Exchange springs in antiferromagnetically coupled $DyFe_2$-YFe_2 superlattices

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Molecular beam epitaxy methods have been used to grow single-crystal Laves phase $DyFe_2$-YFe_2 superlattice samples with a (110) growth direction. In this paper, magnetization curves for the multilayer samples $[w\mathrm{DyFe_2}/w\mathrm{YFe_2}]\times 40$, with $w=50$, 75, 100, 150, and 300 Å, are presented and discussed. Below T_C, the magnetic moments of the $DyFe_2$ layers are coupled antiferromagnetically to those of the YFe_2 layers. However, in applied fields, it is possible to generate magnetic exchange springs in the YFe_2 layers, with well-defined bending (onset) fields B_B. In particular, it is demonstrated, both experimentally and theoretically, that the bending field B_B is closely proportional to $1/w^2$, where w is the thickness of the magnetically soft YFe_2 layer.

I. INTRODUCTION

In recent years, research into magnetic exchange springs has received fresh impetus, because it holds out the promise of increasing $(BH)_{\mathrm{MAX}}$ in permanent magnet applications.[1–4] For example, it has been argued that it should be possible to produce magnets with a giant energy product $(BH)_{\mathrm{MAX}}$ of 120 MGOe, by exploiting the exchange-spring mechanism in nanostructured oriented magnets.[3,4] In general, this research has been directed towards sputtered compounds, such as $SmCo_5$ and Fe, where the hard and soft magnetic layers are coupled ferromagnetically. Such a combination enables the magnetic designer to increase the net magnetization, while maintaining the coercivity associated with the hard layers.

In this paper, we report magnetic measurements on antiferromagnetically coupled hard and soft multilayer samples, grown epitaxially with nanometer control. While antiferromagnetic coupling leads naturally to a decrease in the maximum energy product, it does permits interesting applications. These include engineering magnetic compensation points (in a digital fashion), increasing coercivity, and fabricating magnetic exchange springs. In this paper, we present results for molecular beam epitaxy (MBE) grown, antiferromagnetically coupled, $DyFe_2$-YFe_2 superlattice films. In zero field, the larger Dy moments are coupled antiferromagnetically to the weaker Fe moments, and hence to their Fe counterparts in neighboring YFe_2 layers. However, this is not necessarily the case in applied fields above a critical value, known as the bending field B_B. For fields in excess of B_B, model exchange springs can be set up in the YFe_2 layers, whose properties are dictated by the thicknesses of the component layers. A schematic drawing of an exchange spring in a single YFe_2 layer can be seen in Fig. 1.

II. SAMPLE DETAILS, CRYSTAL GROWTH, AND MEASUREMENT

The two magnetic layers in question, $DyFe_2$ and YFe_2, belong to the cubic Laves O_h^7-$F3dm$ $MgCu_2$-type intermetallic compound.[5,6] They are both characterized by high Curie temperatures in excess of 600 K, as a result of strong Fe-Fe magnetic exchange. In addition, they also exhibit relatively strong RE-Fe magnetic exchange fields ~100 T (~100 K), which gives rise to antiferromagnetic (ferromagnetic) coupling in the heavy (light) RE compounds, respectively. Finally, the direction of easy magnetization is controlled primarily by crystal-field interaction at the RE ion.[7]

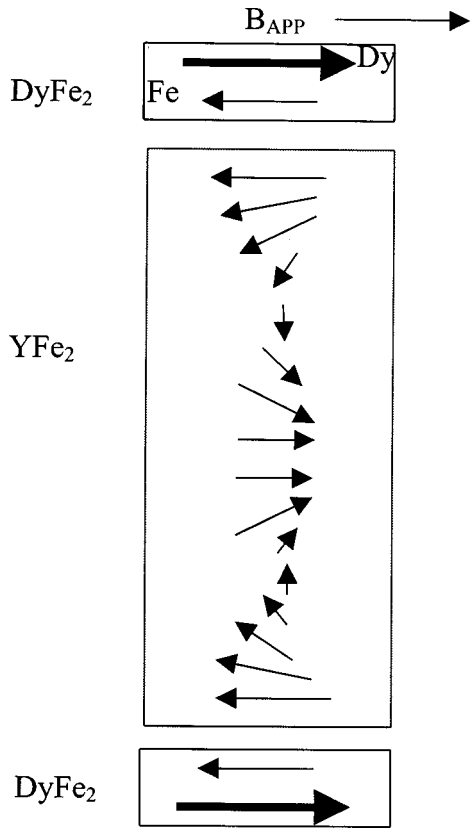

FIG. 1. A schematic drawing of an exchange spring in a YFe_2 layer.

For bulk $DyFe_2$ the easy direction of magnetization is [001] at all temperatures, whereas for YFe_2 it is [111]. But since the magnetic anisotropy of YFe_2 is very much weaker than that of $DyFe_2$, the YFe_2 layers can be considered as magnetically soft. Thus the direction of easy magnetization in the samples is likely to be dictated by the properties of the $DyFe_2$ layers. Finally, it should be noted that the easy direction of magnetization in an MBE grown (110) $DyFe_2$ film differs from that of the bulk, because of strain induced during crystal growth. Nevertheless, at low temperatures the direction of easy magnetization in MBE $DyFe_2$ reverts to [001].[8,9]

All of the above-mentioned features have recently been used to engineer a magnetic compensation point in a [25-Å $DyFe_2$/50-Å YFe_2]×40 sample, with relatively thin individual layers.[9] Because of the strong Fe-Fe exchange fields, all the Fe atoms, in both the $DyFe_2$ and YFe_2 layers, are constrained to be parallel to each other. So it is possible to balance the net magnetic moments associated with the Dy and Fe atoms, in a digital fashion.

In this paper, we present results for [$w$$DyFe_2$/$w$$YFe_2$] ×40 samples, with $w=50$, 75, 100, 150, and 300 Å. In these superlattices, exchange springs can be set up in the magnetically soft YFe_2 layers. This leads to an increase in the saturation magnetization, in applied fields greater than a critical field. We shall refer to this field as the bending field B_B, after Ref. 10.

The $DyFe_2$-YFe_2 superlattice samples were grown by molecular beam epitaxy (MBE) techniques using the Balzers UMS 630 UHV facility at Oxford, following a procedure described in Ref. 8. The samples were grown on epi-prepared $(11\bar{2}0)$ sapphire substrates with a 1000-Å (110) Nb buffer and a 30-Å layer of Fe. The $w=50$-Å sample only was grown with an additional YFe_2 seed layer to facilitate crystal growth, as reported in Ref. 9. However, no significant improvement in quality was observed and the additional YFe_2 layer only serves to confuse the magnetic moment analysis. The Laves phase compounds were grown by co-deposition of elemental fluxes at a substrate temperature of 400 °C in (110) orientation and with the major axes parallel to those of niobium.[9] Because the bulk lattice parameters for $DyFe_2$ and YFe_2 are 7.325 and 7.363 Å, respectively, the mismatch at the multilayer interfaces is only ~0.5%. *Ex situ* the samples were analyzed by x-ray-diffraction techniques, to confirm their single-crystal nature and check the bilayer repeat distance.

The magnetic measurements reported here were made using an Oxford Instruments Aerosonic 3001 vibrating sample magnetometer (VSM). The temperature range covered was 10–300 K, in applied fields of up to 12 T. Typical in-plane magnetization curves for fields applied along the [001] axis can be seen in Figs. 2(a)–(c). Note that the magnetization is given in terms of magnetic moment per formula unit, which we define as $Dy_{0.5}Y_{0.5}Fe_2$ for all the samples studied.

It will be observed that there is a small jump in the magnetization, in all the magnetization curves, as the magnetic field passes through zero. This step could be due either to (i) nonepitaxial layers at the bottom of the sample, (ii) slight misorientation of the sample, or (iii) contamination of the sapphire substrate, which could occur during sample shaping, with a diamond/steel cutting wheel. Nevertheless, this annoying feature does not negate any of the principle conclusions reached in this paper. Finally, it should also be acknowledged that while the accuracy of the magnetic moment does not allow us to distinguish between the bulk and epitaxial magnetic moments, the relative high accuracy of the VSM still allows us to follow the temperature dependence of the magnetization accurately.

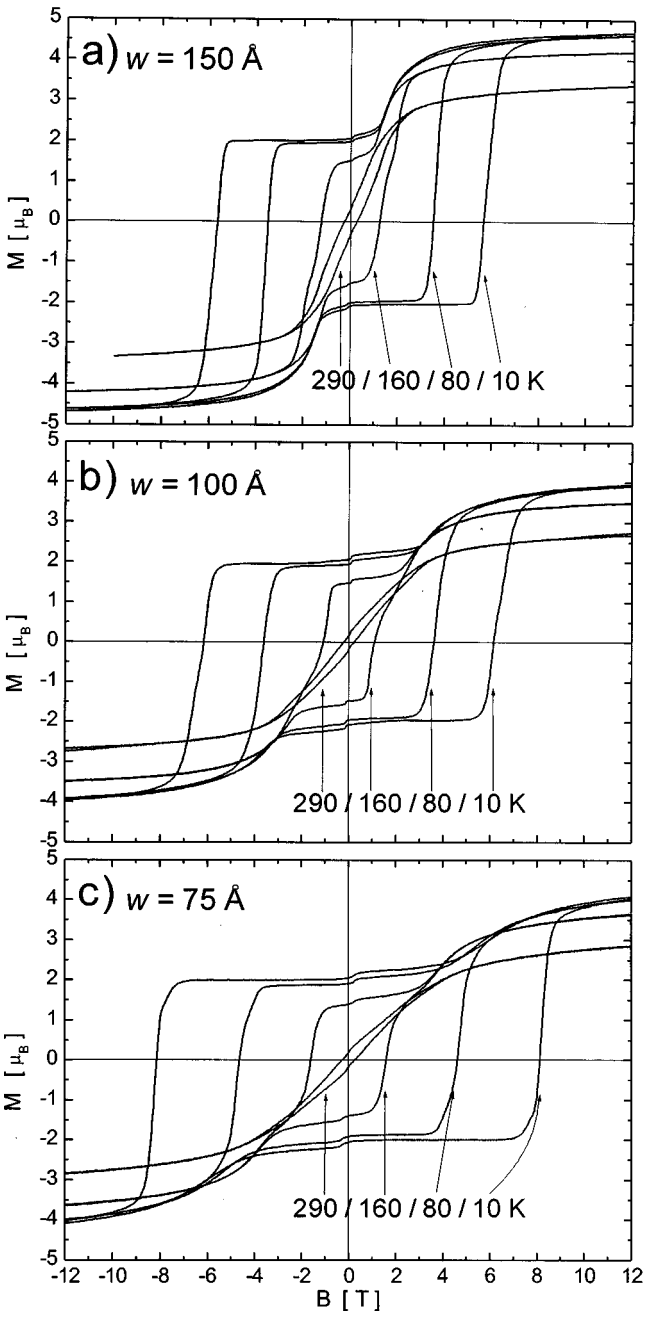

FIG. 2. Selected in plane [001] magnetization curves (magnetic moment per average formula unit $Dy_{0.5}Y_{0.5}Fe_2$) for (a) [150-Å $DyFe_2$/150-Å YFe_2]×40, (b) [100-Å $DyFe_2$/100-Å YFe_2]×40, and (c) [75-Å $DyFe_2$/75-Å YFe_2]×40.

III. RESULTS

From an examination of Figs. 2(a)–(c), it will be observed that all three *M-B* loops show a relatively sharp increase in

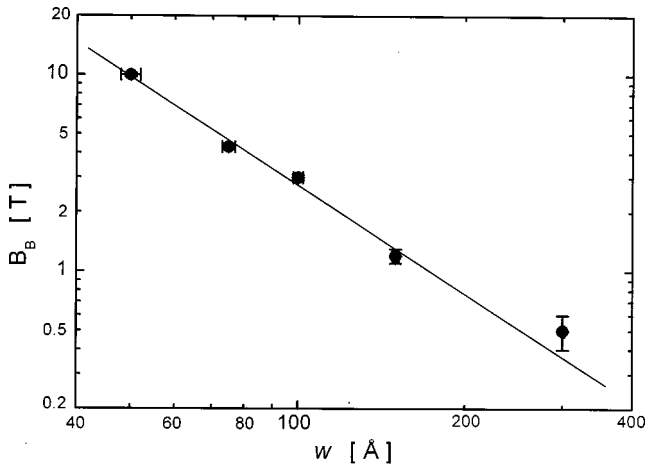

FIG. 3. The bending field B_B as a function of the width of the YFe_2 layer w, plotted on a log-log scale.

magnetization, in the positive right-hand quadrant. For example, in Fig. 2(a), the overall increase in magnetization, from 2 to $4\mu_B$, commences at the bending field of $B_B = 1.2\,\mathrm{T}$, at 10 K. This increase in magnetization is readily understood in terms of exchange springs. If we ascribe moments of 10 and $1.5\mu_B$ to the Dy and Fe atoms, respectively, then in the antiferromagnetic state, the net magnetic moment per formula unit ($Dy_{0.5}Y_{0.5}Fe_2$) is $2\mu_B$. Thus the increase from 2 to $4\mu_B$ is entirely due to the creation of exchange springs. Furthermore, after saturation, the magnetization curve is found to be reversible if the field is ramped up and down between the coercive field $-B_C$ and $+12$ T. This reversibility is a characteristic feature of magnetic exchange springs.[1] Moreover, from an examination of Figs. 2(a)–(c), it is clear that larger bending fields B_B are required when the thickness of the YFe_2 layers is reduced. This property is highlighted in Fig. 3, which shows the measured bending fields B_B versus the thickness w of the YFe_2 layer, plotted on a log-log scale. It will be observed that the points fall almost on a straight line. From a least squares fit we find that

$$B_B = kw^n, \tag{1}$$

where

$$n = -1.83 \pm 0.12,$$

$$\log_{10}(k) = 4.1 \pm 0.2. \tag{2}$$

This behavior can be explained in terms of a nearest-neighbor exchange model, which we outline briefly below.

IV. DISCUSSION

Following Ref. 1, but with minor changes in notation, the total energy for a soft domain wall, with nearest-neighbor interactions only, takes the form

$$E = \sum_n \varepsilon(n), \tag{3}$$

where

$$\varepsilon(n) = -\mu_{Fe}B_{APP}\cos\theta_n - \tfrac{1}{2}\mu_{Fe}B_{EX}[\cos(\theta_n - \theta_{n-1}) + \cos(\theta_{n+1} - \theta_n)]. \tag{4}$$

Note that any anisotropy associated with the YFe_2 layer has been neglected. Also care must be exercised with the spins at the beginning and end of the magnetic exchange spring, which are pinned by the $DyFe_2$ layers.

For stability, the energy of the entire domain E wall must be stable with respect to small partial variations in θ_n:

$$\frac{\partial E}{\partial \theta_n} = 0. \tag{5}$$

Proceeding in this fashion, it is possible to determine the set of equilibrium angles $\{\theta_n\}$, in an iterative fashion.[11] Moreover, given the equilibrium set $\{\theta_n\}$, it is a straightforward matter to calculate the energy of the entire exchange spring and to compare it with the energy of the simple ferromagnetic state (i.e., no exchange spring). In practice, exchange springs will only form when the gain in Zeeman energy outweighs the concomitant loss in magnetic exchange energy. The transition takes place at the bending field B_B, and from both computer and analytical calculations we find that

$$\frac{B_B}{B_{EX}} = \left(\frac{\pi}{N}\right)^2, \tag{6}$$

where B_{EX} is the Fe-Fe exchange field and N is the number of soft YFe_2 monolayers. A more detailed proof of this simple result will be given elsewhere.[11] But for the present we note that $B_B \propto 1/w^2$, where w is the thickness of the soft layer. This inverse square law was first derived, many years ago, for bilayer samples.[10] More recently, a similar result has been obtained for the case of a soft magnetic sphere of radius R, embedded in a hard magnetic medium.[12]

As noted earlier, the experimentally determined exponent of w falls slightly below that of -2. However, we believe that the experimentally determined figure of -1.83 ± 0.12 is acceptable, given the uncertainties associated with spin pinning at the edges of the YFe_2 layer. In this regard, preliminary TEM measurements show that the surface roughness of the interfaces could be as high as ± 5 Å.[13]

Finally, we can use Eq. (6), and the bending fields shown in Figs. 2(a)–(c), to make an estimate of the magnetic exchange field. For example, for the $w = 100$-Å sample, the effective number of Fe monolayers N is ~ 55, where we have assumed that there are four Fe layers per unit cell. Thus on using $B_B = 3.0\,\mathrm{T}$, we find $B_{EX} \sim 910\,\mathrm{T}$. This figure is in excess of the figure of 680 T, given by Ref. 6 and may reflect the inadequacies of the nearest-neighbor effective-field model.

V. CONCLUSIONS

We have presented magnetization results for MBE grown $DyFe_2$-YFe_2 superlattice samples, with antiferromagnetic coupling between the two component layers. The results show the existence of model exchange springs in the magnetically soft YFe_2 layers, pinned at their edges by the mag-

netically hard $DyFe_2$ layers. These exchange springs are characterized by well-defined bending (onset) fields B_B, which are proportional to $1/w^n$, where the experimentally determined exponent n is almost 2.

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[1] E. E. Fullerton, J. S. Jiang, M. Grimsditch, C. H. Sowers, and S. D. Bader, Phys. Rev. B **58**, 12 193 (1998).
[2] E. E. Fullerton, J. S. Jiang, and S. D. Bader, J. Magn. Magn. Mater. **200**, 392 (1999).
[3] J. M. D. Coey and R. Skomski, Phys. Scr. **T49**, 315 (1993).
[4] R. Skomski and J. M. D. Coey, IEEE Trans. Magn. **29**, 2860 (1993).
[5] For a review, see K. H. J. Buschow, Rep. Prog. Phys. **40**, 1179 (1977).
[6] J. J. M. Franse and R. J. Radwanski, in *Handbook of Magnetic Materials*, edited by K. H. J. Buschow (North-Holland, Amsterdam, 1993), Vol. 7.
[7] G. J. Bowden, D. St. P. Bunbury, A. P. Guimaraes, and R. E. Snyder, J. Phys. C **1**, 1376 (1968).
[8] V. Oderno, C. Dufour, K. Dumesnil, Ph. Bauer, Ph. Mangin, and G. Marcal, Phys. Rev. B **54**, R17 375 (1996).
[9] M. Sawicki, G. J. Bowden, P. A. J. de Groot, B. D. Rainford, R. C. C. Ward, and M. R. Wells, J. Appl. Phys. **87**, 6839 (2000).
[10] E. Goto, N. Hayashi, T. Miyashita, and K. Nakagawa, J. Appl. Phys. **36**, 2951 (1965).
[11] G. J. Bowden, P. A. J. de Groot, B. D. Rainford, and M. Sawicki (unpublished).
[12] R. Skomski and J. M. D. Coey, *Permanent Magnetism* (IOP Publishing Ltd., Bristol, 1999), p. 178.
[13] E. Grier (private communication).