Long-Range Incommensurate Magnetic Order in a Dy-Y Multilayer

M. B. Salamon,[1] Shantanu Sinha,[1] J. J. Rhyne,[2] J. E. Cunningham,[1] Ross W. Erwin,[2] Julie Borchers,[1] and C. P. Flynn[1]

[1]*Department of Physics and Materials Research Laboratory, University of Illinois at Urbana–Champaign, Urbana, Illinois 61801*

[2]*National Bureau of Standards, Gaithersburg, Maryland 20899*

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A multilayer sample of Dy(47 Å)/Y(40 Å), produced by molecular-beam-epitaxy techniques, is shown to order magnetically in an incommensurate helix that is coherent over several multilayer periods. The magnetic satellite peak has harmonics resulting from the multilayer modulation. The dc magnetic properties are suggestive of a metamagnet.

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The ability to produce artificial structures such as superlattices with atomic-scale dimensions is one of the most exciting recent developments in materials science. While many properties of superlattices are of fundamental and practical importance, most relate more to the structure of the thin layers than to coherent effects caused by the artificial modulation. Notable exceptions are folded phonon behavior in semiconductor multilayers and high-efficiency reflectors for x-ray and neutron scattering.

In this Letter we report the fabrication of high-quality multilayers of yttrium and dysprosium, thickness matched to the natural periodicity of the helical magnetic order of Dy. Neutron-diffraction data demonstrate for the first time the existence of long-range incommensurate magnetic order with Fourier components at the multilayer periodicity. The measured coherence length of magnetic order, greater than 300 Å, confirms that the helical periodicity of the Dy spins remains coherent across several bilayers. It is modulated, but not interrupted, by the intervening layers of "nonmagnetic" Y.

Dy has hexagonal-close-packed structure with $a = 3.593$ Å (basal plane) and with $c = 5.655$ Å; Y has $a = 3.650$ Å and $c = 5.641$ Å. The 2% lattice mismatch does not prevent the growth of coherent crystalline multilayers. Bulk Dy develops[1] incommensurate helical magnetic order of the $4f$ spins below its Néel temperature of 178 K, and has a ferromagnetic transition at 85 K, induced[2] by a combination of anisotropy and magnetostrictive interactions. In the helical state the wavelength increases smoothly from about 25 Å at the Néel temperature to about 40 Å just above the first-order collapse to the ferromagnetic state. We have produced Y/Dy multilayers with Dy nominal thickness near 40 Å and Y thicknesses between 12 and 40 Å. We restrict the results reported here to a sample of 64 bilayers of Dy(47 Å)/Y(40 Å). Elemental Y has no $4f$ electrons and can be considered as a nonmagnetic analog of Dy. Alloys of Dy and Y with as little as 5% Dy show only helical order down to 0 K and exhibit an initial period[3] at T_N of ≈ 20 Å in the dilute-Dy limit. This is close to the wavelength at which calculations of the generalized conduction electronic susceptibility $\chi(Q)$ show[4] a maximum for Y. We suggest that this enhanced $\chi(Q)$ for Q near the maximum found[5] for Dy is responsible for the phase coherence of the Dy spins across several superlattice periods.

The key to the growth of these superlattice structures is the discovery[6] that Y grows epitaxially on a Nb [110] surface, thereby avoiding the reactivity of rare-earth elements with the sapphire substrate. To produce these samples, 1500 Å of Nb was first grown on $(11\bar{2}0)$ sapphire at 1000 °C in an ultrahigh-vacuum molecular-beam-epitaxy chamber (base pressure $\leq 10^{-9}$ Torr). About 500 Å of Y and subsequently Y and Dy multilayers were grown over a 1-cm^2 area of the Nb [110] at about 320 °C at a rate near 0.5 Å/sec. The Y and Dy sources were temperature-controlled effusion cells with computer-controlled shutters. A mass spectrometer monitored the growth rate. *In situ* reflection high-energy-electron diffraction measurements gave long streaks and faint Kikuchi lines during growth, both evidence for high-quality, atomically flat growth planes. X-ray data taken after growth showed several superlattice sidebands, as expected for a square-wave modulation. The Bragg-peak widths of 0.1° to 0.2° and rocking-curve widths $< 0.5°$ are comparable to values for bulk crystals. The (0002) peak is at an average lattice constant of 5.6 Å. Subsequent α-step measurements gave total thicknesses of 0.30 μm of Dy and 0.26 μm of Y for an actual layer thickness of Y(40 Å)/Dy(47.4 Å).

Magnetic properties of the multilayers were measured in a SQUID magnetometer. They are distinctly different from those of Dy[1] and of a $Dy_{0.5}Y_{0.5}$ alloy.[3,7] Figure 1 shows magnetization data for a 2-kOe field oriented in the basal plane. The contribution of the substrate and Nb underlayer was obtained by the fitting of the higher-temperature susceptibility to a Curie-Weiss law [Fig. 1, inset (a)] with a Curie temperature $\theta_C = 165$ K and Curie constant, $C = 119.2$ emu/g·kOe, corresponding to $(12.4 \pm 1.7)\mu_B$/Dy atom. The values for pure Dy are 178 K and

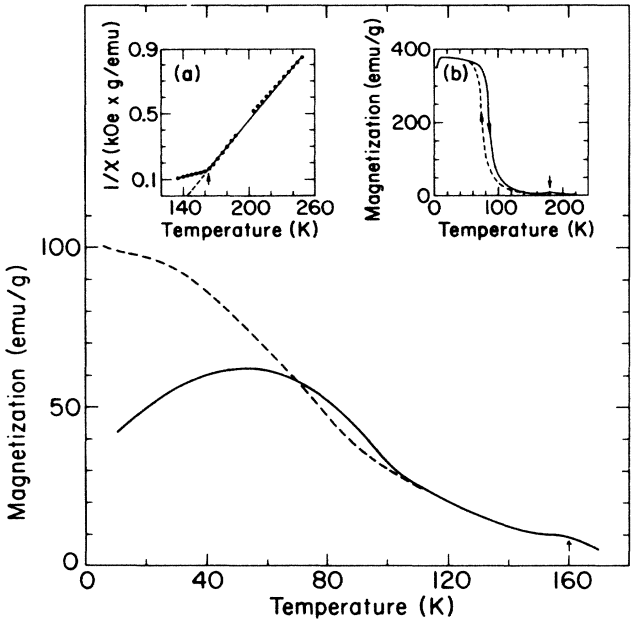

FIG. 1. Magnetization of Dy(47 Å)/Y(40 Å) superlattice at 2.0 kOe. The solid and dashed curves correspond to zero-field–cooled and field-cooled data, respectively. Inset (a): Inverse of susceptibility for the Dy(47 Å)/Y(40 Å) sample. An arrow marks the Néel temperature at the point where the data deviates from the best-fit line. Inset (b): Magnetization of a 2000-Å Dy film at 1.0 kOe prepared in an identical manner to that of the superlattice.

$10.64\mu_B$/atom. The susceptibility maximum near 160 K is similar to that in pure Dy, as shown in Fig. 1, inset (b), for a sample grown under identical conditions but with no Y interlayers. At low temperatures the behavior of the multilayer sample is nevertheless quite distinct from the Dy reference sample. There is no evidence of a first-order ferromagnetic transition. Instead, the sample shows strongly irreversible behavior with distinct values of the magnetization in field-cooled (see Fig. 2) and zero-field–cooled states reflecting a gradual spin-flop transition as a function of applied field. Since the neutrons show that long-range order exists and since there is no observable time dependence in either state, the system cannot be a spin-glass. Slope changes evident in the variation of M with H are similar to those observed in other rare earths that have intermediate "fan" states.

Neutron-diffraction studies were performed on 0.5 cm^2 of the sample in reflection geometry at the National Bureau of Standards reactor. A triple-axis spectrometer with a pyrolytic-graphite analyzer set was employed at an elastic-scattering position which significantly reduced the large incoherent inelastic background and improved the signal-to-noise ratio. The crystallographic c axis was perpendicular to the plane of the specimen, oriented along the scattering vector.

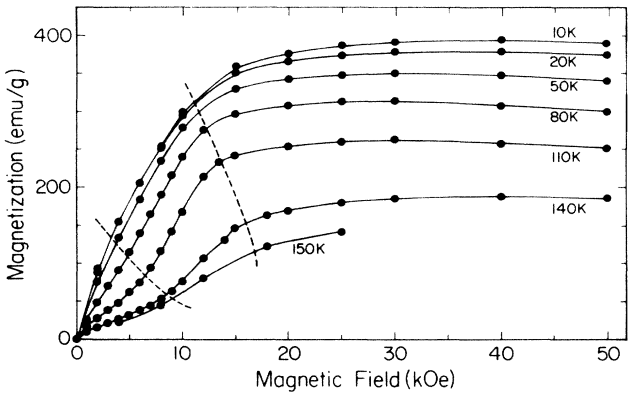

FIG. 2. Magnetization as a function of applied field for the field-cooled superlattice. The slope changes are connected by dashed lines.

Scans were perfomed along [000L], which is the direction associated with incommensurate magnetic order. Nuclear Bragg peaks, magnetic satellites, and harmonics induced by the multilayer periodicity were observed. Data were taken principally in the zone about the (0002) reflection at an incident neutron wave-

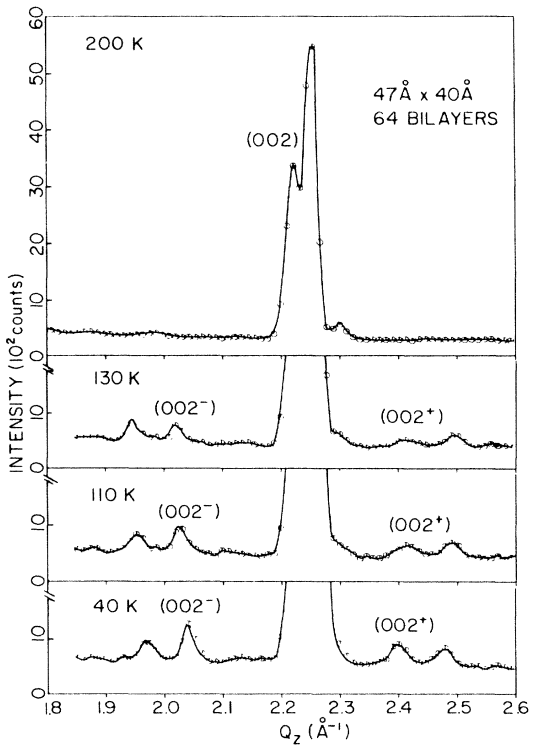

FIG. 3. Neutron-diffraction scans along (00L) direction for temperatures above and below $T_N \approx 160$ K. The principal magnetic satellites of the (002) nuclear reflection are marked (002^-) and (002^+). Other peaks represent the bilayer harmonics of both nuclear and magnetic reflections (see Fig. 4 and text).

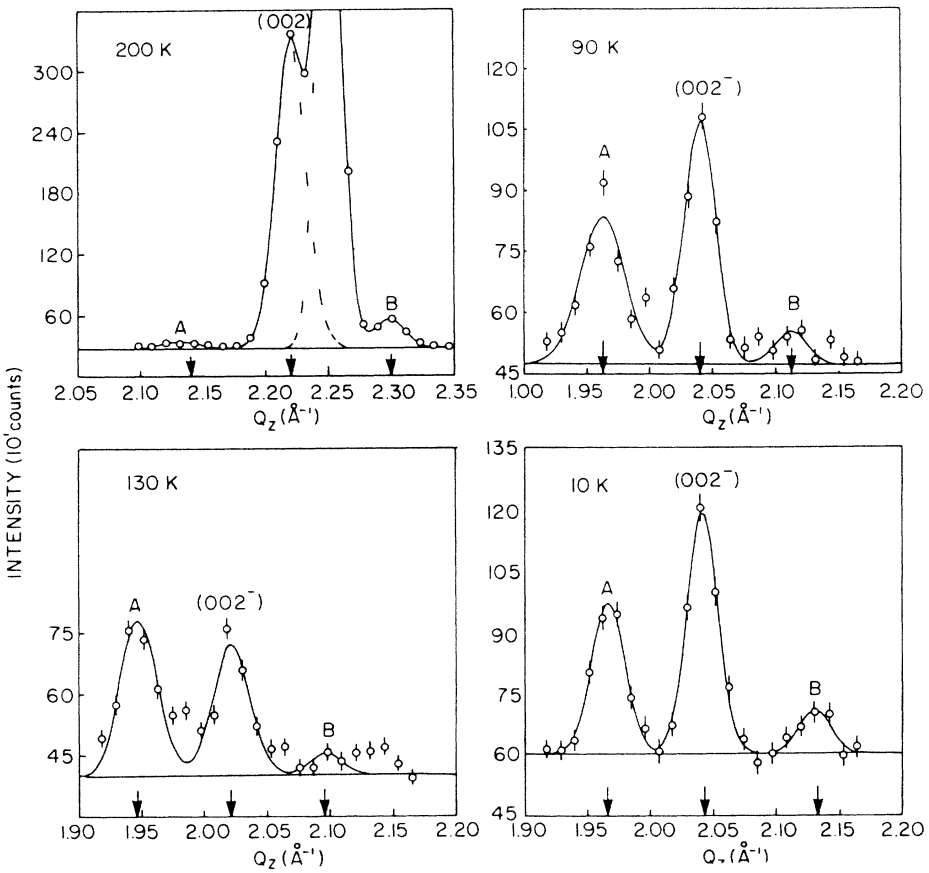

FIG. 4. Symmetric bilayer harmonic peaks (A and B) of the (002) nuclear peak shown at 200 K, and of the (002^-) magnetic satellites shown at 130, 90, and 10 K. All intensities are on the same scale.

length of 2.34 Å from a pyrolytic graphite (002) monochromator. Soller-slit collimators of 40′-20′-20′-40′ angular width were inserted before and after the monochromator and analyzer. A graphite filter suppressed half-wavelength contamination. Scans of the scattered intensity along the [000L] direction as a function of q are shown in Fig. 3 for temperatures of 200 (above $T_N \approx 165$ K), 130, 110, and 40 K. The magnetic reflections are stronger at low temperatures and move toward the (002) nuclear Bragg peak. Thus the order parameter and the spiral period increase as the temperature decreases. From fits of the data by Gaussian line shapes as shown in Fig. 4, it is observed that both the nuclear peak and the magnetic satellites exhibit harmonics at the modulation periodicity. These split symmetrically about the fundamental peak by the modulation wave vector $\Delta Q = 2\pi/\lambda_{\text{multilayer}} = 2\pi/(87\ \text{Å}) = 0.07\ \text{Å}^{-1}$. The intensities of the harmonics are apparently controlled by the lattice-parameter mismatch between the Dy and Y layers and by the magnetic and nuclear scattering amplitudes. The harmonics of the nuclear (002) peak are quite weak and compare well with the more precise x-ray data.[8] Since the intensity of the Bragg peak is independent of temperature, there can be no ferromagnetic component to the magnetic order.

The neutron data show a double peak at the (0002) position (see Fig. 4) which suggests the presence of a second, slightly mismatched crystal. Residual half-wavelength contamination and multiple Bragg scattering from the substrate are alternative explanations; no unambiguous identification was made. The magnetic satellites are symmetrically placed about the lower-Q Bragg peak. This appears to represent the principal c-axis reflection of the multilayers. The temperature dependence of the turn angle and the intensity of the (0002) magnetic satellite are plotted in Fig. 5. The magnetic intensity is not directly proportional to the square of the order parameter as in a bulk crystal because of modulation arising from the T dependence of the lattice mismatch. The turn angle (Fig. 5) decreases continuously with temperature; no ferromagnetic transition occurs above 10 K, in agreement with the magnetic measurements. At all temperatures the 47-Å thickness of one Dy layer remains greater than the wavelength of the magnetic helical ordering.

A measure of the coherence length of magnetic and atomic order in the c direction follows from the mag-

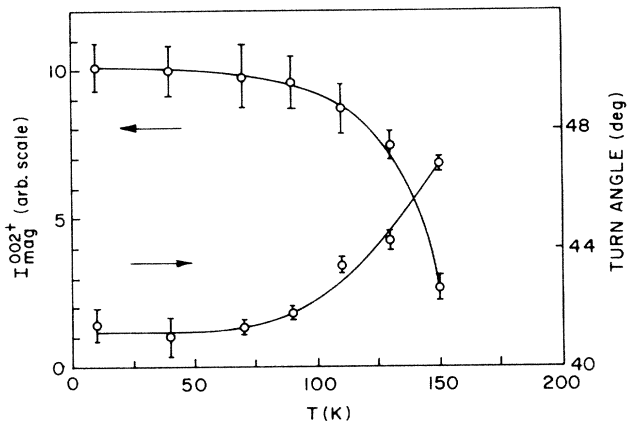

FIG. 5. Temperature dependence of the (002^+) magnetic satellite intensity and of the turn angle expressed in degrees per layer.

netic and nuclear peak widths. That the magnetic coherence remains over many periods is immediately obvious from Fig. 4 because the width of the magnetic satellites is significantly less than the spacing between each satellite and the harmonic from the 87-Å bilayer thickness. If the magnetic order were confined to single Dy layers, then the Q width of the central magnetic satellite peak would instead be comparable with the spacing between two harmonics. After deconvolution of instrumental broadening the (0002) Bragg peaks retained a width $\Delta Q = 0.012$ Å^{-1} reflecting the mosaic spread and c-axis angular wander through the 5000-Å thickness of the superlattice. The magnetic satellite peaks, on the other hand, have a larger intrinsic width of $\Delta Q \approx 0.020$ Å^{-1}, which corresponds to a coherence range of approximately 300 Å. This is convincing evidence that the Dy magnetic order is *not* interrupted at the layer boundaries or by the intervening Y layers, but instead exhibits phase coherence over at least four bilayers. The apparent limiting of the range of the magnetic order may arise from phase slips or incoherence of the spin coupling from one Dy layer to another.

In conclusion, the data suggest that the magnetic helical order is square-wave modulated as the conduction-electron spin-density wave passes between Dy and Y layers, but that it maintains coherence. The wavelength is set by the electronic properties of the multilayer structure. The helical state is evidently energetically weak since a relatively small applied magnetic field suffices to induce a "fan" or metamagnetic structure. Further neutron-scattering studies of this sample with an applied magnetic field are in progress together with measurements on samples in which the Dy and Y layers are of unequal thickness and in which the Dy thickness is less than the helical wavelength. We also plan to prepare similar samples using an intervening layer without a susceptibility peak at finite Q.

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