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2004 J. Phys.: Condens. Matter 16 L213

(http://iopscience.iop.org/0953-8984/16/15/L02)

J. Phys.: Condens. Matter **16** (2004) L213–L220 PII: S0953-8984(04)76039-7

LETTER TO THE EDITOR

Oscillatory temperature dependence of exchange bias for Fe/Gd ferrimagnets

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Received 11 February 2004
Published 2 April 2004
Online at stacks.iop.org/JPhysCM/16/L213
DOI: 10.1088/0953-8984/16/15/L02

Abstract

We investigated the exchange bias effect for Fe/Gd ferrimagnetic multilayers. Our results show that there is a −cosine type of oscillatory exchange bias with temperature for the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer. For the first time, exchange bias was observed to oscillate between positive and negative fields with a period of approximately 130 K. In addition, the coercive field was observed to oscillate within itself for the same temperature range studied. We speculate that oscillatory exchange bias is related to the underlying magnetic phase diagram of the ferrimagnetic Fe/Gd multilayer. When the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] system is cooled under a high magnetic field, negative exchange bias is forced to shift to positive values irregularly. Nevertheless the system shows oscillatory behaviour.

Exchange bias (H_{EB}), which is the shift of the hysteresis loop with respect to the zero magnetic field axis, was discovered by Meiklejohn and Bean [1] in 1956 at low temperatures when the CoO/Co system is cooled under an external magnetic field. Since then various antiferromagnets (AF), which are exchange coupled to ferromagnets (F), have shown H_{EB} [2]. In addition, H_{EB} and related effects have also been observed for the ferrimagnet (ferri)/F and ferri/AF interfaces and even antiferromagnetically coupled F/F systems [3–6]. Generally, negative H_{EB} with respect to the applied external magnetic field direction is observed in the AF/F systems [2]. Positive H_{EB} is very rare. There are examples of positive H_{EB} where the explanation is thought to be due to the coupling of the AF/F interface to very high external fields [7].

There are well written reviews on the subject listing possible configurations [2, 8–10] and there are several theories [11–16] applicable to A/AF systems. In an early theory of H_{EB}, Meiklejohn [11] considers the coherent rotation of F and AF layers across an uncompensated interface. This pedagogical work estimates the H_{EB} to be orders of magnitude larger than the observed value. In Malozemoff's model [12], random interface roughness gives rise to a random magnetic field acting on the interface spins. Therefore, the AF breaks up into domains, yielding a unidirectional anisotropy. Mauri *et al* [13] proposed in their model that a domain wall formation in the AF sets a limit for H_{EB} irrespective of exchange coupling. Koon's [14] micromagnetic calculations show that F magnetization is perpendicular to that of AF in the ground state configuration. Schulthess and Butler [15] combines Malozemoff's random field model with Koon's micromagnetic calculations and shows that interfacial defects can lead to H_{EB} of the correct order of magnitude as the observed values. In Stiles and McMicheal's model [16], F is coupled to the polycrystalline interfacial AF grains with random orientations. The unidirectional anisotropy comes from the grains where the antiferromagnetic order is stable as the magnetization is rotated. All the models (except the earliest [11]) give reasonable values in agreement with experiment.

Fe/Gd is a theoretically and experimentally well studied rare earth/transition metal (RE/TM) system [17–34]. The Fe/Gd multilayer becomes an artificial ferrimagnet due to the antiferromagnetic coupling between the Gd and Fe moments. Since the magnetizations of the two antiferromagnetically coupled materials act differently with temperature there is a particular temperature which is defined as the compensation temperature (T_{comp}) for ferrimagnets [35]. Generally, the RE magnetization is larger below T_{comp} since the Curie temperature (T_C) of the RE ($T_C = 292$ K for Gd) is much lower than that of the TM ($T_C = 1043$ K for Fe).

For Fe/Gd ferrimagnets, the Gd-aligned and Fe-aligned states are below and above T_{comp}, respectively. In the Gd-aligned state the Gd layer magnetization is aligned with the external magnetic field while the Fe magnetization is antiparallel to that of the Gd. Furthermore the Fe magnetization aligns with the magnetic field above T_{comp}, which is the opposite of Gd magnetization. In the original work of Camley and Tilley [18] there are bulk and surface twisted states along with the Gd- and Fe-aligned states. The different phases arise from the competition between exchange and Zeeman energies. At low fields, exchange energy is dominant over the Zeeman energy and favours the system to be in one of the aligned states (Gd-aligned or Fe-aligned). The net magnetic moment of the ferrimagnet determines which aligned state is favoured. For moderate fields, both Zeeman and exchange energies are effective and the system favours the twisted states which are the mixed states of Gd- and Fe-aligned states [17–20].

In this letter, we discuss the anomalous magnetic behaviour of the particular [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer. First, we show that the H_{EB} oscillates with temperature, and then discuss the possible explanations of the H_{EB} oscillations. Second, we match the simultaneous coercivity (H_C) oscillations with the oscillatory exchange bias. Finally, we look into the high cooling field effects in the oscillatory H_{EB} and coercivity (H_C) patterns.

Samples were prepared in a dc magnetron sputtering system at room temperature (RT). The unbaked base pressure of the UHV deposition chamber was 10^{-9} Torr. High purity argon gas was used and deposition pressure was 3 mTorr. Pure Ag, Gd and Fe targets were used. Deposition rates were 0.038, 0.028 and 0.031 nm s^{-1} for Ag, Gd and Fe, respectively. Samples were deposited on Corning glass substrates. 20 nm thick Ag layers were used as buffer and cap layers in all samples. Deposition thicknesses were monitored *in situ* by a quartz thickness monitor calibrated by a stylus profilometer. Hysteresis measurements were made using a superconductor quantum interference device (SQUID) between 4 and 300 K. Samples were cooled from RT to 4 K under either 0.2 T (low) or 1 T (high) external magnetic field which is applied in the plane of the multilayers, Then successive hysteresis measurements in ± 0.1 and ± 0.5 T intervals were taken with increasing temperature.

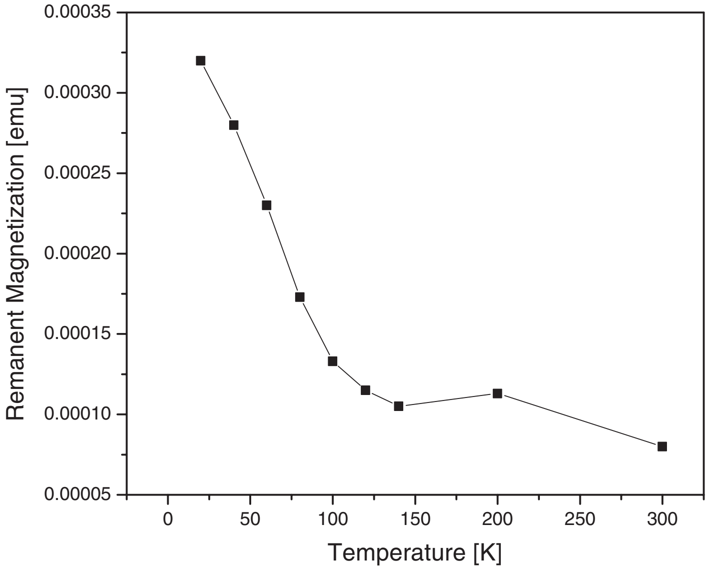

Figure 1. Remnant magnetization versus temperature for the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer. The T_{comp} occurs approximately at 140 K.

The [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] system has a T_{comp} of approximately 140 K as seen from the local minimum in figure 1. In our previous work [5], the (Fe 4 nm/Gd 4 nm)$_4$ part of the multilayer was observed to have a T_{comp} around 90 K. Here, the addition of the Gd underlayer enhances T_{comp} up to approximately 140 K. In figure 1, as we approach room temperature, the system loses its ferrimagnetic property due to the paramagnetic behaviour of the Gd. Thus, the system is left only with the Fe moment which has slightly lower remnant magnetization at 300 K than at 200 K.

In figure 2, H_{EB} of the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] system cooled under an external field of 0.2 T is shown as a function of temperature. H_{EB} oscillates between negative and positive values with a nearly −cosine curve. The period of oscillation is around 130 K throughout the temperature range (except near room temperature). The amplitude of the oscillations is approximately 10 Oe. H_{EB} intersects with the temperature axis at 30, 90 and 160 K. Below 30 K, H_{EB} is negative. Between 30 and 90 K it is positive. In the 90–160 K temperature interval it is again negative and finally above 160 K it is positive. Selected hysteresis loops at various temperatures for the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer are shown in figure 3.

Previous experimental results on GdCo/Co multilayers have shown that H_{EB} changes sign when the system passes through T_{comp} [36, 37] and it was observed in Gd/Fe that H_{EB} becomes zero at T_{comp} [5]. For these systems the temperature axis intercept of H_{EB} may then be interpreted as the T_{comp} of the system. This is obviously not our case here, because T_{comp} of the system (140 K) does not correspond to a zero value of H_{EB} and the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] system has only one T_{comp} as shown in figure 1. However, one should keep this in mind throughout this letter.

The Fe/Gd magnetic phase diagram can be found in the literature [18–20, 22, 23]. It is basically composed of four phases: Gd-aligned, surface twisted, bulk twisted and the Fe-aligned regions. If the system passes from one phase to the other, H_{EB} may change from positive to negative [5, 36, 37]. We speculate that a possible explanation for the oscillatory H_{EB} with

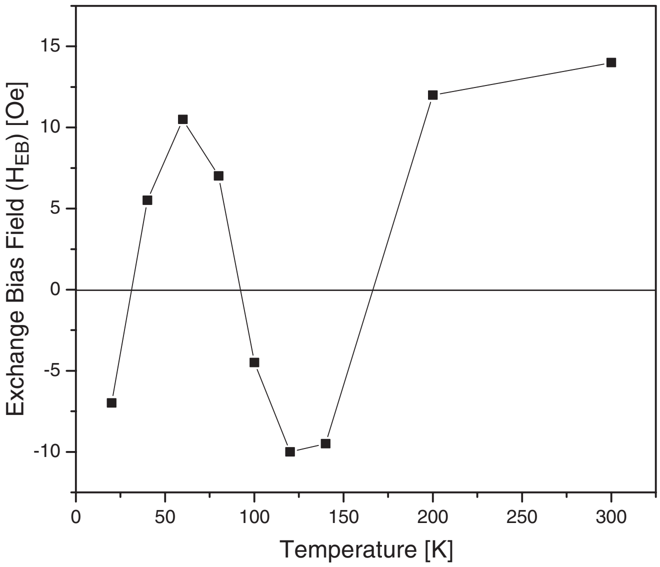

Figure 2. The H_{EB} versus temperature for the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer. H_{EB} is oscillating with a period of 130 K. H_{EB} oscillations can be fitted to a −cosine curve. Positive H_{EB} at 300 K is an example of the proximity effect.

temperature may depend on the magnetic phase diagram of the ferrimagnetic Fe/Gd system. The oscillatory H_{EB} can be tracking the different phases in the magnetic phase diagram. In this view, the negative H_{EB} below 30 K in figure 2, corresponds to the Gd-aligned phase. Then the positive H_{EB} between 30 and 90 K corresponds to the surface twisted phase. The negative H_{EB} between 90 and 160 K corresponds to the bulk twisted phase. Finally, the positive H_{EB} above 160 K corresponds to the Fe-aligned phase.

A simple picture of antiferromagnetically coupled layers with uniform moments does not apply to the real Fe/Gd multilayer case [26, 30–32]. At low temperature, the Gd sublattice shows a non-negligible magnetic anisotropy, as is evident from the enhancement of the H_C (see figures 5 and 6). Camley [18] does not take into account the Gd anisotropy energy in his model for the Fe/Gd multilayers. Therefore we suggest that anisotropy energy has to be put into the Hamiltonian for a more precise picture. In an earlier work of Hosoito and coworkers [32] the Gd moments show exchange spring behaviour in the Gd sublayer. The moments at the core of each Gd sublayer are reduced even at the lowest temperatures compared to the Gd moments in the proximity of the Fe layers. Therefore the core Gd moments can deviate from the rest of the Fe/Gd multilayer alignment even at zero external magnetic field. Furthermore, moments at the core of the Gd sublayer have a temperature dependence that is proportional to $(1 - T/T_C)$ with $T_C < T_{C(Bulk)}$. In figure 2, at 5 K ($T \ll T_{comp}$) the Gd-aligned state shows $-H_{EB}$ and at $T \gg T_{comp}$ (above 180 K) the Fe-aligned state shows $+H_{EB}$. Therefore positive and negative H_{EB} are the labels for the Fe- and Gd-aligned states, respectively [5, 6]. The H_{EB} oscillation in between these extremes is the evidence of some additional transitions such as an Fe-aligned state in the Gd-aligned region below T_{comp} [28] and the opposite, Gd-aligned state in the Fe-aligned region above T_{comp} [33]. Thus the Fe/Gd phase diagram deviates from Camley's model [18] at low fields and oscillatory H_{EB} gives a new physical insight for understanding the real behaviour of the Fe/Gd multilayers at low external magnetic fields.

As the temperature approaches RT in figure 2, the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] system is left with the Fe layer magnetization due to the paramagnetic behaviour of the Gd. Positive

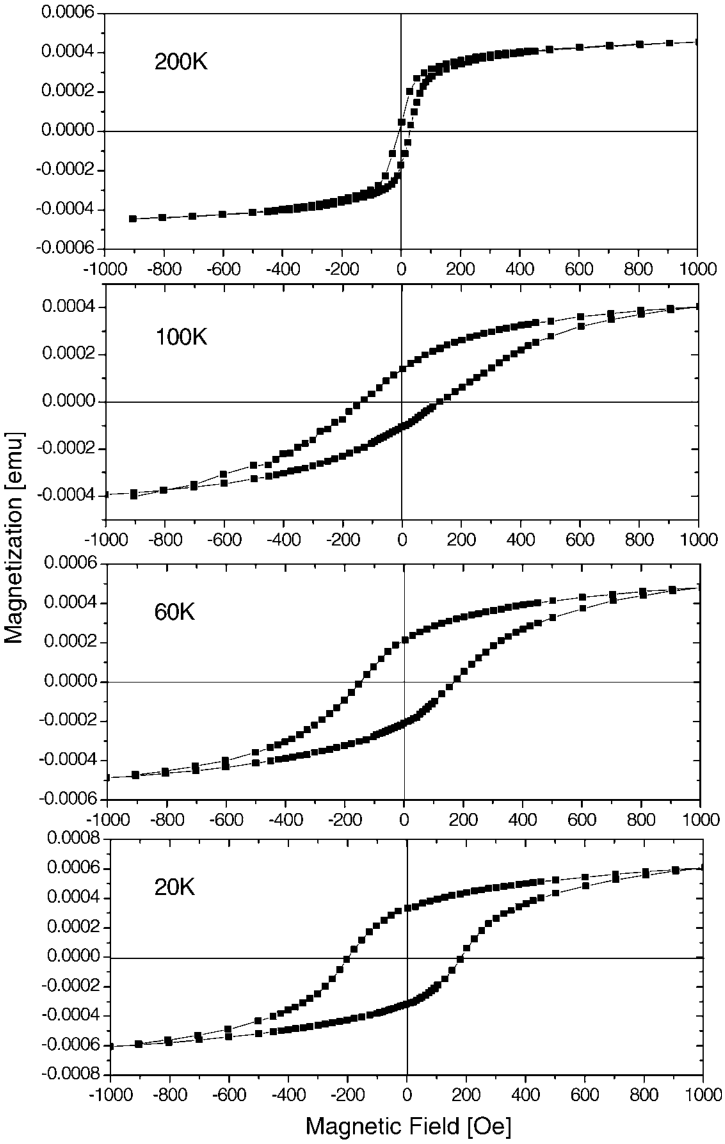

Figure 3. Selected hysteresis loops at various temperatures. The sample was cooled under a 0.2 T external field applied in the plane of the multilayer. Then successive hysteresis loops were taken at ± 0.1 T field intervals with increasing temperature. For the H_{EB} corresponding to each temperature see figure 2.

H_{EB} near room temperature is due to the antiferromagnetic interaction of the Fe magnetization with that of the Gd in the proximity of the Fe layer. H_{EB}, which is due to the proximity effects, is not a part of the oscillatory behaviour, since the effective magnetic Gd layer thicknesses are solely confined to the proximity of the Fe layers. Hysteresis loop shifts close to or at higher temperatures than the Curie temperature of the Gd are a unique example of the proximity effect.

When the same experiment is repeated under a cooling field strength of 1 T, the oscillatory H_{EB} pattern is shifted irregularly to positive values as shown in figure 4. Nevertheless, the

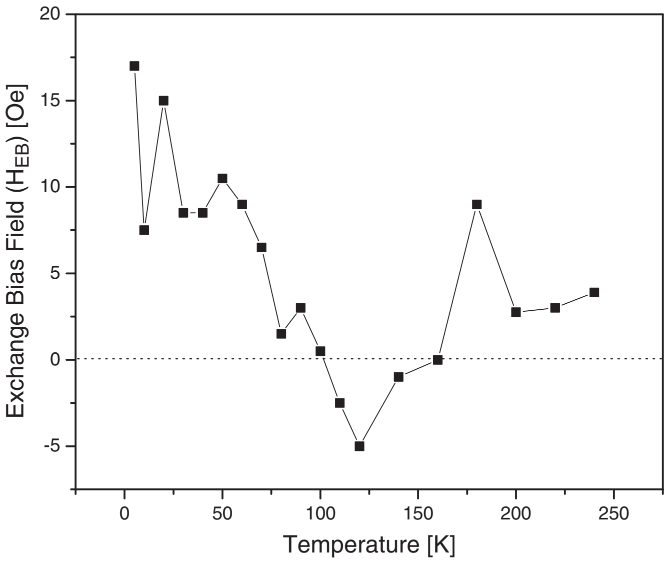

Figure 4. The H_{EB} field versus temperature for the 1 T field cooled [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer. Hysteresis loops were taken at ±0.5 T field intervals (not shown). Negative H_{EB} below 100 K tends to move to the positive side.

H_{EB} conserves its overall oscillatory behaviour. Although the 1 T cooling field is sufficient enough to shift part of the negative H_{EB} to positive values, it is not sufficient enough to change the negative bias totally near the T_{comp} of the system. Basically, high cooling fields are able to break or deform the antiferromagnetic coupling (of the Fe and Gd layers) at the interfaces, forcing them to couple ferromagnetically to the cooling field direction. Thus, the negative H_{EB} tends to shift to positive fields [7]. We think that even higher cooling fields than 1 T can move the negative H_{EB} totally to positive fields throughout the entire temperature range studied.

The H_C of the [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer in 0.2 and 1 T external cooling fields is shown in figures 5 and 6, respectively. While H_C is decreasing from a high value at low temperatures to a low value at RT, H_C is also oscillating within itself throughout the measured temperature range. Comparing figures 5 and 6, H_C oscillations become more prominent for the high field (1 T) cooled sample, as shown in figure 6. Although the oscillatory H_{EB} pattern is deformed when the cooling field is 1 T, the oscillatory behaviour of H_C with temperature is even larger under high field cooling.

In conclusion, we observe for the first time the temperature-dependent oscillatory H_{EB} and the related effects for the ferrimagnetic [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer. Appearance of the H_{EB} oscillation may lead us to identify the twisted phases as well as the aligned phases by measuring the exchange bias. The Fe/Gd phase diagram deviates from Camley's model [18] at low fields. Therefore oscillatory H_{EB} gives a new insight into understanding the physics of the Fe/Gd multilayers at low fields. Moreover the H_C is oscillating within itself for the same temperature range. High cooling fields deform the natural antiferromagnetic coupling of the Fe and Gd layers and this causes the oscillatory H_{EB} pattern to shift irregularly to the positive fields. Furthermore the H_{EB} near room temperature is one of the examples of the proximity effect where the Gd is paramagnetic except in the proximity of the Fe layers. Finally the publication of these results will motivate further experimental and theoretical work in order to understand the temperature-dependent magnetic processes in

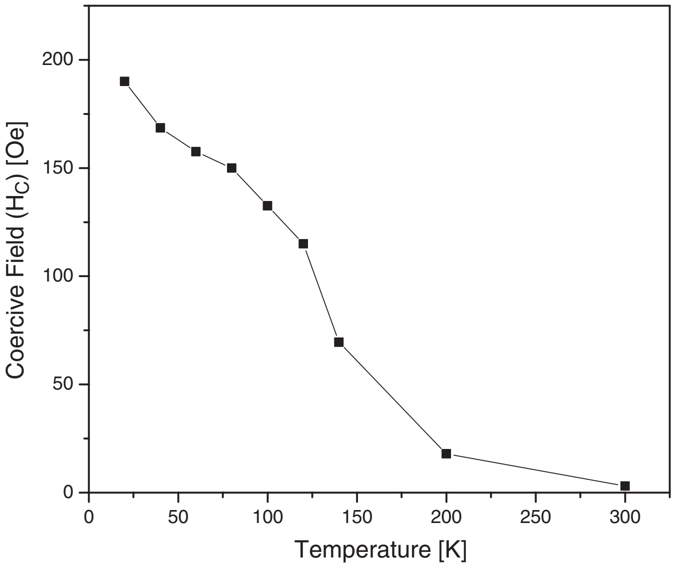

Figure 5. The H_C versus temperature for the 0.2 T field cooled [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer.

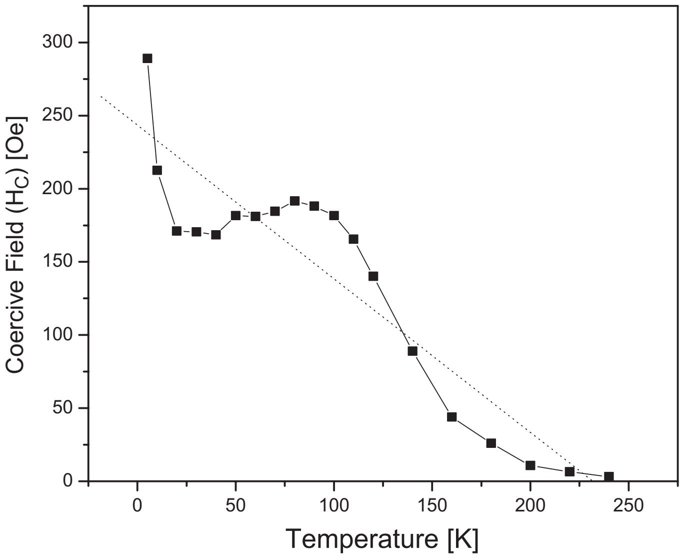

Figure 6. The H_C versus temperature for the 1 T field cooled [(Fe 4 nm/Gd 4 nm)$_4$/Gd 16 nm] multilayer. The linear fit is a guide to the eye.

the Fe/Gd multilayer systems. Our future work will focus on minor loop measurements and studying the effect of cooling fields.

We thank Dr D J Sellmyer for his encouragement. We also thank Drs K Gschneidner and J P Liu for the discussions about Gd anisotropy. This work is supported by a grant from The Welch Foundation.

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