Suppression of Biquadratic Coupling in Fe/Cr(001) Superlattices below the Néel Transition of Cr

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(Received 13 February 1995)

The Néel temperature (T_N) of Cr in sputtered, epitaxial Fe/Cr(001) superlattices is identified via transport and magnetic anomalies. The onset of the antiferromagnetism is at a thickness t_{Cr} of 42 Å. The bulk value of T_N is approached asymptotically as t_{Cr} increases and is characterized by a three-dimensional shift exponent. Most strikingly, the biquadratic interlayer magnetic coupling (90° orientation of adjacent Fe layers), observed in the thick Cr regime, is suppressed below T_N. These results are attributed to finite-size effects and spin frustration near rough Fe-Cr interfaces.

PACS numbers: 75.70.Fr, 73.50.Jt, 75.30.Kz, 75.50.Rr

Fe/Cr superlattices exhibit a variety of intriguing magnetic properties, including oscillatory interlayer coupling [1–4], giant magnetoresistance [5], non-Heisenberg biquadratic coupling [6], and the first experimental realization of the surface spin-flop transition [7]. For the case of Cr(100) spacers, two periods in the interlayer coupling have been observed, a short period [2 monolayers (ML)] and a long period (18 Å or 12 ML) [2–4,8]. The short-period oscillations result from the nested feature in the ⟨100⟩ direction of the Cr Fermi surface which also is responsible for the spin-density-wave (SDW) antiferromagnetism (AF) of Cr. The long period has also been observed in (110)- [2] and (211)- [9] oriented films, which suggests that it is not related to the nesting, but results instead from a short spanning vector associated with the relatively isotropic "lens" feature of the Fermi surface [10]. The observed coupling depends on the roughness of the spacer layers, with atomic-scale fluctuations of the Cr thickness (i.e., steps) being sufficient to suppress the short period, leaving only the long-period coupling [8]. However, Slonczewski [11] has shown that fluctuations in the short-period interactions can give an additional nonoscillatory, biquadriatic coupling term in which the magnetization orientation between adjacent Fe layers is 90°, rather than 180° or 0°.

An outstanding problem in the Fe/Cr system is to understand the role of the magnetic ordering within the Cr spacers. Bulk Cr is an itinerant antiferromagnet with a Néel temperature (T_N) of 311 K. The incommensurate SDW is characterized by a wave vector $\mathbf{Q}$ determined by the nesting. At high temperature, the Cr spin sublattices $\mathbf{S}$ are transverse to $\mathbf{Q}$ ($\mathbf{S} \perp \mathbf{Q}$), while below the spin-flip transition at 123 K $\mathbf{S}$ rotates 90° to form a longitudinal SDW with $\mathbf{S} \| \mathbf{Q}$ [12]. The AF order of thin-film Cr is less well studied, particularly in proximity to Fe layers. Two recent studies [13,14] reported observations that the surface-terminated ferromagnetic layer of Cr on an Fe(001) substrate oscillates in its magnetization orientation relative to that of the Fe with an $\approx$2 ML period, consistent with the SDW AF anticipated for Cr. Reference [13] further reported observation of the oscillations well above the bulk T_N value, which suggests that the substrate, a relatively perfect Fe whisker, stabilizes the antiferromagnetic spin structure of the Cr. In a complementary study [15] it was shown that the magnetism of Fe films on a Cr(001) substrate is modified by the AF order of the Cr. In this Letter, we investigate the AF ordering of Cr spacer layers in sputtered, epitaxial Fe/Cr(001) superlattices and report a number of new observations. First, we find that the AF order is suppressed for Cr spacers of thickness $t_{Cr} < 42$ Å. Second, for $t_{Cr} > 42$ Å, T_N initially rises rapidly, asymptotically approaches the bulk value for the thickest spacers studied (165 Å), and exhibits a transition-temperature shift exponent $\lambda = 1.4 \pm 0.3$ characteristic of three-dimensional (3D) Heisenberg or Ising models. Third, the AF ordering of the Cr spacers results in anomalies in a variety of physical properties, including the interlayer coupling, remanent magnetization (M_r), coercivity (H_c), resistivity (ρ), and magnetoresistance (MR). Finally, and most intriguingly, the biquadratic coupling of the Fe layers observed for $T > T_N$ vanishes below T_N.

Epitaxial Fe/Cr(001) superlattices were grown by dc magnetron sputtering onto MgO(001) single-crystal substrates. The growth procedure and structural characterizations are given in detail elsewhere [9]. The Fe thickness was held constant at 14 Å and t_{Cr} was varied from 8 to 165 Å. The magnetic properties were measured by a SQUID magnetometer from 10 to 350 K with the applied field H in plane along either the Fe ⟨100⟩ easy axis or ⟨110⟩ hard axis. Transport properties were measured using a standard four-terminal dc technique with a constant current of 10 mA and H in plane along the Fe ⟨001⟩.

 0031-9007/95/75(2)/330(4)$06.00

Transport measurements are often used as a probe of the AF ordering in Cr and Cr alloys, where ρ is enhanced above its extrapolated value as the temperature T decreases through T_N [12,16]. This anomaly in ρ, attributed to the formation of energy gaps opening on the nesting parts of the Fermi surface, is commonly used to locate T_N. Shown in Fig. 1 are T-dependent transport results for an [Fe(14 Å)/Cr(70 Å)]$_{13}$ superlattice. Figure 1(a) shows ρ vs T for $H = 500$ Oe, which is a sufficient field to align the Fe magnetization. An anomaly in ρ below ~200 K is observed as an increase above its expected linear behavior, as shown by the dotted line. The difference between the measured ρ and the linear extrapolation ρ_{lin} is plotted in Fig. 1(b). The magnitude of the ρ enhancement of 7% is consistent with similar measurements in bulk Cr and Cr(001) films [12,17]. The reduced value of $T_N = 195$ K is determined by the point of inflection of the ρ vs T [see Fig. 1(c)].

The AF ordering of the Cr dramatically alters the interlayer coupling of the Fe. Shown in Fig. 2 are the magnetic and MR data measured above and below T_N for the same superlattice with H applied in plane along the Fe ⟨100⟩ easy axis. Below T_N, M/M_s, where M_s is the saturation value, has a nearly square hysteresis loop and the low MR value (≈0.3%) is comparable to that for ferromagnetically coupled or uncoupled superlattices. However, increasing T above T_N to 220 K changes both M/M_s and the MR: M_r decreases to ≈0.5M_s and the MR increases. The reduction in the M_r value in Fig. 2(a) is not attributable to a change in the easy-axis direction of the Fe; magnetization measurements with H along the ⟨110⟩ hard axis have an $M_r = 0.71M_s$ at both temperatures, which is expected for a magnetization that rotates 45° from the field direction to the Fe ⟨100⟩ easy axis. Thus, we conclude that the reduction in M_r results from a change in the Fe interlayer coupling as T increases. The M_r value for $T > T_N$ is consistent with a 90° alignment of the magnetization of adjacent Fe layers, indicative of biquadratic interlayer magnetic coupling [18], while the M_r value for $T < T_N$ is consistent with a vanishing of the biquadratic coupling that leaves the layers relatively uncoupled.

Shown in Fig. 3 is the T dependence of M_r, H_c, the saturation field (H_s), and the MR of the superlattice in Figs. 1 and 2. All four quantities exhibit anomalous behavior which is directly related to the Néel transition of the Cr. The M_r shows a transition at T_N from a value of 0.53M_s for $T > T_N$ to 0.95M_s for $T < T_N$, as discussed above. The H_c values peak at T_N in a manner often observed in systems which undergo magnetic phase transitions [19]. H_s in Fig. 3(c), the field at which M reaches 90% of M_s, is roughly proportional to the interlayer coupling strength [20], and increases strongly with decreasing $T > T_N$, reaches a maximum at ~230 K, and then decreases sharply and approaches zero at T_N. The MR in Fig. 3(d) also shows an anomaly at T_N, and is consistent with a loss of interlayer coupling below T_N.

Utilizing both the transport and magnetic properties to identify T_N, Fig. 4(a) shows T_N vs t_{Cr} for a series of (001)-oriented Fe(14 Å)/Cr(t_{Cr}) superlattices. For $t_{\text{Cr}} <$ 42 Å there is no evidence of the Cr ordering. For $t_{\text{Cr}} >$

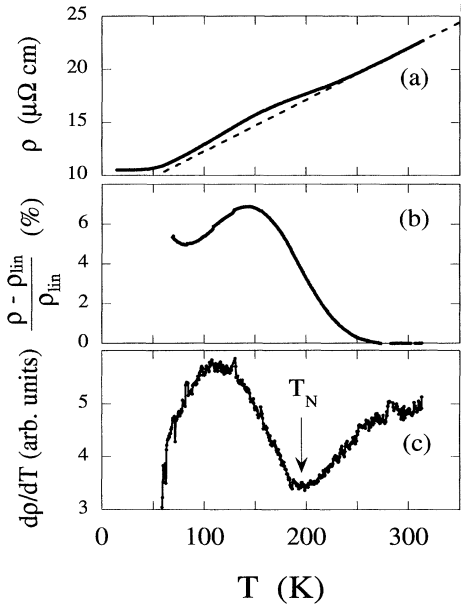

FIG. 1. Resistivity of an [Fe(14 Å)/Cr(70 Å)]$_{13}$ superlattice. (a) ρ vs T measured at $H = 500$ Oe. The dashed line is a linear extrapolation ρ_{lin} of the data above 280 K. (b) The difference between the measured ρ and ρ_{lin} normalized to ρ_{lin}. (c) Numerical derivative of ρ smoothed for clarity. The minimum in $d\rho/dT$ locates T_N.

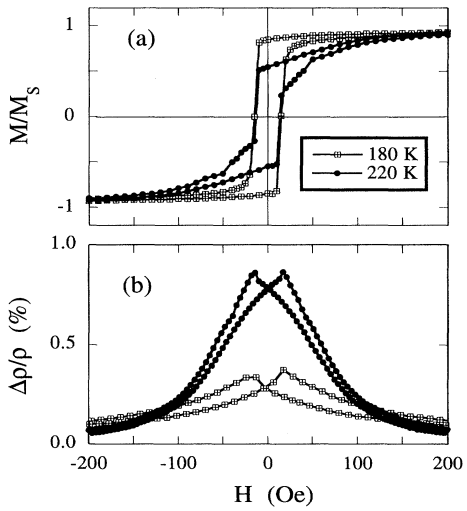

FIG. 2. (a) Magnetic and (b) magnetoresistance results for the [Fe(14 Å)/Cr(70 Å)]$_{13}$ superlattice in Fig. 1 measured above and below $T_N = 195$ K. H is applied along the ⟨100⟩ easy axis.

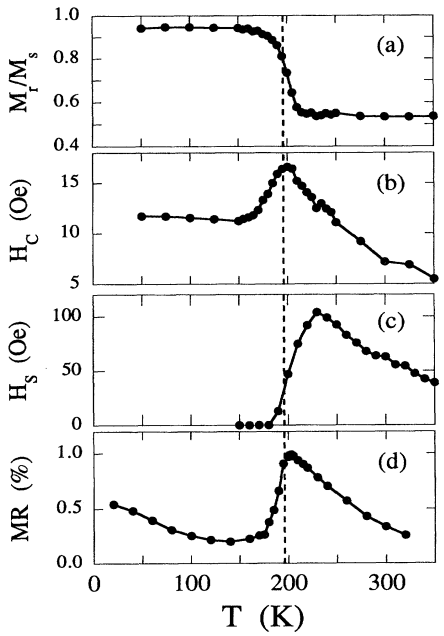

FIG. 3. Temperature dependent magnetization results for the [Fe(14 Å)/Cr(70 Å)]$_{13}$ superlattice in Figs. 1 and 2. (a) Squareness ratio M_r/M_s, (b) coercivity, (c) saturation field defined at 90% of M_s, and (d) the magnetoresistance. The vertical dashed line locates T_N of the Cr layers.

42 Å, T_N increases rapidly and reaches a value of 265 K for a 165 Å Cr thickness. For a 3000 Å thick Cr film grown in similar fashion to the superlattices, a T_N value of 295 K was obtained. A number of factors [12] can alter the T_N value of Cr, including impurities, pressure, defects, grain size [21], and epitaxial strain [17]. The unusual

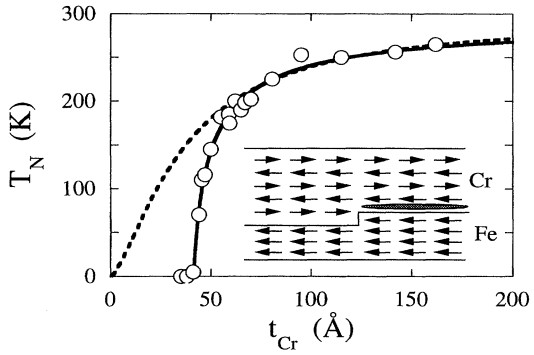

FIG. 4. T_N for a series of [Fe(14 Å)/Cr(70 Å)]$_{13}$ superlattices vs Cr thickness. The open circles are the measured values, and the dashed and solid curves are fitted by Eqs. (1) and (2), respectively. The measured T_N value for a 3000 Å Cr film (not shown) is 295 K. The inset shows a possible spin configuration of Cr on a stepped Fe surface in which the region of spin frustration at the Fe-Cr interface is shown schematically by the shaded ellipse to the right of the atomic step.

behavior of T_N vs t_{Cr} reported here can be understood as arising from a combination of finite-size effects within the Cr spacer and spin-frustration effects at the Fe-Cr interface, as will be discussed in the remainder of the paper.

In thin films, magnetic properties are altered due to the surface contribution to the free energy [22]. Since this contribution is generally positive, the magnetic order is weakened at the surface and the ordering temperature is reduced. Scaling theory predicts that T_N should have the form

$$[T_N(\infty) - T_N(t_{Cr})]/T_N(t_{Cr}) = b\, t_{Cr}^{-\lambda}, \qquad (1)$$

where $\lambda = 1/\nu$ is the shift exponent, b is a constant, and ν is the correlation-length exponent for the bulk system. The theoretically expected λ values are $1/0.7048 = 1.419$ and $1/0.6294 = 1.5884$ for the 3D Heisenberg [23] and Ising [24] models, respectively. Fitting the data for $t_{Cr} > 70$ Å by Eq. (1) is shown by the dashed line in Fig. 4, where $T_N(\infty) = 295$ K has its thick-film value. These data are well represented by $\lambda \approx 1.4 \pm 0.3$, which is in agreement with expectation from scaling theory. To fit the complete data set, we used the empirical expression

$$[T_N(\infty) - T_N(t_{Cr})]/T_N(t_{Cr}) = b\,(t_{Cr} - t_0)^{-\lambda'}, \qquad (2)$$

where $t_0 = 42.3$ represents the zero offset in the Cr thickness and $\lambda' = 0.8 \pm 0.1$ as shown by the solid line in Fig. 3. The sharp drop in the value of T_N near $t_{Cr} \sim 50$ Å and the nonuniversal value of λ' indicate the presence of an additional effect for the thinnest Cr spacers that we identify below as being due to spin frustration.

We need to understand the differences between the thin and thick Cr regimes and the relationship to isolated Cr films. For an ideal Fe/Cr(001) interface, $T_N(t_{Cr})$ should *increase* with decreasing Cr thickness due to the Fe-Cr exchange coupling since the Fe Curie temperature is much higher than T_N for Cr. This agrees with the experimental observations of Ref. [13]. In the present study, however, we find $T_N(t_{Cr})$ *decreases* for thin Cr layers. We believe that this behavior arises from spin-frustration effects in the vicinity of the rough Fe-Cr interfaces [25]. Such interfaces contain atomic steps as shown in the Fig. 4 inset and discussed in Ref. [15]. The interfacial exchange energy can be minimized only locally, and frustration of the interfacial spins will occur if the Fe and Cr magnetically order long range. For a superlattice, assuming random monatomic steps at both the Fe and Cr surfaces, 25% of the Cr layer will be frustrated at both interfaces, 50% will be frustrated at one interface, and 25% will match with the Fe layers. The value of T_N, therefore, should be influenced by a balance between the energy gained from long-range AF ordering of the Cr and the energy cost due to magnetic frustration at the Fe-Cr interfaces. For thin Cr layers, the frustration energy is sufficiently high to suppress long-range ordering of the Cr. As t_{Cr} increases, the system overcomes the

frustration energy and begins to order. The crossover thickness for the present samples is 42 Å of Cr.

Given that the Fe-Cr exchange is much stronger than the Cr-Cr exchange interaction, a more realistic picture would have the frustrated magnetic interface located *near* the Fe-Cr interface but within the Cr layers. Then the interfacial Cr atoms could be polarized by the Fe, and the magnetically "dead" Cr atoms (or nodes in the AF SDW) could be moved towards the interior of the Cr layer. Electronic structure calculations [26] of Fe/Cr/Fe trilayers determine that the energy cost in suppressing the Cr moment (at 0 K) is only 0.8 meV/atom, as compared to 200 and 80 meV/atom for Fe and Mn, respectively. Calculations of diffuse or stepped Fe/Cr interfaces demonstrate that the presence of frustrated Fe-Cr bonds can strongly suppress the Cr moment over extended distances [26–28]. Thus, Cr is highly sensitive to its local environment with local distortions being capable of causing strong moment reductions.

For the thicker Cr films ($t_{Cr} > 65$ Å) above T_N, the long-period oscillatory coupling energy decreases to a value of <0.01 erg/cm^2 and the observed interlayer coupling becomes dominated by the nonoscillatory biquadratic term. The surprising result that the biquadratic coupling disappears below T_N gives some insight into its origin. We consider two recent theories which involve extrinsic mechanisms for the biquadratic coupling [11,29]. The first results from fluctuations in the Cr thickness that average over the short-period oscillation in the coupling; 90° (biquadratic) alignment results from an energetic compromise between 0° and 180° orientations [11]. Below T_N if the Cr spins participate in long-range AF ordering, they must decouple from the rough interface with Fe and no longer respond locally to the thickness fluctuations. The second theory posits that paramagnetic "loose spins" *in* the spacer or *near* the interface mediate the coupling by being polarized by the RKKY exchange with the ferromagnetic layer [29]. Below T_N, these loose spins are frozen in the AF matrix and, therefore, no longer contribute to the coupling. The relative importance of the two mechanisms can, in principle, be assessed by characterizing the different functional forms they yield for the temperature dependence of the biquadratic coupling.

In summary, we have investigated the AF ordering of Cr layers in epitaxial Fe/Cr(001) superlattices. AF order is suppressed for $t_{Cr} < 42$ Å and is attributed to finite-size and spin-frustration effects. For $t_{Cr} > 42$ Å, T_N initially rises rapidly, and then asymptotically approaches the bulk value for the thickest spacers studied (165 Å) with a transition-temperature shift exponent characteristic of 3D Heisenberg or Ising models. Finally, the AF ordering of the Cr layers dramatically alters the interlayer coupling in the sense that the biquadratic coupling of the Fe layers observed for $T > T_N$ vanishes below T_N.

We thank J. Mattson and D. Stoeffler for helpful discussions. This work was supported by the U.S. Department of Energy, Basic Energy Sciences-Materials Sciences, under Contract No. W-31-109-ENG-38. One of us (A. B.) gratefully acknowledges support from the Alexander von Humboldt-Stiftung through a Feodor Lynen Research Fellowship.

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