Positive Giant Magnetoresistance in Dy/Sc Superlattices

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We have discovered large positive magnetoresistance in Dy/Sc superlattices at low temperatures. These and other magnetotransport phenomena lack the hysteresis of the observed Dy magnetization. We offer a speculative interpretation in terms of interfacial reflectivity.

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In this Letter we describe measurements of large *positive* magnetoresistance (MR) in thin film systems. Large negative effects have previously been reported for transition metal multilayers [1] and for granular alloys [2]. They have been interpreted as the consequence of either (1) spin-dependent potentials that scatter itinerant magnetic charge carriers at interfaces between the different materials that form the composite system [3]; or (2) the spin-dependent density of states at the Fermi level [4]. Interfacial magnetism and scattering by interfacial states have also been considered [5]. A variety of mechanisms are indeed possible in the transition metals because the same carriers are responsible for both magnetism and charge transport. This is not the case for rare earths, where the magnetism derives mainly from $4f$ states, while transport is associated with $6s$ and $5d$ conduction orbitals. The materials employed here are the rare-earth dysprosium and the nonmagnetic metal scandium, which is rare-earth-like in its conduction properties. The small MR effects observed in superlattices (SLs) of the rare earth and Y (Lu) [6] indicate the weak spin-dependent scattering potential for the conduction states of the rare earth. We report here that a substitution of the nonmagnetic interlayer in the SLs by Sc causes large MR effects.

The MR effects in the Dy/Sc SLs were measured with the current along the *a* axis (in the growth plane) and the magnetic field along various crystal orientations. It is remarkable how well the SLs conduct [7]: 4000 Å of SL conducts as well as 2000 Å of Nb and Sc buffers. Corrected residual resistance ratios between 40 and 100 in the SLs confirm that the structures are notably defect-free. All the SLs studied here exhibit similar field and temperature dependences, so that the behavior is not highly sensitive to the Sc layer thickness. Large positive MR effects were observed in the SLs at low temperatures. Typical behavior at 10 K is shown for $(Dy_{20\,Å}/Sc_{40\,Å})_{65}$ in Fig. 1(a), together with anisotropy (inset) and the measured magnetization [Fig. 1(b)]. All resistivity results presented here are corrected for the buffer layer contribution (see below). No corrections were made to other transport coefficients reported here.

The behavior of resistance, MR, and magnetization shown in Fig. 1 leads immediately to two important conclusions. First, the strong hysteresis of the magnetization and its complete absence in the MR provide a compelling proof that the two phenomena arise from separate causes. Thus the giant MR cannot be caused by the spins in the magnetized Dy layers. Therefore the interactions between the dominant magnetic electrons and the dominant carriers of the present structures *cannot* explain the magnetotransport. Second, the high conductivity and residual resistance ratio of the SL [7], combined with the absence

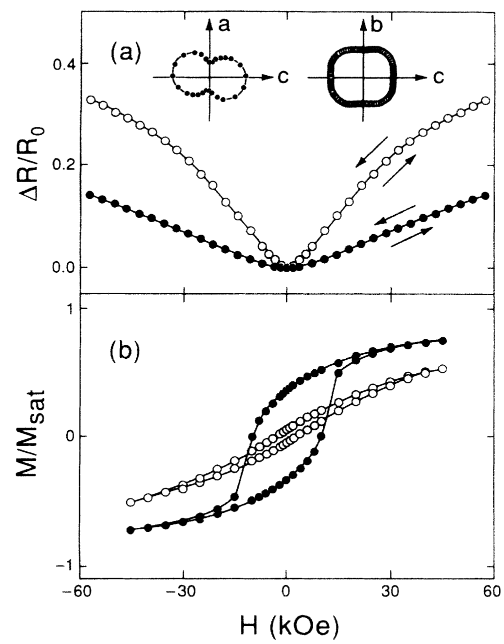

FIG. 1. Typical MR and magnetization for $(Dy_{20\,Å}/Sc_{40\,Å})_{65}$ at 10 K. (a) Open circles, transverse MR with field along the *c* axis perpendicular to the current along the *a* axis; filled circles, longitudinal MR with field parallel to the current along the *a* axis. Insets: polar plots of $\Delta R/R_0$ with a 57 kOe field applied along various orientations in the *a*-*c* plane (left) and in the *b*-*c* plane (right) and with the current along the *a* axis. R_0=0.97 $\mu\Omega$ cm. Note that the buffer layer conductances were subtracted out and that the raw MR before the subtraction has roughly the same field dependence and is slightly smaller (e.g., at 10 K the raw transverse and longitudinal MR are 22% and 12%); note also that the MR of the buffer layer is quadratic in field which corresponds to the cyclotron motion of electrons [13] and is about several percent at low temperatures. (b) Magnetic hysteresis curves for fields applied along the *c* axis (open circles) and in the basal plane (closed circles). M_{sat} is about 350 emu/g.

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of Dy scattering, lead us to conclude that the transport is channeled down the thin Sc layers. In order to achieve this conduction, Sc carriers must reflect off the interfaces largely without change of parallel momentum.

SLs used in this research were grown along the hcp c axis by molecular beam epitaxy (MBE) techniques on $(11\bar{2}0)$ sapphire substrates with buffer layers of (110) Nb and (0001) Sc. X-ray analysis revealed structural coherence lengths along the [0001] growth direction of about 500 Å and interdiffusion limited to about 3–4 interfacial atomic layers. Samples with nonmagnetic Sc layer thicknesses of 20, 30, 40, and 60 Å were prepared with typical Dy layer thickness of ~20 Å. Details of the sample growth, structure, and magnetic properties are given elsewhere [9]. The magnetic behavior of interest here may be summarized as follows. Helimagnetic order, which occurs in bulk Dy below 180 K [8], was not observed in any of the SLs. Instead, an apparently second order ferromagnetic transition occurs in individual Dy layers at a typical Curie temperature near 150 K. This is much higher than the first order transition at $T_C=85$ K in the bulk material. The enhancement of T_C arises from compression of the Dy basal planes by the Sc layers [10]. Owing to the confinement of magnetization within each Dy layer, the ferromagnetic coherence length along the growth direction, measured by neutron scattering, is equal to the Dy layer thickness. Weak interlayer coupling of this type may be attributed to the absence of nesting Fermi surface features in the SL.

The Dy-Sc alloys were prepared by vacuum annealing Dy/Sc SLs at 650 °C for 10 min at a pressure below 5×10^{-10} torr. X-ray diffraction experiments on the annealed alloys indicate crystal quality comparable to that of homogeneous alloys prepared by codeposition [10], and structural coherence > 500 Å along the c axis.

Samples were prepared for magnetotransport measurements using standard lithography techniques, including inert Ar beam etching, to pattern the metallic film on its sapphire substrate into 0.5 mm wide current channels and voltage terminals. Care was taken to avoid contamination or oxidation of the reactive rare-earth material during sample preparation. Neither structural and compositional analysis, nor resistivity measurements, indicate any deterioration of the sample quality during postgrowth procedures. Resistivity measurements were made by standard dc four-terminal techniques. Conductance contributions from buffer layers of Nb and Sc were subtracted using measured values for Nb and Sc films identical to those in the SLs. Thermopower measurements were carried out using calibrated fine Cu wires with known thermopower as electrodes, and constantan-chromel differential thermocouples. The temperature difference was maintained at about 1 K by a heater attached to one end of the 15 mm long and 2 mm wide sapphire substrate, with the other end thermally anchored to the Cu block of the cryostat. Experimental details will be provided in a more complete publication.

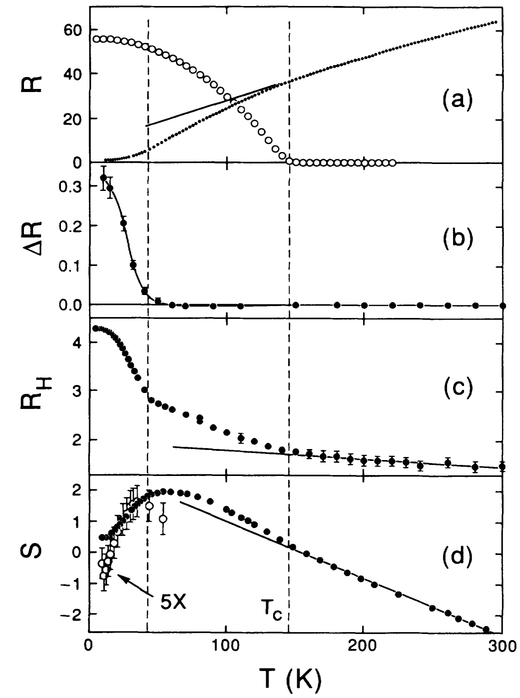

FIG. 2. Typical temperature dependence of the transport properties for $(Dy_{20\,Å}/Sc_{40\,Å})_{65}$. (a) Dots, zero-field resistivity in units of $\mu\Omega$ cm (the residual resistance ratio is ~60); open circles, basal plane magnetization in relative units. (b) The difference between resistances at 57 kOe transverse field and at zero field in units of $\mu\Omega$ cm. (c) Hall coefficients in units of 10^{-10} m^3/C. The field was applied along the c axis, perpendicular to the current along the a axis. The contribution from the buffer layers is about 0.6×10^{-10} m^3/C. (d) Filled circles, Seebeck coefficient in μV/K; open circles, difference between thermopowers at a 57 kOe transverse field and at zero field in units of 0.2 μV/K. Vertical dashed lines indicate the onset of the strong low temperature magnetotransport effects (left), and the onset of the high temperature transport effects at T_C. Above T_C, the transport effects vary linearly with T (lines), owing to diffusive phonon scattering.

Temperature dependences of the transport properties are shown in Fig. 2 for $(Dy_{20\,Å}/Sc_{40\,Å})_{65}$. Here we compare the MR in Fig. 2(b) with the Hall coefficient in Fig. 2(c) and the thermopower in Fig. 2(d), using the magnetization and resistance in Fig. 2(a) to define the physical regimes. The behavior shown in Fig. 2 can be divided into the three temperature regions indicated by vertical dashed lines. Above T_C, the transport behavior is linear in temperature, which indicates that diffusive phonon scattering effects are dominant. The resistivity R changes at a rate of about 0.16 $\mu\Omega$ cm/K, which is comparable to values for most heavy rare earths [8]. The Hall effect and thermopower in the SL exhibit opposite signs which indicate the complexity of the transport processes, and the presence of at least two carrier bands, with holes being the more mobile carriers, as indicated by the positive Hall coefficient. This is the opposite of both bulk Dy and Sc [8,11,12]. Moreover, the temperature dependence of

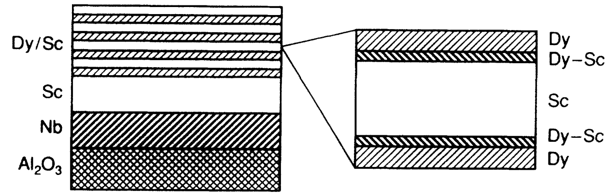

FIG. 3. Sketch of the Dy/Sc SLs, with the enlarged section to the right indicating the Dy-Sc alloyed layers on both sides of each Sc layers.

the thermopower fails to reflect the temperature dependence of the bulk materials [8]. This again points to a conduction band of the SL that differs from that of the bulk constituent materials.

A distinct regime of large transport anomalies can be identified at temperatures below about 40 K. There, the magnetization is saturated and the resistivity reaches its characteristic low temperature behavior, comprising summed residual and T^4 components. Both the MR and the Hall coefficients exhibit striking increases in this regime, the former reaching 0.3 $\mu\Omega$ cm at 57 kOe and 10 K. Both the normal and magnetic thermopowers pass through a maximum near 40 K and decrease markedly at lower temperatures, with a weak minimum near 12 K.

In between the diffusion regime at high temperature and the anomalies below 40 K lies a regime in which the transport properties reflect the temperature dependence of ferromagnetic order. Departures from the high temperature trend may indicate that spin disorder scattering plays a role, although the small negative MR demonstrates that the effect is relatively weak, and the strong magnetization-dependent Hall effect observed in Dy [8,11] is absent in the SL [12]. As mentioned above, other rare-earth SLs we have examined yield small magnetotransport effects [6], like the bulk materials, and lack the anomalies reported here for Dy/Sc.

We now return to the giant MR and the low temperature regime that provide the main topic of this Letter. There is no evidence to suggest that cyclotron motion of conduction electrons [13], for unknown reasons, simply causes unusually large effects with unexpected field dependence. Instead, we advance a speculative interpretation of the results based on the unusually large conductivity of the Sc layers [7], which assures a mean free path many times greater than the Sc layer thickness. Under these circumstances the carriers reflect many times from the interfaces before scattering, in a manner made familiar by thin film studies [13–17] and this enhances the sensitivity of resistance to momentum loss upon reflection. The extra resistance is [13,17]

$$R \sim \frac{1-p}{1+p} \sim \frac{1-p_0}{1+p_0}\left[1-\frac{2\Delta p}{1-p_0^2}\right], \qquad (1)$$

with $p=1$ for specular reflection and $p=0$ when all parallel momentum is lost. The last term shows how small changes Δp are enhanced when p_0 is close to 1. Our proposal is that an applied field may decrease the specular reflectance to cause a large positive MR.

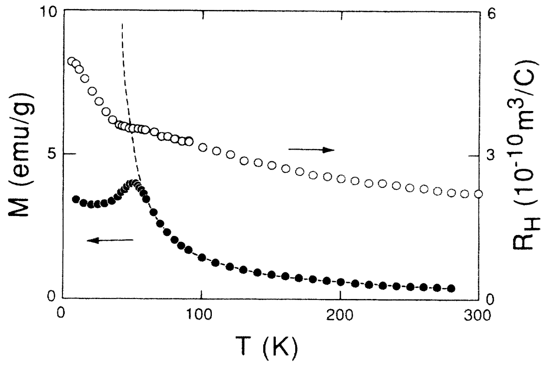

FIG. 4. Temperature dependences of magnetization (filled circles) and Hall coefficient (open circles) for 40 at.% Dy alloyed in Sc. The basal plane magnetization at 500 Oe exhibits a cusp at 43 K indicating the onset of antiferromagnetic order. The dashed line is a Curie-Weiss fit of the high temperature behavior with a paramagnetic Curie temperature of 33 K. The Hall coefficient is nearly identical to that of the Dy/Sc SL shown in Fig. 2(c).

The magnetic measurements indicate that about 10% of the Dy spins are missing from the Dy layer susceptibility. We speculate that they are contained in interfacial alloy and that this alloy causes the field-induced change of interfacial reflectivity (Fig. 3). To explore this possibility we have interdiffused SL samples and determined the field dependent magnetizations and magnetotransport behavior of the resulting alloys. As illustrated in Fig. 4, a typical 40% Dy alloy has $T_N=43$ K, its magnetization at 10 K shows negligible hysteresis [Fig. 5(a)], and the Hall effect in Fig. 4 is similar to that of the SLs (Fig. 2).

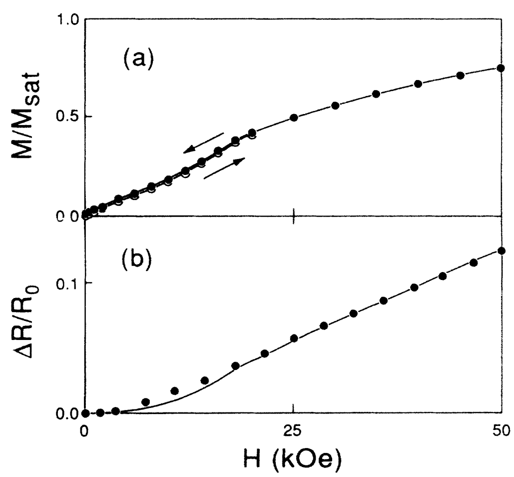

FIG. 5. (a) Basal plane magnetization for 40 at.% Dy alloyed in Sc at 10 K, used to model the interfacial moment for the MR shown in (b), with $\alpha/(1-p_0^2)=-0.13$. (b) Longitudinal MR of a Dy/Sc SL at 10 K (filled circles) and the model fit (line) discussed in the text.

Most important, the alloy exhibits a weak positive MR ~3% at 10 K. While the system is too poorly characterized for quantitative prediction it remains clear that the fractional alloy magnetization $\mu = M/M_{sat}$ must be the parameter relevant to the reflectance; p can depend only on μ^2, so that $p = p_0 + \alpha\mu^2 + \cdots$ and $\Delta p = \alpha\mu^2$ in Eq. (1), to the neglect of higher terms. Here, α may depend on orientation but not on μ.

We have fit $\Delta p = \alpha\mu^2$ to the longitudinal MR of Fig. 1 to obtain the result $\alpha/(1-p_0^2) = -0.13$. If the MR of 3% in the alloy causes a 3% decrease of interfacial reflectance, the required enhancement factor is just $(1-p_0^2)^{-1} = 4$, which gives $p_0 = 0.9$. This is certainly consistent with the observed high conductivity [7]. Also, comparable results for other SLs are assured by the similarities among the data. While the mechanism described here thus remains speculative, the description it affords for the results appears reasonable, and efforts to pursue these processes further appear warranted.

In summary, we report magnetotransport measurements on Dy/Sc SLs grown by MBE. The result of central interest here is the observation of a *positive* giant MR that lacks the hysteresis of the measured SL magnetization. A speculative explanation is that the MR originates in a decrease of interfacial reflectivity caused by magnetization of the interfacial alloy. Future research in which the SLs have conducting strata spaced by controlled alloy layers instead of Dy offers a clear and interesting opportunity for more quantitative modeling. In a larger context, the use of thin film conductance changes stimulated by carrier reflectances perturbed by surface phenomena of various kinds may offer timely applications to other scientific problems.

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