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Spin-density-wave antiferromagnetism of Cr in Fe/Cr(001) superlattices

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The antiferromagnetic spin-density-wave (SDW) order of Cr layers in Fe/Cr(001) superlattices was investigated by neutron scattering. For Cr thicknesses from 51 to 190 Å, a transverse SDW is formed for all temperatures below the Néel temperature with a single wavevector **Q** normal to the layers. A coherent magnetic structure forms with the nodes of the SDW near the Fe-Cr interfaces. For thinner Cr layers, the magnetic scattering can be described by commensurate antiferromagnetic order.

Keywords: Fe/Cr superlattices, antiferromagnetic, spin density waves, neutron scattering

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I. Introduction

Fe/Cr(001) trilayers and superlattices are the prototypical coupled-layered magnetic system being the first to exhibit oscillatory interlayer coupling[1, 2], giant magnetoresistance (GMR)[3] and biquadratic interlayer coupling[4]. Although there has been over 10 years of work on this system, the nature of the magnetic ordering of the Cr layers and its influence on GMR and interlayer coupling are still not fully understood. Bulk Cr is an intinerant antiferromagnet (AF) which forms an incommensurate spin density wave (SDW) below the Néel temperature T_N=311 K.[5] The SDW is characterized by a wavevector **Q** determined by the nested feature of the <100> direction of the Cr Fermi surface. At high temperature, the Cr spin sublattices **S** are transverse to **Q** (**S**$\perp$**Q**), while below the spin-flip transition at 123 K **S** rotates 90° to form a longitudinal SDW with **S**$\parallel$**Q**.

Only recently have experiments focused directly the magnetic properties of the Cr layers in Fe/Cr superlattices.[6-8] Transport experiments on epitaxial, sputtered Fe/Cr(001) superlattices determined T_N values for relatively thick Cr spacers.[7] The Néel transition of the Cr spacers is suppressed for Cr thicknesses t_{Cr} < 42 Å. For >42 Å of Cr, T_N increases rapidly and asymptotically approaches the bulk value for thick Cr spacers. The magnetic properties of the superlattices are dramatically altered at T_N. For T> T_N, adjacent Fe layers are coupled at 90° resulting from biquadratic interlayer coupling. For T < T_N the biquadratic coupling is suppressed resulting in anomalies in the remanent magnetization, coercivity, and magnetoresistance in the vicinity of T_N as has been studied in detail by neutron reflectivity.[9, 10]

Perturbed-angular-correlation spectroscopy (PACS) measurements for Fe/Cr(001) superlattices grown by molecular beam epitaxy (MBE) indicate suppression of AF order for t_{Cr}<60 Å.[8] For t_{Cr}>60 Å, the PACS study reports (i) T_N > 500 K, which is significantly greater than that of the bulk, (ii) Cr moments that are enhanced over the bulk value, and (iii) Cr moments that are oriented *normal* to the layers and *perpendicular* to the Fe moments. The PACS spectra were

analyzed as arising from a longitudinal SDW order. Both the enhanced value for T_N and the perpendicular moments were attributed to strain.[8]

In the present paper, we highlight our recent quantitative neutron diffraction measurements of AF-SDW Cr ordering in Fe/Cr(001) superlattices. Magnetic scattering of neutrons from a transverse SDW with Q||(001) results in satellites at the Cr $(0,0,1\pm\delta)$ positions. The incommensurability, δ, is given by $1 - aQ/2\pi$, (a=2.884 Å is the Cr lattice constant) and is directly related to the nested wave vector. In bulk Cr, δ varies continuously from 0.037 at T_N to 0.049 at 10 K, corresponding to SDW periods (a/δ) of 78 and 59 Å, respectively. Thus, neutron scattering provides a powerful technique for characterizing the magnetic ordering within the Cr layer.

II. Experimental Procedures

Epitaxial Fe/Cr(001) superlattices were grown by dc magnetron sputtering onto single-crystal MgO(001) substrates. The Fe thickness was held constant at 14 Å for each sample and t_{Cr} was varied from 190 and 31 Å. Details about the growth and structural characterization are given in Ref. 9. Neutron diffraction measurements were performed on triple-axis spectrometers HB-2 and HB-1A at the HFIR reactor at Oak Ridge National Laboratory. The samples were mounted on the cold finger of a closed-cycle refrigerator which allowed diffraction measurement to be made from 12 - 300 K. Initial temperature-dependent measurements were made on the HB-2 spectrometer using a focused Si(111) monochromator and were published previously.[9] The present low-temperature measurements were made on the HB-1A spectrometer using a double crystal graphite (002) monochromator with a pyrolytic graphite filter to suppress harmonic contamination. A graphite (002) analyzer crystal was used for all experiments. Most scans were performed with the scattering vector normal to the layers. In this scattering geometry, the measurement is sensitive to in-plane moments which are ordered normal to the layers.

III. Experimental Results and Discussion

Shown in Fig. 1 are low-temperature neutron scans about the Cr (001) position for the 115-, 63-, 51-, and 31-Å samples. The dashed lines indicate the expected $(1+\delta)$ and $(1-\delta)$ satellites positions for bulk Cr. Symmetrically split satellite peaks observed about the Cr(001) position confirm that the Cr layers (for $t_{Cr} \geq 51$) form a transverse SDW with **Q** normal to the layers. No evidence of a longitudinal SDW is observed indicating that the spin-flip temperature has been suppressed at least to T<12 K, the lowest measurement temperature. For the 115-Å Cr sample, additional scans were performed with the scattering vector in-plane along the [100] and [010] directions. No magnetic scattering is observed which indicates that the Cr layers are in a single-**Q** configuration to better than one part in a thousand. This is consistent with measurements of single-**Q** configurations for Cr(001) films on Nb,[11] and for the near-surface regions of Cr single crystals.[12] For all samples, the Cr moments are in-plane is contrary to the PACS results on strained MBE-grown samples. The intensity of the satellite peaks decreases with increasing temperature until they are no longer observable at room temperature. This result differs from the PACS results which found T_N values well above room temperature.

Although the presence of SDW order can be inferred, the spectra exhibit a more complex dependence on Cr thickness than might be expected for simple SDW order. Quantitative analysis of the SDW ordering requires fitting the measured intensities to a model calculation which includes both the SDW ordering of the Cr layer as well as the superlattice structure. To accomplish this, we use a Hendricks-Teller approach which has proved successful in analysing x-ray and neutron scattering results from superlattices.[13, 14] The ferromagnetic-Fe and AF-SDW-Cr layers are described by the magnetic scattering factors:

$$F_{Cr}(q) = \sum_{n=0}^{N-1} (-1)^n p_{Cr} \sin\left(2\pi n d_{Cr}/P + \Phi\right) \exp(iqnd_{Cr})$$

$$F_{Fe}(q) = \sum_{n=0}^{M-1} p_{Fe} \exp(iqnd_{Fe}), \qquad (1)$$

where N (M) is the number of Cr (Fe) atomic planes with a lattice spacing of d_{Cr} (d_{Fe}) within a Cr (Fe) layer, and Φ and P are the phase and period of the SDW, respectively. The quantities p_{Cr} and p_{Fe} are the magnetic form factors for Cr and Fe, respectively. The scattering vector q equals $4\pi\sin\theta/\lambda$ where λ=2.351 Å is the neutron wavelength. Interfacial roughness is calculated by introducing 1-monolayer fluctuations in the Cr and Fe layer thicknesses. The scattered intensity is determined by ensemble averaging random fluctuations in the layer thicknesses, as outlined in Ref. 13. The calculated intensity is then convoluted with the measured instrumental resolution.

We least-squares fitted the measured spectrum. The fitting parameters are: (i) the layer thicknesses t_{Cr} and t_{Fe}, (ii) the Fe and Cr lattice spacings, (iii) the period and phase of the Cr SDW, and (iv) the ratio of the Fe to Cr moments. The most important parameters are the layer thicknesses, Cr lattice spacing, and the period and phase of the SDW. The thick solid lines in Fig. 1 are the results of the fitting procedure. This model reproduces the shift and splitting of the SDW peaks and quantitatively fits both the relative intensities and line widths of the experimental data. For the $t_{Cr} \leq 63$-Å samples, the observed structure in the peaks results from coherent scattering of adjacent Cr layers. For a superlattice, the true periodicity of the system is the repeat distance of the superlattice Λ, and, therefore, Bragg peaks are expected to be located at $q = na/\Lambda$ (in units $2\pi/a$). The SDW ordering within the Cr layers only modulates the intensities of these peaks, thus the SDW period cannot be determined directly from the peak positions, but quantitative analysis is required to fit the peak intensities.

The fitting parameters are given in Table 1, and the magnetic moment distributions determined from the fits are shown schematically in Fig. 2. A symmetric spin structure is formed in the Cr layers with the nodes of the SDW near the interfaces. Unlike T_N, which is strongly thickness dependent, the period of the SDW is independent of t_{Cr} and is in agreement with that for bulk Cr. This even holds for the 51-Å Cr sample which only supports a half-period of the SDW. This all suggests that strain or impurities are not driving the change of T_N in the present samples. Both strain and impurities alter the Fermi surface and dramatically alter T_N and the SDW period. For example, the strains induced in making a crushed Cr powder are sufficient to completely suppress the SDW order resulting in commensurate AF state with T_N=450 K. Since the SDW period in the present study is independent of thickness, this suggests that the changes in T_N arise from a combination of finite-size effects within the Cr layers and spin-frustration effects at the Fe-Cr interface as was previously proposed.[7]

The nature of the Cr ordering is quite different for the thinner layers. Although, previous magnetic and transport studies failed to detect a Néel transition for t_{Cr}<42Å, there still is magnetic scattering from the 31-Å sample. The scattering persists up to at least T=175 K (above T_N of the 51-Å Cr samples) and consists of two sharp peaks about the Cr (001) and a broad diffuse component centered near the Cr (001) reflection. Similar results are obtained for a 44-Å Cr sample. The scattered intensity is fitted assuming a commensurate AF structure. The splitting of the peaks results from the superlattice periodicity ($\Delta q=a/\Lambda$), and cannot be interpreted as arising from SDW order.

In summary, we have investigated the AF SDW order of Cr layers in Fe/Cr(001) superlattices by neutron scattering. For $t_{Cr}\geq$51 Å, we find a transverse SDW is formed for all temperatures below T_N with a single **Q** normal to the layers and the nodes of the SDW near the Fe-Cr interfaces. The observed SDW period is close to the bulk value. For the t_{Cr}=31 Å and 44 Å, the magnetic scattering can be described by a commensurate AF ordering. The crossover from commensurate to SDW AF order at $t_{Cr}\approx$45Å with increasing Cr thickness coincides with the onset

of the observed Néel transition of the Cr layers.[7] This suggests that nodes in the SDW near the Fe-Cr interface isolates the interior of the Cr layer from the frustrated Fe-Cr interfaces which allows homogeneous AF-SDW order and an independent Néel transition to develop for thicker Cr layers.

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Table 1: Fitted results for the [Fe (14 Å)/Cr (t_{Cr})]$_N$ superlattices shown in Fig. 1 measured at T < 30 K. The layer thicknesses were confined to ± 1 ML of those determined from x-ray diffraction. Samples marked with an asterick indicates parameters which could not be determined in the fitting proceedure and were fixed. The bulk value for d_{Cr}, d_{Fe} and the SDW period are 1.442, 1.433, and 59 Å, respectively. The 31-Å data were fitted assuming a commensurate AF structure.

nominal t_{Cr} (Å)	d_{Cr} (Å) (±0.003)	d_{Fe} (Å) (±0.03)	SDW period (Å)	Φ
31	1.440	1.43*	—	—
51	1.445	1.45	60 ± 5	2.2 ± 0.4
63	1.446	1.42	61 ± 5	-0.1 ± 0.3
115	1.445	1.44	58 ± 4	0.0 ± 0.5
190	1.446	1.44*	59 ± 3	0.0*

Figure 1: Neutron diffraction results for [Fe (14 Å)/Cr]$_N$ superlattices measured at 20 K. Cr thicknesses are listed to the left of the spectra. The spectra are offset and the scale factor to the right indicates the relative intensities normalized to the counting time and total Cr thickness. The open circles are the measured intensities, and the solid lines are calculated intensities described in the text. Parameters for the calculated intensities are given in Table 1. The vertical dashed lines indicate the expected (1+δ) and (1-δ) satellite positions for bulk Cr at 20 K.

Figure 2: Schematic representation of the magnetic moments for Fe and Cr layers determined from the fitting results of Fig. 1 and Table 1 for the 115-, 63-, and 51-Å samples. The filled circles indicate the Fe moments and the open circles the Cr moments.

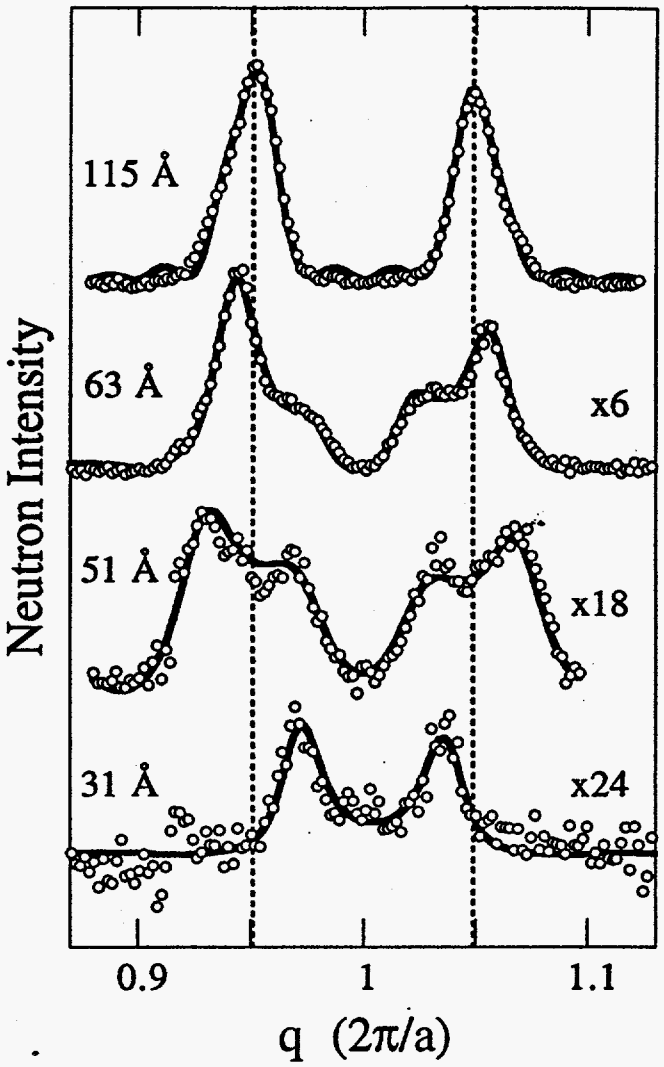

Fig. 1

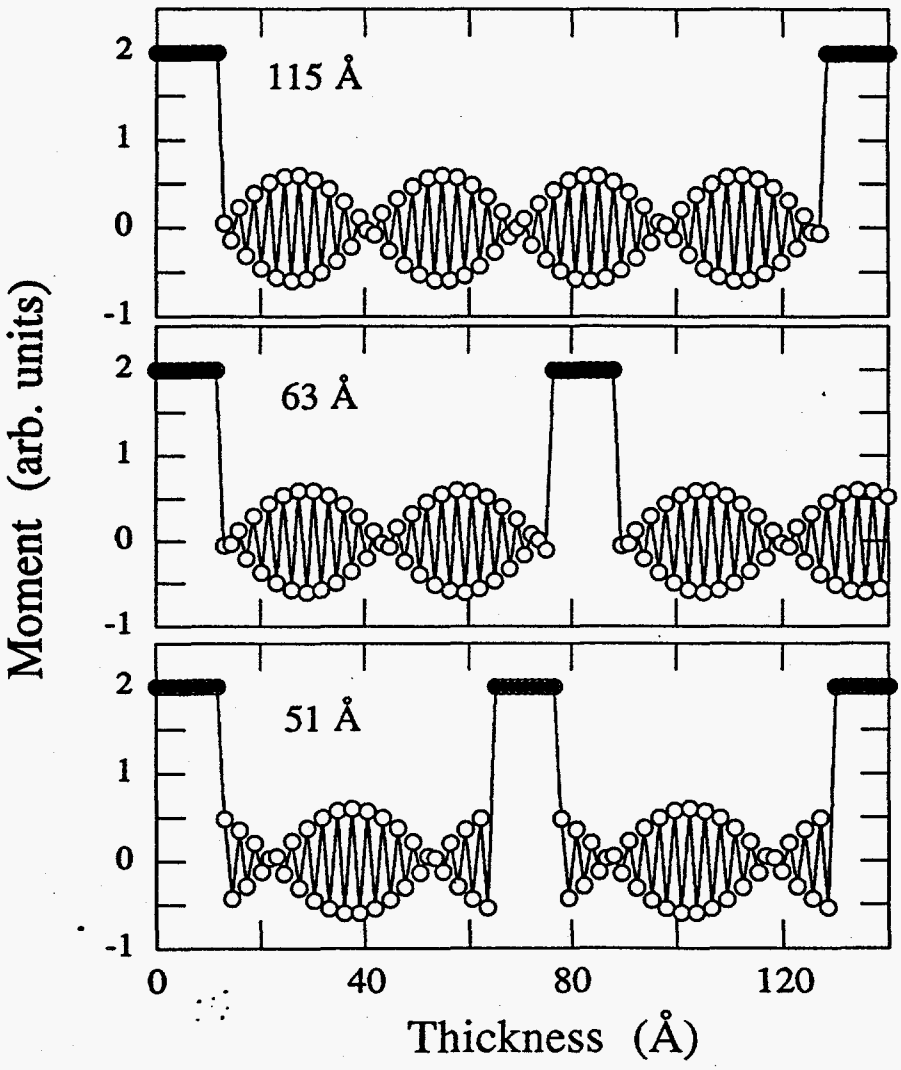

Fig. 2