be stable in a perfect Fe/Cr trilayer when the orientation of the Fe moments frustrates C ordering. In the presence of steps at the Fe-Cr interface, a single C SDW domain would be totally decoupled from the neighboring Fe moments. Yet as shown in Fig. 1, two sets of H SDW domains with opposite helicity [4] can maintain their antiferromagnetic coupling with Fe moments that are oriented 90° apart [11].

A H SDW is unstable in bulk Cr [12] because its free energy is always higher than that of a C or I SDW. Remarkably, a H SDW was the first SDW predicted by Overhauser [13] in 1960. Soon afterwards, however, polarized neutron-scattering measurements [14] revealed that the SDW in pure Cr was collinear.

All three SDW states are produced by the nearly perfect nesting [15,16] of electron and hole Fermi surfaces which are roughly octahedral in shape. Because the hole Fermi surface is slightly larger than the electron Fermi surface, the nesting wave vectors $Q_\pm = (G/2)(1 \pm \delta)$ differ from $G/2 = 2\pi/a$. To maximize the nesting on both sides of the Fermi surfaces [17], the ordering wave vectors of the SDW $Q'_\pm = (G/2)(1 \pm \delta')$ are slightly closer to $G/2$ than the nesting wave vectors with $0 \leq \delta' < \delta$.

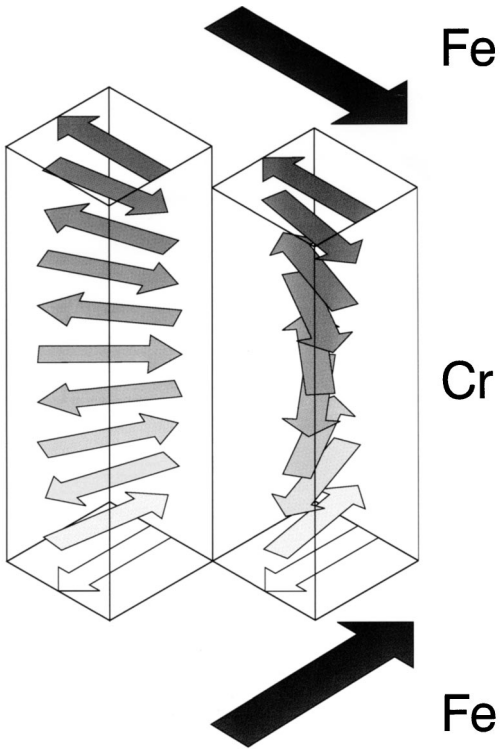

FIG. 1. A sketch of two H SDW's, one right handed ($m = 1$) and the other left handed ($m = -1$), coupling Fe moments 90° apart due to a step at the interface.

Since the Bloch wave functions are sharply peaked at the lattice sites, they may be replaced by delta functions in the spin density. With $\mathbf{Q}'_{\pm}$ along the z axis, the local spins in the I and H SDW phases can be written

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

$$\mathbf{S}_{\mathrm{H}}(z) = \alpha_s g(-1)^{2z/a}\left\{\hat{x}\cos\left(\frac{2\pi}{a}\,\delta' z - \theta\right) + \hat{y}\sin\left(\frac{2\pi}{a}\,\delta' z - \theta\right)\right\}, \quad (2)$$

where α_s is a constant, $\hat{m}$ is the polarization of the I SDW, θ is an arbitrary phase, and $g(T)$ is the order parameter. At low temperatures in bulk Cr [3], $\alpha_s g = 0.6\mu_{\mathrm{B}}$. For an I SDW, the distance between nodes is $1/\delta'$ ML's. For a H SDW, this is the distance for a π twist. Just below the Néel temperature of pure Cr, the SDW nodes are separated by $1/\delta' \approx 27$ ML's [3].

The mismatch between the electron and hole Fermi surfaces is measured by the energy $z_0 = 4\pi\delta v_F/\sqrt{3}a$, where v_F is the Fermi velocity. As z_0 and δ are decreased by doping with Mn or Fe, δ' is also diminished. At some critical value of z_0, δ' drops to zero and the SDW becomes commensurate with $Q'_{\pm} = G/2$. In the limit $\delta' \to 0$, $\mathbf{S}_{\mathrm{I}}(z)$ and $\mathbf{S}_{\mathrm{H}}(z)$ reduce to C SDW's with amplitude $\alpha_s g$.

Within the random-phase approximation, the change in the bulk SDW free energy below T_{N} is given by [17,18]:

$$\Delta F_{\mathrm{I}}(g,\delta',T,z_0) = \rho_{eh} g^2 \ln\left(\frac{T}{T^*_{\mathrm{N}}}\right) + \rho_{eh}\sum_{n=0}^{\infty} \times \left\{g^2\,\frac{1}{n+1/2} - T\int_{-\infty}^{+\infty} d\varepsilon \ln\left|1 - g^2\,\frac{2i\omega_n - z_0 + 2\varepsilon}{(i\omega_n - \varepsilon)[(i\omega_n - z_0/2 + \varepsilon)^2 - (z_0\delta'/2\delta)^2]}\right|\right\}, \quad (3)$$

$$\Delta F_{\mathrm{H}}(g,\delta',T,z_0) = \rho_{eh} g^2 \ln\left(\frac{T}{T^*_{\mathrm{N}}}\right) + \rho_{eh}\sum_{n=0}^{\infty} \times \left\{g^2\,\frac{1}{n+1/2} - \frac{1}{2}\,T\int_{-\infty}^{+\infty} d\varepsilon \ln\left|\left(1 - \frac{2g^2}{(i\omega_n - \varepsilon)(i\omega_n - z_0/2 + \varepsilon - z_0\delta'/2\delta)}\right) \times \left(1 - \frac{2g^2}{(i\omega_n - \varepsilon)(i\omega_n - z_0/2 + \varepsilon + z_0\delta'/2\delta)}\right)\right|\right\}, \quad (4)$$

where $\omega_n = (2n+1)\pi T$ are the Matsubara frequencies, ρ_{eh} is the density of states of the nested portions of the Fermi surfaces, and $T^*_{\mathrm{N}} \approx 100$ meV is the Néel temperature of a perfectly nested alloy with $\delta = 0$ and $z_0 = 0$. We shall use a value for the mismatch energy of $z_0 = 5T^*_{\mathrm{N}}$, which is appropriate for pure, unstressed Cr.

In the limit $\delta' \to 0$, ΔF_{I} and ΔF_{H} reduce to the same C free energy. The bulk values of the SDW order parameter and wave vector are obtained by minimizing these free energies with respect to g and δ'. Both H and I SDW states have the same Néel temperature and the same period $1/\delta'$ at T_{N} [19]. Below T_{N}, however, a H SDW has a shorter period than an I SDW. For any fixed z_0 and $T < T_{\mathrm{N}}$, the minimum value of ΔF_{H} exceeds the minimum value of ΔF_{I} so that a H SDW (with $\delta' > 0$) always has a higher free energy than an I SDW. Nonetheless, the stability of the H phase in the presence of interfaces is possible only because the H SDW state already provides a local minimum of the bulk Cr free energy.

The total energy E of the multilayer is modeled by simply adding the free energy $\Delta F a^2 L$ of the spacer (with area a^2) and the interfacial coupling energy $E_{\mathrm{coup}} = A[\mathbf{S}^{\mathrm{I}}_{\mathrm{Fe}}\cdot\mathbf{S}(0) + \mathbf{S}^{\mathrm{II}}_{\mathrm{Fe}}\cdot\mathbf{S}(L)]$, where $L = (N-1)a/2$ is the width of the Cr spacer. In accord with the observations of Fullerton *et al.* [7], we assume that the SDW is rigid with the same amplitude and wave vector throughout each domain. Indeed, the inherent "softness" of bulk Cr—as evidenced by its rotational and translational Goldstone modes [20]—is broken by its interactions with the two interfaces. But as mentioned latter, some softness may be retained by an I SDW in a large spacer. In the absence of interface steps, Shi and Fishman [21] employed this model to evaluate the magnetic phase diagram of an Fe/Cr wedge [22] with nearly perfect interfaces.

To calculate the coupling energy, we assume that the regions of the spacer with thicknesses N and $N+1$ (or $N-1$) are the same. For the H phase, this implies that adjacent Fe moments lie 90° apart [4]. For the collinear I phase, adjacent Fe moments are either parallel (F) or antiparallel (AF). Minimizing E_{coup} with respect to the arbitrary phase θ of the I SDW for F or AF Fe moments, we find

$$E^{(\mathrm{F})}_{\mathrm{coup}} = -A\alpha_s g S_{\mathrm{Fe}}|\cos\phi - \cos(\pi\delta' - \phi)|, \quad (5)$$

$$E^{(\mathrm{AF})}_{\mathrm{coup}} = -A\alpha_s g S_{\mathrm{Fe}}|\sin\phi + \sin(\pi\delta' - \phi)|, \quad (6)$$

where $\phi = (\pi L/a)(1+\delta')$. As expected, both coupling energies vanish in the C phase with $\delta' = 0$. In the I phase, the coupling energies are minimized when the SDW nodes lie precisely at the Fe-Cr interfaces. However, the actual spin configuration is obtained by minimizing the *total* energy $E_{\mathrm{F,AF}} = E^{(\mathrm{F,AF})}_{\mathrm{coup}} + \Delta F_{\mathrm{I}} a^2 L$ with respect to g and δ'.

Despite the rotation of the SDW in the H phase, neighboring Cr and Fe moments are still assumed to be antiparallel [23] at the interfaces. Hence, the H coupling energy in each domain is given by $E_{\rm coup} = -2A\alpha_s g S_{\rm Fe}$. The boundary conditions imposed on the H SDW in a domain with thickness N restrict the wave vector to values $\delta'(m,N) \equiv m/2(N-1)$, where $m = 2n+1$ is an odd integer and the helix rotates through the angle $m\pi/2$ from $z = 0$ to $z = L$. For $m > 0$ and $\delta' > 0$, Eq. (2) indicates that the helix is right handed; for $m < 0$, the helix is left handed. Because $\delta'(m,N) \approx \delta'(m,N \pm 1)$ for large N, the bulk free energies $\Delta F_{\rm H} a^2 L$ of H SDW's in the two sets of domains with thicknesses N or $N \pm 1$ are taken to be the same.

Since the bulk free energies are proportional to $\rho_{eh} T_{\rm N}^{*2}$, the total energy E depends on the single dimensionless parameter $\gamma = A\alpha_s S_{\rm Fe}/(V/N)\rho_{eh} T_{\rm N}^{*}$. For a perfect Fe-Cr interface with $AS_{\rm Fe}^2 \approx 100$ meV [21], $\gamma \approx 12$. However, interfacial roughness and interdispersion suppress γ by an unknown amount. The phase diagram of an Fe/Cr wedge is fit rather well with $\gamma = 3$ [21].

Interfacial steps have two important effects within this model. First, steps frustrate C ordering so that a C SDW does not gain any coupling energy at the interfaces. Second, steps reduce the coupling between an I SDW and the neighboring Fe moments to the point that a H SDW has the lower total free energy for high temperatures or small thicknesses.

In Fig. 2, the IH phase boundary for $\gamma = 2$ is plotted in the solid curve. Above $T_{\rm IH}$, a H SDW with twist parameter $|m|$ has the lowest free energy between the thin dashed lines. So a helix with a single $\pm\pi/2$ twist is stable for thicknesses below 25 ML's. Higher-order helixes with $|m| > 1$ become stable as N increases. Different $|m|$ states may be distinguished by polarized neutron-scattering measurements with the scattering wave

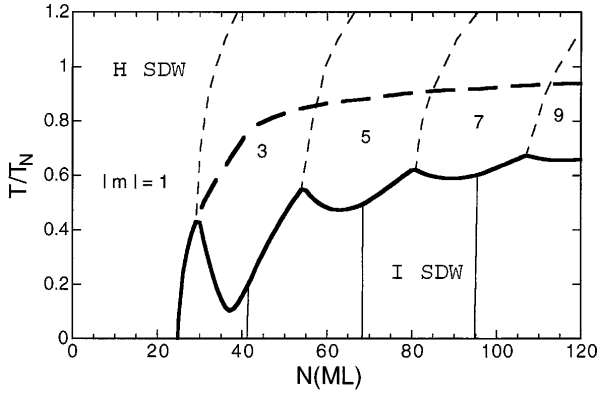

FIG. 2. Phase diagram of an Fe/Cr multilayer with $\gamma = 2$ and $z_0/T_{\rm N}^{*} = 5$. The thick solid curve gives the IH transition temperature between an I SDW and a H SDW with an $m\pi/2$ twist. The thick dashed curve gives the phase boundary between an I SDW and a H SDW with a single $\pm\pi/2$ twist. Below $T_{\rm IH}$, the magnetic coupling between adjacent Fe moments experiences a phase slip across the thin, nearly vertical lines.

vector in the plane of the multilayer. This figure clearly indicates that the I-to-H phase transition occurs below the bulk Néel temperature of the Cr spacer. For perfect interfaces, on the other hand, the I-to-C phase transition predicted by Ref. [21] happens above the bulk $T_{\rm N}$.

With increasing thickness, the IH transition temperature passes through consecutive valleys. The maxima in $T_{\rm IH}$ occur as $|m|$ changes by 2. The minima in $T_{\rm IH}$ occur when the H SDW wave vector passes closest to its bulk value. Similar oscillations in the transition temperature are produced by a model [24] which forces the I SDW nodes to lie at the Fe-Cr interfaces. For larger values of γ, the lower critical thickness at $T = 0$ increases and $T_{\rm IH}$ decreases as the larger coupling energy of the H SDW gains it more of an advantage over the I SDW.

Even with steps at the interface, the I SDW continues to magnetically couple adjacent Fe layers below $T_{\rm IH}$. However, $J_{\rm coup} = E_{\rm AF} - E_{\rm F}$ is roughly an order of magnitude smaller than in the absence of interfacial steps [21]. The magnetic coupling and order parameters of the I SDW at $T/T_{\rm N} = 0.05$ are plotted in Fig. 3. With increasing thickness, the coupling alternates between F and AF except on either side of the nearly vertical lines in Fig. 2, when the magnetic coupling repeats. As in Fe/Cr wedges, these phase slips are roughly separated by the bulk value of $1/\delta' \approx 26.4$ ML's. Between phase slips, the SDW stretches to keep its nodes near the interfaces. Across a phase slip, both the SDW amplitude and period change discontinuously as the SDW suddenly contracts. A similar series of oscillations about the bulk order parameters, except with much larger magnitudes, was predicted for Fe/Cr wedges [21].

Unlike a H SDW, an I SDW in a large spacer can adjust to the presence of lattice defects by shifting the position of its nodes with very little cost in free energy. Higher-order helixes with $|m| > 1$ may be especially frustrated by the presence of defects in the Cr spacer. Assuming that only $|m| = 1$ helixes are stable, the IH phase boundary $T_{\rm IH}^{|m|=1}$ is plotted in the thick dashed curve of Fig. 2. As expected, $T_{\rm IH}^{|m|=1}$ is always larger than $T_{\rm IH}$ but still lies below $T_{\rm N}$. For realistic Fe/Cr multilayers, we expect that the I SDW and higher-order H SDW's coexist in the region between $T_{\rm IH}$ and $T_{\rm IH}^{|m|=1}$. With increasing γ, this coexistence region becomes even larger as $T_{\rm IH}$ decreases, so this calculation may explain the IH transition region observed by Schreyer *et al.* [1] between 200 and 300 K in epitaxially grown multilayers.

By contrast, higher-order helixes may be unable to form in rougher, sputtered multilayers. Then the thick dashed curve would correspond to the monotonically increasing transition temperature measured by Fullerton *et al.* [25]. Fitting this curve to the expression $1 - T_{\rm IH}^{|m|=1}(N)/T_{\rm N} = b(N - N_0)^{-\lambda'}$ yields the exponent $\lambda' \approx 0.86$, consistent with the result $\lambda' \approx 0.8 \pm 0.1$ of the Argonne group [25].

Because the coupling energy of the I SDW is so small in the presence of a step, almost exactly the same IH phase

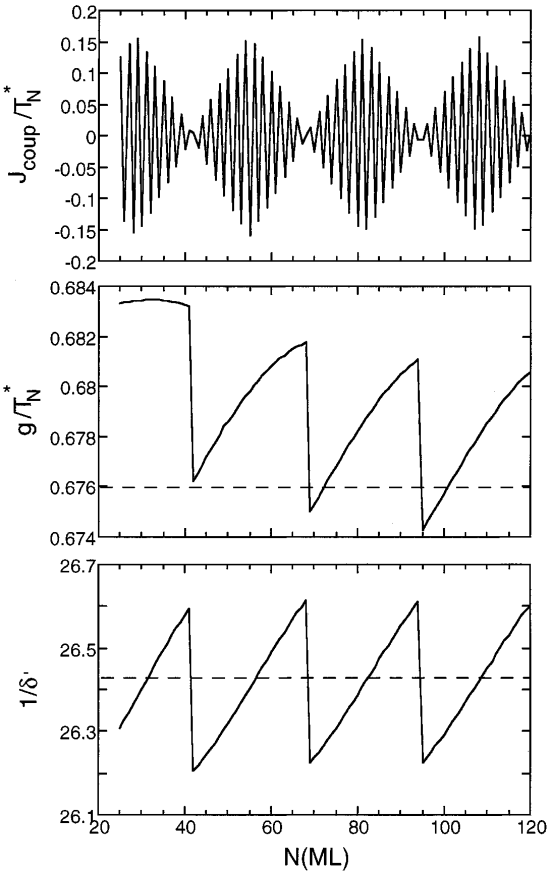

FIG. 3. The magnetic coupling and order parameters of the I SDW for the same parameters as in Fig. 2 and $T/T_N = 0.05$. Dashed horizontal lines denote the bulk values of the SDW amplitude and period.

boundaries $T_{IH}(N)$ and $T_{IH}^{|m|=1}(N)$ would be obtained if the I SDW were completely decoupled from the Fe moments [so that $E_{coup}^{(F,AF)} = 0$ in Eqs. (5) and (6)]. In that case, $J_{coup} = 0$ and the I SDW amplitude and wave vector would be given by their bulk values. The condensation energy itself may then favor placing the I SDW nodes close to a rough interface [24].

To summarize, we have evaluated the magnetic phase diagram of Fe/Cr multilayers with steps at each interface. A series of H SDW's are stabilized by the interfacial coupling at small thicknesses or high temperatures. In an intermediate range of temperatures between T_{IH} and $T_{IH}^{|m|=1}$, the I SDW and H SDW may coexist due to the frustration experienced by higher-order H SDW's in the presence of lattice defects.

The author would like to thank Dr. Sam Liu, Dr. Lee Robertson, Dr. Andreas Schreyer, and Dr. Zhupei Shi for helpful discussions. This research was supported by Oak Ridge National Laboratory managed by Lockheed Martin Energy Research Corp. for the U.S. Department of Energy under Contract No. DE-AC05-96OR22464.

[1] A. Schreyer *et al.*, Europhys. Lett. **32**, 595 (1995); Phys. Rev. Lett. **79**, 4914 (1997).
[2] P. Bödeker *et al.*, Phys. Rev. Lett. **81**, 914 (1998).
[3] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988); E. Fawcett *et al.*, Rev. Mod. Phys. **66**, 25 (1994).
[4] J. C. Slonczewski, J. Magn. Magn. Mater. **150**, 13 (1995).
[5] S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).
[6] M. Donath *et al.*, Phys. Rev. B **43**, 13 164 (1991).
[7] E. E. Fullerton, S. D. Bader, and J. L. Robertson, Phys. Rev. Lett. **77**, 1382 (1996).
[8] S. Adenwalla *et al.*, Phys. Rev. B **53**, 2474 (1996).
[9] J. C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991).
[10] D. Stoeffler and F. Gautier, J. Magn. Magn. Mater. **121**, 259 (1993).
[11] Two sets of C SDW domains with polarizations 90° apart can also couple perpendicular Fe moments with Cr spins at $z = 0$ and L making 135° angles with their neighboring Fe moments. However, each C SDW will be twisted by the Fe moments, turning this spin configuration into the pair of H SDW's sketched in Fig. 1.
[12] K. Hirai, J. Phys. Soc. Jpn. **65**, 586 (1996).
[13] A. W. Overhauser, Phys. Rev. Lett. **4**, 462 (1960).
[14] P. J. Brown *et al.*, Proc. Phys. Soc. London **85**, 1185 (1965).
[15] W. M. Lomer, Proc. Phys. Soc. London **80**, 489 (1962).
[16] P. A. Fedders and P. C. Martin, Phys. Rev. **143**, 8245 (1966).
[17] R. S. Fishman and S. H. Liu, Phys. Rev. B **48**, 3820 (1993).
[18] R. S. Fishman (unpublished).
[19] A. Shibitani, K. Motizuki, and T. Nagamiya, Phys. Rev. **177**, 984 (1969).
[20] R. S. Fishman and S. H. Liu, Phys. Rev. Lett. **76**, 2398 (1996).
[21] Z. P. Shi and R. S. Fishman, Phys. Rev. Lett. **78**, 1351 (1997); R. S. Fishman and Z. P. Shi, J. Phys. C **10**, L277 (1998).
[22] J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett. **67**, 140 (1991); **69**, 1125 (1992).
[23] Relaxing this approximation and allowing the angle between neighboring Cr and Fe moments to deviate from 180° lowers the IH transition temperature slightly but does not affect any of our conclusions.
[24] R. S. Fishman, Phys. Rev. B **57**, 10 284 (1998).
[25] E. E. Fullerton *et al.*, Phys. Rev. Lett. **75**, 330 (1995).