

Magnetic properties of Dy-Lu alloys

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Although many binary heavy rare-earth alloy systems have been studied extensively, there has been little work reported on the Dy-Lu system. The properties of single-crystal Dy_xLu_{1-x} films grown by molecular beam epitaxy are reported. SQUID magnetometer and neutron diffraction measurements on samples with $x=0.4, 0.5,$ and 0.6 show that the samples order helimagnetically with Néel temperatures of $T_N=90, 105,$ and 120 K, respectively. The helical turn angle was mapped as a function of temperature for each of the three alloys. Magnetic x-ray scattering, the first in an alloy, was observed at the $(002)^\pm$ positions at 15 and 60 K in the $x=0.4$ sample using resonant exchange scattering of synchrotron radiation at the Dy L_{III} edge.

Due to the similarity of their chemical properties, the heavy rare earths and yttrium are almost completely mutually soluble. In binary alloys, the concentration may be varied continuously to change such properties as the ordering temperature and lattice spacings.¹ Ordering temperatures and wave vectors for the heavy rare-earth binary alloys with each other and with Y have been shown to follow universal curves.² While many rare-earth alloys have been studied in the past, little work has been reported on the Dy-Lu alloy system. This system is of interest as part of our ongoing work on Dy/Lu superlattice structures.³

We have studied three c -axis samples grown by molecular beam epitaxy (MBE) at the University of Illinois Epicenter. The $Dy_{0.4}Lu_{0.6}$, $Dy_{0.5}Lu_{0.5}$, and $Dy_{0.6}Lu_{0.4}$ alloy films are 2300, 2400, and 20 540 Å thick, respectively. The films were grown on Y and Nb buffer layers upon a sapphire substrate and with Lu cap layers. Sample quality was verified by x-ray diffraction. The 2300- and 2400-Å films have in-plane mosaics of 0.3° and 0.4° , respectively. The thick (2.05- μ m) film has an in-plane mosaic of 0.1° . The c -axis coherence lengths of the films were 760 Å for the $Dy_{0.4}Lu_{0.6}$ film, 600 Å for the $Dy_{0.5}Lu_{0.5}$ film, and 1330 Å for the thickest film ($Dy_{0.6}Lu_{0.4}$).

In bulk Dy, the moments initially order at $T_N=179$ K in a c -axis helimagnetic structure; the moments are confined to the basal plane, and their direction rotates through a turn angle $\omega(T)$ in going from layer to layer along the c axis. The initial turn angle ω_i is 43° just below T_N , decreasing to 26.5° at $T_C=89$ K. Below T_C , bulk Dy is ferromagnetic. Bulk Lu is weakly paramagnetic. We will contrast the properties of Dy-Lu alloys with bulk Dy.

The magnetic moment was measured as a function of temperature using a SQUID magnetometer in zero-field cooled (ZFC) and field-cooled (FC) configurations. Cusps in the field-cooled susceptibility indicate that the Néel temperatures occur at 90, 105, and 120 K for the $Dy_{0.4}Lu_{0.6}$, $Dy_{0.5}Lu_{0.5}$, and $Dy_{0.6}Lu_{0.4}$ films, respectively (see Fig. 1). The Néel temperatures of the alloys follow the $2/3$ power of the effective spin parameter (de Gennes factor)

$$x = c(\lambda - 1)^2 J(J + 1), \quad (1)$$

where c is the concentration of the rare earth with angular momentum J , and λ is the Landé factor. Néel temperatures of many other rare earth-rare earth, rare earth-yttrium, and rare earth-lutetium binary alloys have been shown to follow this universal curve⁴ (see inset, Fig. 2). Consistent with previous results,⁵ which show a sharp suppression of T_C in heavy rare earths diluted with Y and Lu, no ferromagnetic transition was observed at temperatures above 10 K in any of the three Dy-Lu alloy samples.

The Néel temperatures were confirmed by neutron diffraction, and the films were shown to order helimagnetically (see Fig. 2). The magnetic coherence length along the c axes of the films is approximately the same as the structural c -axis coherence length at low temperatures. The intensity and position of the magnetic satellites were determined as a function of temperature for the three films. The separation Q of

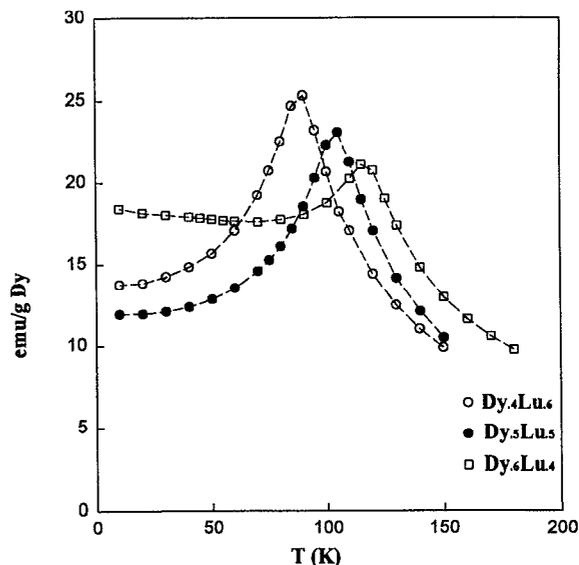


FIG. 1. Field-cooled SQUID magnetization data in a 5-kOe field. Cusps in the susceptibility indicate Néel temperatures of 90, 105, and 120 K for the $Dy_{0.4}Lu_{0.6}$, $Dy_{0.5}Lu_{0.5}$, and $Dy_{0.6}Lu_{0.4}$ alloys, respectively.

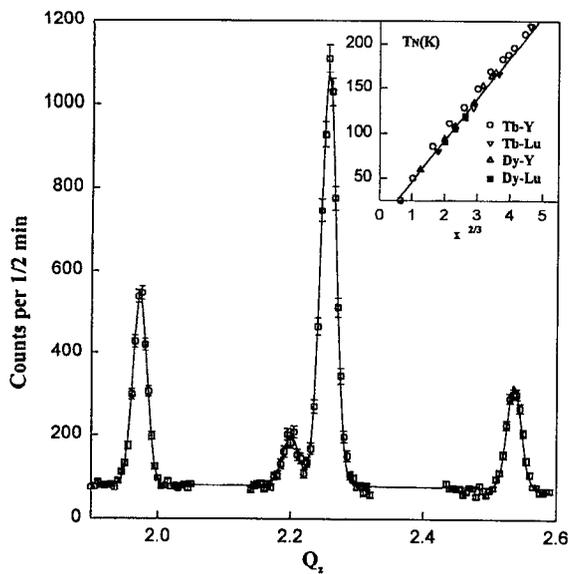


FIG. 2. Neutron scan along the c^* hcp axis of sample $\text{Dy}_{0.5}\text{Lu}_{0.5}$ at 30 K. The structural (002) Bragg peak at $Q_z = 2.26 \text{ \AA}^{-1}$ has magnetic satellites at 1.97 and 2.54 \AA^{-1} , indicating helimagnetic order. The structural peak at 2.20 \AA^{-1} is from the (002) Y buffer layer. *Inset.* Néel temperature as a function of the $2/3$ -power of the de Gennes factor for selected alloys. Tb-Y, Tb-Lu, and Dy-Y data are from Ref. 5. The data are seen to follow the curve given by $T_N = 46x^{2/3}$.

the magnetic satellites from the central (002) Bragg peak determines the turn angle per atomic layer. The turn angles $\omega = Qc/2$, where c is the lattice constant, for the $\text{Dy}_{0.4}\text{Lu}_{0.6}$ and $\text{Dy}_{0.5}\text{Lu}_{0.5}$ films varied with temperature from 47° at T_N to 45° at low temperature (see Fig. 3). The turn angle for the thicker film varied over a wider range; from 47.5° at the Néel temperature to 43.5° at low temperature. Previous work has shown similar results; Child⁵ *et al.* have found that the turn

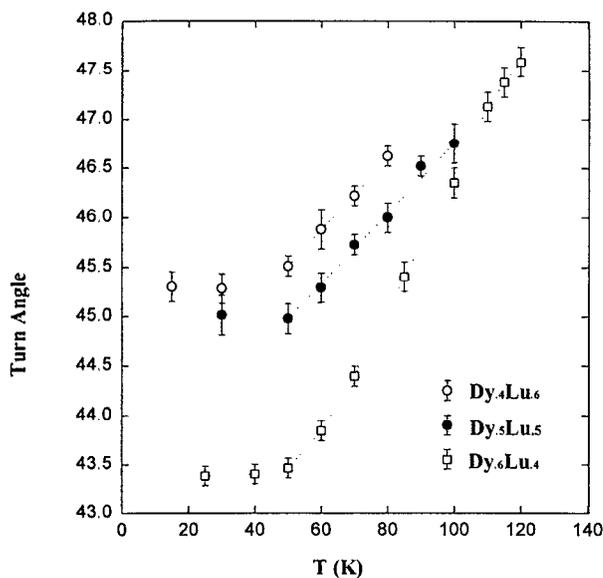


FIG. 3. Helimagnetic c -axis turn angle per atomic layer for each of the three alloys as a function of temperature. Lines through data points serve to guide the eye.

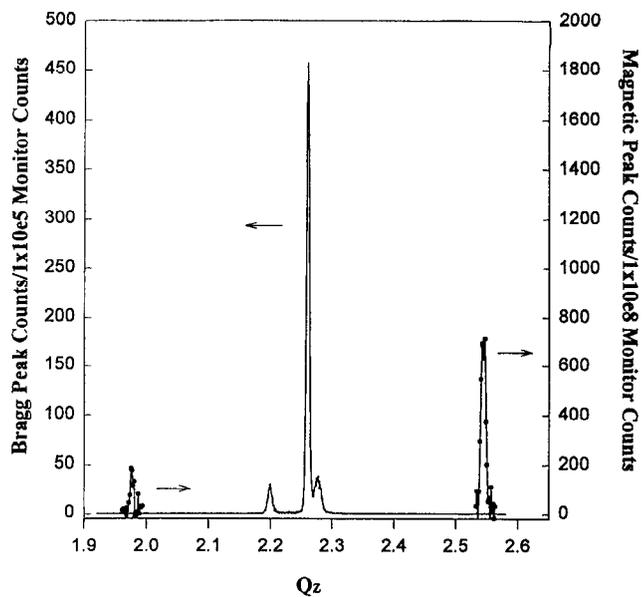


FIG. 4. Synchrotron x-ray diffraction data showing the Dy (002) Bragg peak ($Q_z = 2.26 \text{ \AA}^{-1}$) and x-ray resonant enhanced magnetic satellites ($Q_z = 1.974$ and 2.541 \AA^{-1}) of sample $\text{Dy}_{0.4}\text{Lu}_{0.6}$ at 15 K. The structural peaks at 2.20 and 2.27 \AA^{-1} are due to the Y buffer layer and Lu cap layer, respectively. Note the different scales for the nuclear and magnetic scattering.

angle initially decreased linearly with temperature from T_N in heavy rare earth-yttrium alloys, then remained constant at low temperatures. As the concentration of Y was increased, $\omega(T)$ varied over a smaller range, and at high Y concentrations approached 50° per layer at all temperatures below T_N . The Tb-Lu⁵ and Dy-Lu systems have qualitatively similar behavior to the rare earth-yttrium systems, although the limiting turn angle appears closer to 48° .

Synchrotron x-ray magnetic scattering, the first in an alloy, was observed from the $(002)^\pm$ magnetic satellites using resonant exchange scattering (XRES) at the Dy L_{III} edge.⁶ The $\text{Dy}_{0.4}\text{Lu}_{0.6}$ alloy was studied at the National Synchrotron Light Source at Brookhaven National Laboratory. The first-order harmonic was used to study the helimagnetic structure of the alloy at low temperature. A polarimeter was used to separate magnetic from charge scattering by exploiting the fact that the polarization of magnetically scattered radiation is rotated 90° relative to the incident polarization. Here, we label the major polarization component of the incident radiation (97%) σ and the orthogonal polarization π . Radiation scattered from the sample is diffracted a second time from the polarimeter analyzer crystal at a Bragg angle (θ) of 44.8° in order to select out a single polarization component.

The Dy $(002)^\pm$ magnetic satellites of the (002) Bragg peak were seen when scattering from the incident σ polarization into the detector π configuration. At 15 K, magnetic satellites are located at $Q(002)^+ = 2.541 \text{ \AA}^{-1}$ and $Q(002)^- = 1.974 \text{ \AA}^{-1}$ (Fig. 4), corresponding to a turn angle of 45.2° per atomic layer. At 60 K, the integrated intensity of the magnetic peaks has decreased by a factor of 0.7 from the 15 K data, and the turn angle is 45.7° per atomic layer. These results are consistent with the earlier neutron diffraction data

taken at NIST. At 120 K no magnetic satellites were visible. The satellites were not observed in the analyzer σ - σ scattering configuration at any temperature.

We have studied the magnetic ordering of $\text{Dy}_x\text{Lu}_{1-x}$ alloys using a variety of techniques. The Néel temperatures of the three alloys ($x=0.4, 0.5,$ and 0.6) and suppression of the Dy ferromagnetic transition are consistent with the behavior of other rare-earth alloy systems. A helimagnetic structure was confirmed by neutron diffraction, and the turn angle $\omega(T)$ was shown to have a qualitatively similar temperature dependence to Tb-Y, Tb-Lu, and Dy-Y alloys.⁵ Magnetic x-ray scattering was observed at the $(002)^\pm$ positions at 15 and 60 K in the $x=0.4$ sample using resonant exchange scattering of synchrotron radiation at the Dy L_{III} edge.

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¹J. Jensen and A. R. Mackintosh, *Rare Earth Magnetism: Structures and Excitations* (Oxford University Press, New York, 1991), p. 247.

²W. C. Koehler, *Magnetic Properties of Rare Earth Metals*, edited by R. J. Elliott (Plenum, New York, 1972), p. 94.

³R. S. Beach, J. A. Borchers, A. Matheny, R. W. Erwin, M. B. Salamon, B. Everitt, K. Pettit, J. J. Rhyne, and C. P. Flynn, *Phys. Rev. Lett.* **70**, 3502 (1993).

⁴B. Coqblin, *The Electronic Structure of Rare-Earth Metals and Alloys: The Magnetic Heavy Rare Earths* (Academic, New York, 1977), p. 83.

⁵H. R. Child, W. C. Koehler, E. O. Wollan, and J. W. Cable, *Phys. Rev. A* **138**, 1655 (1965).

⁶E. D. Isaacs, D. B. McWhan, D. P. Siddons, J. B. Hastings, and D. Gibbs, *Phys. Rev. B* **40**, 9336 (1989).