

## Engineering coercivity in epitaxially grown (110) films of DyFe<sub>2</sub>-YFe<sub>2</sub> superlattices

M. Sawicki, G. J. Bowden,<sup>a)</sup> P. A. J. de Groot, B. D. Rainford, and J. M. L. Beaujour  
*Department of Physics and Astronomy, University of Southampton, Southampton S017 1BJ, United Kingdom*

R. C. C. Ward and M. R. Wells  
*Clarendon Laboratory, Oxford University, Oxford OX1 3PU, United Kingdom*

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Molecular beam epitaxial methods have been used to grow single crystal Laves phase DyFe<sub>2</sub>-YFe<sub>2</sub> superlattice samples with a (110) growth direction. It is shown that it is possible, in principle, to engineer a desired coercivity between the limits  $K_{\text{DyFe}_2} \leq K \leq \infty$ . This can be achieved by adjusting the relative thickness of the individual DyFe<sub>2</sub> and YFe<sub>2</sub> layers, in multilayer films. This novel feature is illustrated, using the superlattice films  $[x \text{ \AA} \text{ DyFe}_2 / (100-x) \text{ \AA} \text{ YFe}_2] \times 40$ , with  $x = 80, 60, 50$ , and 45. It is found that the measured coercivity is in semiquantitative agreement with a simple theoretical expression, for the nucleation fields in both bilayer and multilayer compounds. However, in practice, exchange spring penetration into the DyFe<sub>2</sub> layers can set a limit to the maximum coercivity that can be achieved. © 2000 American Institute of Physics. [S0003-6951(00)03730-X]

In recent years, research into layered magnets has become a hot topic in magnetism.<sup>1-4</sup> For example, it has been argued theoretically that composite magnets with a giant energy product  $(BH)_{\text{MAX}}$  of 120 MGOe might be feasible, if the exchange-spring mechanism in nanostructured oriented magnets could be exploited.<sup>3,4</sup> This work has been complemented experimentally, with magnetic measurements on ferromagnetically coupled hard and soft layers such as sputtered SmCo<sub>5</sub> and Fe layers. For a review of the present status of this field, the reader is referred to Refs. 1 and 2.

However, there are novel applications, which are only possible if the hard and soft layers involved are coupled antiferromagnetically. One of these applications, namely growing superlattice films with increased coercivity, is discussed in this letter. Results are presented for molecular beam epitaxial (MBE) grown, antiferromagnetically coupled DyFe<sub>2</sub>-YFe<sub>2</sub> superlattice samples, at the nanostructure level, which clearly demonstrate that the coercivity of the multilayer films can be engineered. This is achieved, simply by adjusting the relative thickness of the component DyFe<sub>2</sub> and YFe<sub>2</sub> layers, in an appropriate fashion.

Both DyFe<sub>2</sub> and YFe<sub>2</sub> are isomorphic with the cubic Laves  $O_h^7$ -F3dm MgCu<sub>2</sub>-type intermetallic compound.<sup>5,6</sup> They are characterized by: (i) high Curie temperatures in excess of 600 K as a result of strong Fe-Fe magnetic exchange, (ii) relatively strong rare-earth (RE)-Fe magnetic exchange fields  $\sim 100$  K, which gives rise to antiferromagnetic (ferromagnetic) coupling in the heavy (light) RE compounds, respectively, and (iii) strong crystal field interactions at the RE site, which control the direction of easy magnetization.<sup>7</sup> For bulk DyFe<sub>2</sub> the direction of magnetization is [001] at all temperatures, whereas for YFe<sub>2</sub> it is [111]. However since the magnetic anisotropy associated with YFe<sub>2</sub> is very weak, it can be considered as magnetically soft.

While all of the above features are also present in the MBE grown thin films, there is one important difference. Because of strain, induced during crystal growth, the direction of magnetization is temperature dependent.<sup>8</sup> Nevertheless, at temperatures below  $\sim 100$  K, the direction of easy magnetization in MBE grown DyFe<sub>2</sub> films reverts to [001].<sup>8,9</sup>

Recently, all of the above features have been used to engineer: (i) a magnetic compensation point in a  $[25 \text{ \AA} \text{ DyFe}_2 / 50 \text{ \AA} \text{ YFe}_2] \times 40$  superlattice sample, with relatively short individual layers ( $\sim 50$  \AA),<sup>9</sup> and (ii) magnetic exchange springs in YFe<sub>2</sub> with relatively long individual layers ( $\geq 100$  \AA).<sup>10</sup> However, in this letter, we concentrate on a third application: namely engineering the magnetic coercivity of superlattice films of DyFe<sub>2</sub> and YFe<sub>2</sub>. This is illustrated using MBE grown films of  $[x \text{ \AA} \text{ DyFe}_2 / (100-x) \text{ \AA} \text{ YFe}_2] \times 40$ , with  $x = 80, 60, 50$ , and 45. For comparative purposes magnetization curves are also presented for an MBE grown DyFe<sub>2</sub> film, and an MBE grown alloy film Dy<sub>0.5</sub>Y<sub>0.5</sub>Fe<sub>2</sub>. From these results, it will be seen that the coercivity of a given multilayer sample can be increased, either by reducing the relative thickness of the individual DyFe<sub>2</sub> layers, or by growing selected MBE alloy films.

The DyFe<sub>2</sub>-YFe<sub>2</sub> multilayer samples were grown by MBE techniques, using the Balzers UMS 630 ultrahigh vacuum facility at Oxford, in the manner described in Ref. 8. The samples were grown on epi prepared (11 $\bar{2}$ 0) sapphire substrates (10 × 12 mm) with a 1000 \AA (110) Nb buffer and a 30 \AA layer of Fe. The superlattice films were grown by codeposition of elemental fluxes at a substrate temperature of 400 °C in (110) orientation and with the major axes parallel to those of niobium.<sup>9</sup> Since the bulk lattice parameters for DyFe<sub>2</sub> and YFe<sub>2</sub> are 7.325 and 7.363 \AA, respectively, the mismatch at the superlattice interfaces is expected to be on the order 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

<sup>a)</sup> Author to whom correspondence should be addressed; electronic mail: gjb@phys.soton.ac.uk

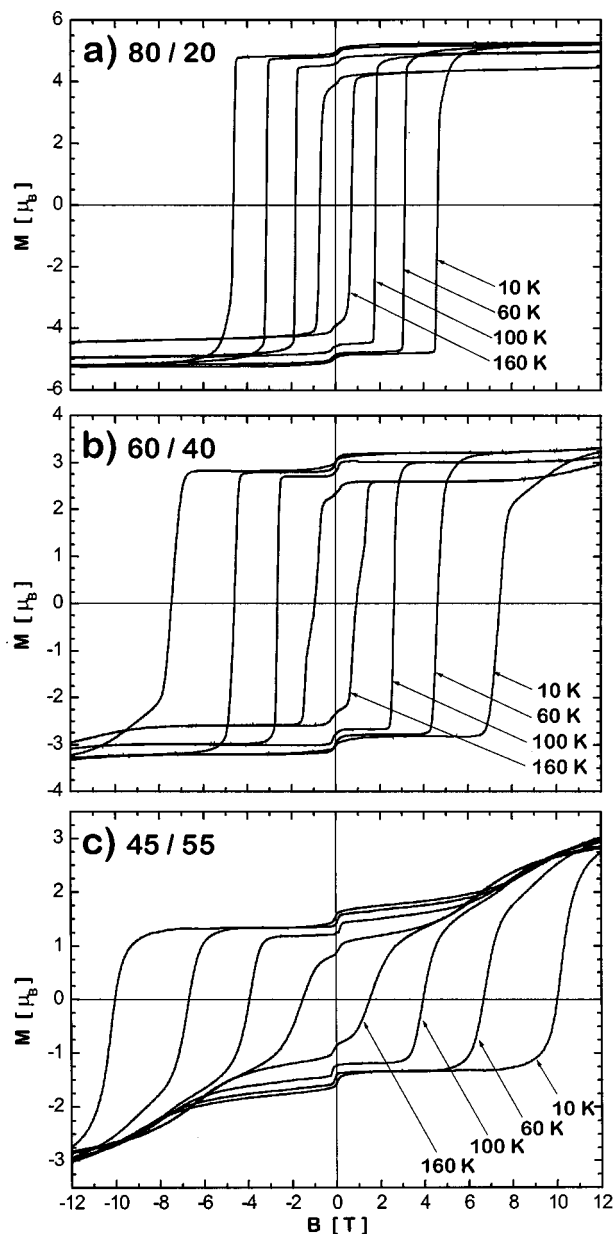


FIG. 1. Selected in-plane magnetization curves for the superlattice samples, for magnetic fields directed along a [001] axis: (a) [80 Å DyFe<sub>2</sub>/20 Å YFe<sub>2</sub>] $\times$ 40, (b) [60 Å DyFe<sub>2</sub>/40 Å YFe<sub>2</sub>] $\times$ 40, (c) [45 Å DyFe<sub>2</sub>/55 Å YFe<sub>2</sub>] $\times$ 40. As an aid to the eye, the ratios of the thickness (80/20, 60/40, and 45/55) have also been included in the diagrams.

using an Oxford Instruments Aeronomic 3001 vibrating sample magnetometer (VSM). The temperature range covered was 10–300 K, in applied fields of up to 12 T. In all cases, the magnetic field was applied along a [001] axis.

Typical in-plane magnetization curves, for  $[x \text{ \AA} \text{ DyFe}_2/(100-x) \text{ \AA} \text{ YFe}_2] \times 40$ , where  $x=80, 60$ , and  $45$ , can be seen in Figs. 1(a), 1(b), and 1(c), respectively. 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 letter. Finally, it should also be acknowledged

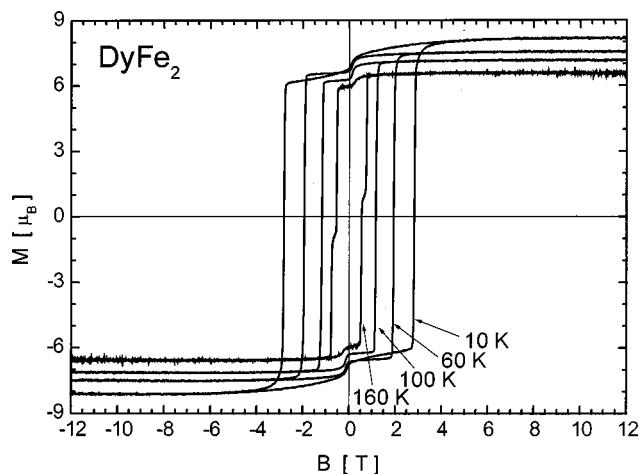


FIG. 2. Selected in-plane magnetization curves for MBE grown [4000 Å DyFe<sub>2</sub>], for magnetic fields directed along a [001] axis.

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.

From an examination of Figs. 1(a), 1(b), and 1(c), it will be seen that the coercivity of the samples increases as the thickness of the DyFe<sub>2</sub> layers is reduced, reaching in excess of 10 T in the [45 Å DyFe<sub>2</sub>/55 Å YFe<sub>2</sub>] $\times$ 40 multilayer sample. For comparative purposes, the measured magnetization curve for ‘pure’ DyFe<sub>2</sub> is shown in Fig. 2. It will be observed that the measured coercivity in DyFe<sub>2</sub> (~3 T) is lower than that of any of the superlattice samples shown in Figs. 1(a)–1(c). Finally, in Fig. 3 the magnetization curves for the alloy Dy<sub>0.5</sub>Y<sub>0.5</sub>Fe<sub>2</sub> can be seen. The coercivity of this alloy is practically identical to that of the [50 Å DyFe<sub>2</sub>/50 Å YFe<sub>2</sub>] $\times$ 40 multilayer sample reported in Ref. 10. Next we will provide a simple theoretical interpretation for all of these results.

It has been known for some time (see Refs. 1, 2, 11, and 12), that the nucleation-field  $B_N$  (coercivity) in layered magnets can be described by the relationship

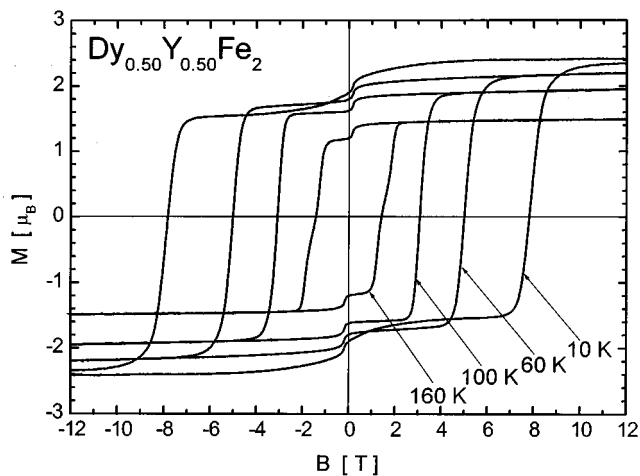


FIG. 3. Selected in-plane magnetization curves for the alloy Dy<sub>0.5</sub>Y<sub>0.5</sub>Fe<sub>2</sub>, for magnetic fields directed along a [001] axis.

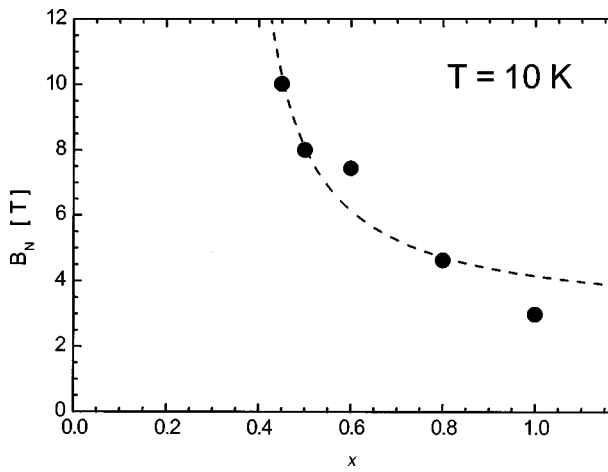


FIG. 4. The experimentally determined coercivity at  $T=10$  K, as a function of the thickness of the DyFe<sub>2</sub> layers (bullets); (dashed line) a theoretical plot, based on Eq. (1).

$$B_N = \frac{2(K_H t_H + K_S t_S)}{M_H t_H + M_S t_S}, \quad (1)$$

where  $K_H$  ( $K_S$ ) are the anisotropies of the hard (soft) layers, and  $t_H$  ( $t_S$ ) the thickness of the hard (soft) layers, respectively.

In the past, Eq. (1) has been used to describe the nucleation fields of ferromagnetically coupled hard and soft magnetic layers. However in the case under discussion, the magnetization of the hard (DyFe<sub>2</sub>) layer is antiparallel to that of the soft (YFe<sub>2</sub>) magnetic layer. To emphasize this difference, we rewrite Eq. (1) in the form

$$B_N = \frac{2K_H t_H}{M_H t_H - M_S t_S}, \quad (2)$$

where: (i) we have explicitly acknowledged the fact that the magnetization of the two layers involved are antiparallel to each other, and (ii) for simplicity, we have set the anisotropy constant for the soft layer  $K_S=0$ . Clearly if we choose  $M_H t_H = M_S t_S$  (i.e., the magnetic compensation point), the nucleation field  $B_N$  should go to infinity. But this interesting result is of little use, because the net magnetization  $M$  is now zero.

In Fig. 4, the experimentally determined coercivity can be seen as a function of the thickness of the DyFe<sub>2</sub> layers. Also included in this diagram is a least squares fit to the experimental data, based on Eq. (2). From this fit we find  $K_H = 9 \pm 3$  K per ion, where we have ascribed  $7\mu_B$  ( $3\mu_B$ ) per formula unit to the magnetic moments of the hard (soft) magnetic layers, respectively. While the agreement is reasonable, it must be acknowledged that Eqs. (1) and (2) are simplistic in that they do not take into account any energy expended in creating magnetic exchange springs. This feature is particularly evident, in the case of the  $[45 \text{ \AA} \text{ DyFe}_2 / 55 \text{ \AA} \text{ YFe}_2] \times 40$  sample, where there is clear evidence for the formation of magnetic exchange springs in the mag-

netization curve at 160 K, in Fig. 1(c). Here the intrinsic  $M$ - $B$  loop associated with the DyFe<sub>2</sub> layers is now quite small, and so the large increase in magnetization, associated with magnetic exchange spring formation, is more pronounced. Note that the magnetic moment in the positive quadrant rises in the classic curved manner seen in magnetic spring formation.<sup>10</sup> Moreover, this part of the  $M$ - $B$  loop is reversible, yet another characteristic of exchange springs.

At first sight, this observation is somewhat surprising. For example, it might be argued that a 55 Å YFe<sub>2</sub> layer is too short for the formation of a magnetic spring of less than  $\sim 10$  T.<sup>10</sup> But, in practice, it is likely that exchange springs, which originate primarily in the soft YFe<sub>2</sub> layers, will penetrate into their neighboring DyFe<sub>2</sub> layers. Moreover, this feature will become more pronounced, as the thickness of the DyFe<sub>2</sub> layers is reduced, and the temperature increased. Thus the effective length of the exchange spring can exceed the thickness of the individual YFe<sub>2</sub> layers, and may even penetrate the entire sample. In summary therefore, exchange spring formation will set a limit to the maximum coercivity that can be achieved in superlattice samples. However, this limitation does not apply to the MBE grown alloy samples, with modest Dy content, because exchange spring formation is now essentially forbidden.

It has been shown how antiferromagnetic coupling between hard and soft magnetic layers can be used to engineer the coercivity of a given bilayer or superlattice film, by adjusting the thickness of the component layers. In addition, it has been noted that exchange spring formation can occur in superlattice films, where the thickness of the hard layers is not thick enough to pin the edges of an exchange spring. However, this difficulty can be circumvented by growing MBE alloy films free of exchange springs.

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