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Tunable high magnetic field thermal hysteresis for exchange-coupled double layers

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Abstract
Two types of tunable thermal hysteresis are observed for the first time in exchange-coupled double layer (ECDL) structures. For low external magnetic fields, ECDLs show one compensation temperature where the measurement of the magnetization as a function of temperature displays a bow-tie shape. For high fields a new type of tunable thermal hysteresis is observed due to an interface wall created between the layers where the ECDL shows two different compensation temperatures. The widths of both forms of thermal hysteresis are tunable with a change in external magnetic field.

Antiferromagnetically coupled Co/Gd, Fe/Gd multilayers and their alloys are well studied structures [1–25]. A key feature of these multilayers/alloys is that the magnetizations of the two materials respond differently to changes in temperature. These systems are called artificial ferrimagnets where the effective concentrations of the two magnetic components are controllable by changing the layering pattern or atomic concentration. The total magnetic moment goes to zero at the compensation temperature (T_{comp}) where equal and oppositely aligned magnetic moments of Co (Fe) and Gd cancel each other. There is a low temperature Gd-aligned state below T_{comp} where the larger Gd magnetization is aligned with the external field (opposite for Co) and there is a high temperature Co-aligned state above T_{comp} where the larger Co magnetization is aligned with the external field (and Gd is opposite). There is also a canted spin state (twisted state) for multilayers where the moments in each atomic layer have a different angle with respect to the applied magnetic field.

Ferrimagnetic multilayers and alloys are subject to a characteristic bow-tie shaped thermal hysteresis under small and moderate magnetic fields [1–3]. Thermal hysteresis arises from the intrinsic uniaxial anisotropy of the system. One may visualize the occurrence of thermal hysteresis as follows: at high temperatures the system is typically a Co-aligned state, with the small Gd moments opposite to the external field. When the temperature is decreased, the Gd

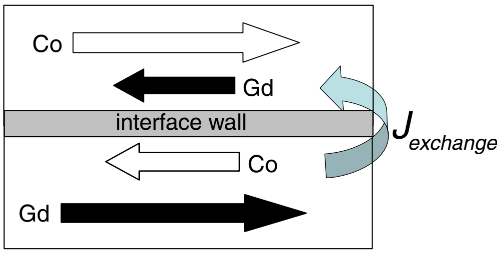

Figure 1. Schematic representation of an exchange-coupled double layer (ECDL) in one of the possible ground states (stable configuration) due to Co–Gd exchange interaction.

(This figure is in colour only in the electronic version)

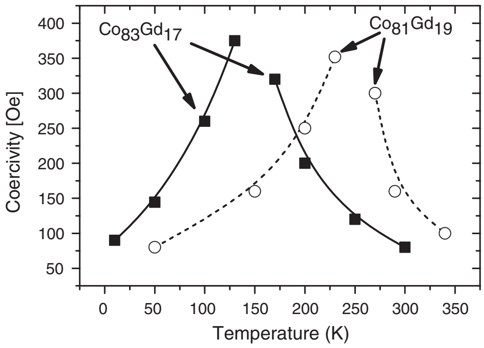

Figure 2. Coercivity as a function of temperature for the $Co_{81}Gd_{19}$ and $Co_{83}Gd_{17}$ alloys.

moments increase, but if some anisotropy holds the Co or Gd spins in place the system will remain in the Co-aligned state; therefore the net magnetic moment can even be opposite to the external field. However, this configuration is no longer stable if the temperature is decreased further, and the system will reverse and have the Gd moments parallel to the applied field (Gd-aligned state) and the Co moments antiparallel. A similar effect occurs on heating the system from low temperatures, except that the Gd-aligned state is held stable by anisotropy as the temperature is increased.

Exchange couple double layers (ECDLs) are the magnetic medium used for magneto-optical recording [26]. If two ferrimagnetic alloys with different atomic compositions are formed, one on top the other, as shown in figure 1, they are called an ECDL [27–34]. Since each layer corresponds to a different atomic concentration and T_{comp}, the coercivity of each layer is different at any temperature. Therefore, at a particular temperature one of the layers can be assumed to be magnetically softer than the other, as shown in figure 2. Coercivity of a ferrimagnetic alloy asymptotically increases around T_{comp}. The coercive field measures how easy it is to reverse the magnetization (figure 2). The field at which this reversal takes place depends on the competition between the anisotropy energy, which would like to hold the moments in their current positions, and the Zeeman energy of the net moment in the applied field. So as the net moment decreases (near T_{comp}) the anisotropy energy wins out and it takes a larger field to reverse the spins. Under an external field, the magnetic alignment of each layer is affected by that of the other, such that an interface wall is created due to the change of alignment between the layers. This is the situation shown in figure 1. Although the coupling within each alloy layer is always antiferromagnetic, magnetic alignment between the alloy layers can be ferromagnetic or antiferromagnetic depending on the exchange coupling, the T_{comp} of each

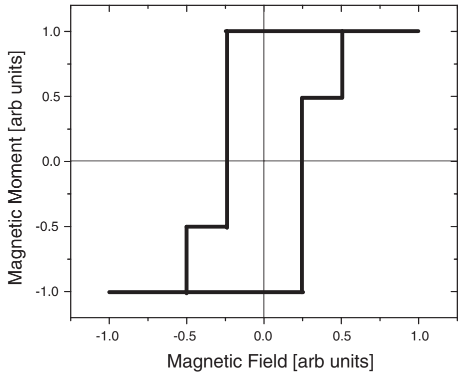

Figure 3. Schematic assumption of hysteresis without exchange coupling for double layers.

layer and the strength of the applied field. For example, ferromagnetic alignment refers to the fact that the net Co moments are aligned parallel to each other across the alloy layers (similarly for the net Gd moments but pointing in the opposite direction).

In this paper, we are interested only in the thermal hysteresis properties of ECDLs. The starting point of this project was the fact that we were observing thermal hysteresis (thermal loops) associated with decoupling in field hysteresis (decoupling refers to steps in field hysteresis). We selected the binary ECDL structure as an example due to its interesting field hysteresis pattern as shown in figure 4. Here, we show for the first time that an ECDL type structure is subject to an unknown high field tunable thermal hysteresis as well as the low field bow-tie thermal hysteresis.

Samples were prepared in a dc magnetron sputtering system at room temperature. They were deposited on Corning glass substrates and 20 nm thick Ag layers were used as the buffer and cap layers. Deposition thicknesses were monitored *in situ* using a quartz thickness monitor calibrated by a stylus profilometer. Co–Gd alloy was created by co-deposition from pure Co and Gd targets. X-ray analysis showed that the formed Co-Gd alloys are x-ray amorphous within the limitations of our measurements. Magnetization (thermal hysteresis) measurements in the temperature interval 10–390 K were taken using a SQUID magnetometer starting from 390 K under a constant in-plane external magnetic field (100 Oe–10 kOe). The temperature was first reduced to 10 K and then increased back to 390 K. The temperature sweep rate was 10 K min^{-1}.

If there is no strong coupling between the ferrimagnetic alloy layers, magnetic hysteresis of an ECDL type bilayer of an arbitrary binary alloy should look like the schematic field hysteresis shown in figure 3. As shown in figure 3, the magnetically softer layer responds to the field reversal first as the field reverses and then the hard layer turns to the field direction at higher fields.

The observed temperature dependent hysteresis loops for the [$Co_{81}Gd_{19}$ 30 nm/$Co_{83}Gd_{17}$ 30 nm] binary ECDL system are shown in figure 4, and are quite different from what was anticipated (figure 3). The difference is due to the existence of exchange coupling between the layers. Whenever one of the layers rotates before the external field reverses direction, an interface wall is formed and this interface wall is exchange biased [27–38] both in forward and reverse fields, as shown in the insets of figure 4. There is a small area created in the hysteresis loop due to the interface wall. Discussions of field hysteresis properties of ECDL type binary structures can be found in the literature. We only use figure 4 as a reference for the remainder of the properties presented below.

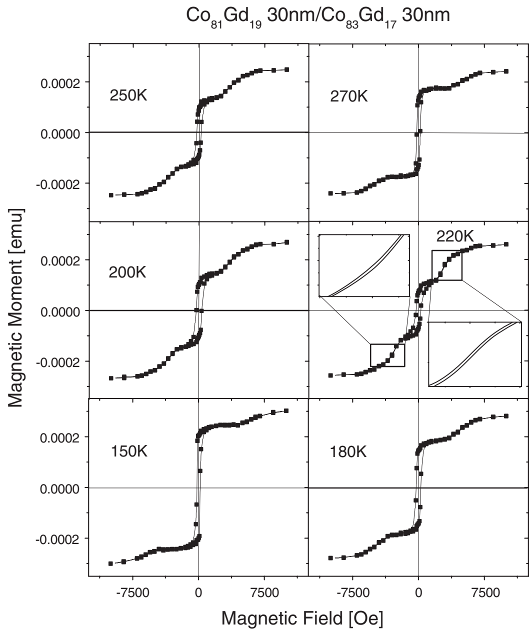

Figure 4. Hysteresis loops at various temperatures for the [$Co_{81}Gd_{19}$ 30 nm/$Co_{83}Gd_{17}$ 30 nm] system. Positive and negative exchange biases are shown as insets to the figure.

The central theme of this paper is to show that ECDLs are subject to more than one type of tunable magnetic thermal hysteresis and we focus our attention solely on the unknown high field thermal hysteresis which we refer to as the second type.

Thermal hysteresis measurements for the [$Co_{81}Gd_{19}$ 30 nm/$Co_{83}Gd_{17}$ 30 nm] system are shown in figure 5 for various external magnetic fields. At low external magnetic fields the [$Co_{81}Gd_{19}$ 30 nm/$Co_{83}Gd_{17}$ 30 nm] system surprisingly shows bow-tie thermal hysteresis, which is the characteristic behaviour of an entire class of antiferromagnetically coupled transition metal/rare earth multilayers [2] and single alloys [3]. The tunable width (80 K at 200 Oe) of the bow-tie thermal hysteresis decays rapidly with increasing field and vanishes at fields around 800 Oe. Here the width of the thermal hysteresis is defined as the temperature interval between the minima of the bow-tie. Detailed discussions of this type of tunable thermal hysteresis can be found elsewhere [1–3]. Above 800 Oe, there is no thermal hysteresis for magnetic fields up to 3 kOe. Although individual layers in the [$Co_{81}Gd_{19}$ 30 nm/$Co_{83}Gd_{17}$ 30 nm] system are assumed to have separate T_{comp}s due to their different atomic concentrations, it turns out that they have a mutual T_{comp} of approximately 220 K under external magnetic fields smaller than 3 kOe. This is why it is surprising to observe such a behaviour in which the individual layer properties of two different alloys are suppressed when they are in contact due to the antiferromagnetic Co–Gd exchange interaction. The binary system acts as if it were just one single alloy with a T_{comp} of 220 K.

For fields of 3 kOe and above a new type of thermal hysteresis is observed, as shown in figure 5. While cooling the [$Co_{81}Gd_{19}$ 30 nm/$Co_{83}Gd_{17}$ 30 nm] system under 3 kOe each layer of the [$Co_{81}Gd_{19}$ 30 nm/$Co_{83}Gd_{17}$ 30 nm] system is in the Co-aligned state at temperatures above 260 K.The hard layer of the system, which corresponds to the higher T_{comp} at 260 K under 3 kOe, switches to the Gd-aligned configuration and creates an interface wall possibly in the hard layer. At 260 K, the net magnetization of the hard layer is assumed to be zero. After

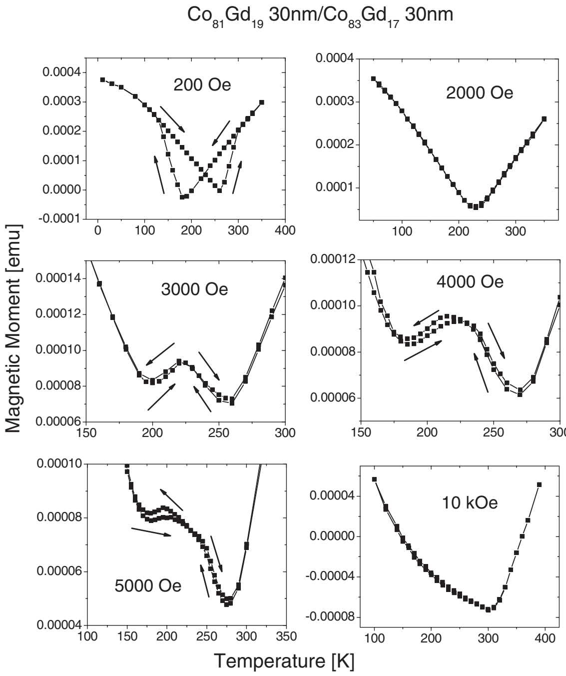

Figure 5. Thermal hysteresis at various external magnetic fields for the [$Co_{81}Gd_{19}$ 30 nm /$Co_{83}Gd_{17}$ 30 nm] system.

the switching of the hard layer at 260 K, the total magnetization of the system increases as the temperature decreases from 260 to 220 K. Around 220 K, which is roughly the mid-point between the corresponding lower and higher T_{comp}s, a coercive hard–soft change of the layers takes place, i.e. the hard layer which has the smaller net magnetization becomes softer and the soft layer which has the larger net magnetization becomes harder (as shown in figure 2). Below 220 K, the total magnetization of the system decreases as the temperature is lowered to 200 K. At temperatures between 200 and 260 K the alignment between the layers is antiferromagnetic since one of the layers is in the Gd-aligned state ($T < T_{\mathrm{comp}}$) and the other is in the Co-aligned state ($T > T_{\mathrm{comp}}$) irrespective of the coercive hardness–softness relationship. At 200 K, which corresponds to the lower T_{comp}, the second layer switches from the Co-aligned state to the Gd-aligned state. Further decrease of the temperature below 200 K enhances the total magnetic moment where both layers are in the Gd-aligned configuration.

On heating from low temperatures exactly the same steps are taken in reverse; however, an interface wall is created in the hard layer which now corresponds to the lower T_{comp} at 200 K. Therefore the path of the total magnetization is not the same for the cooling and heating cycles. Hence, this thermal cycling creates a new form of thermal hysteresis. This new form of thermal hysteresis is not yet fully understood, although it very likely involves an interface domain wall. As the external magnetic field increases from 3 kOe up to 10 kOe, the tunable width of the thermal hysteresis increases from (4 K; 2 K) on each side of the mid-point (220 K) at 3 kOe to (5 K; 2.5 K) at 4 kOe and to (8 K; 5 K) at 5 kOe, as shown in figure 5. The existence of this new form of thermal hysteresis is confined to the temperature interval between the minima of the thermal hysteresis curve which is assumed to correspond to the T_{comp}s of the individual layers in the binary ECDL structure. As can be seen from figure 5, the corresponding lower T_{comp}

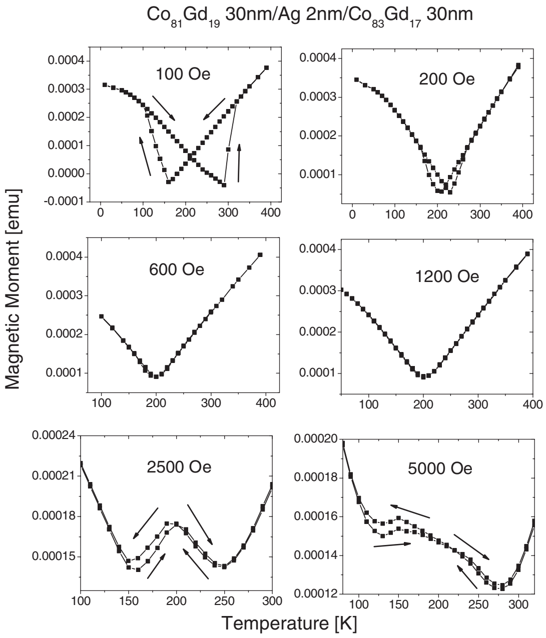

Figure 6. Thermal hysteresis at various external magnetic fields for the [$Co_{81}Gd_{19}$ 30 nm/ Ag 2 nm/$Co_{83}Gd_{17}$ 30 nm] system.

vanishes as the external field increases up to 10 kOe; this is due to the fact that sufficiently high external fields are able to break the antiferromagnetic coupling between the layers (not within the layers) and align each layer to the field direction (saturation). This can be seen from the saturation field values of figure 4.

Similarly as shown in figure 5, the temperature interval between the minima of the second form of thermal hysteresis increases from 60 K at 3 kOe to 85 K at 4 kOe and finally to 100 K as the external field increases to 5 kOe. Therefore, T_{comp} is a function of applied external magnetic field. In other words, the ECDL shows one or two T_{comp}s depending on the external field strength and compensation occurs at relatively different temperatures for the same reason.

The same experiment was repeated for the [$Co_{81}Gd_{19}$ 30 nm/Ag 2 nm/$Co_{83}Gd_{17}$ 30 nm] ECDL system with a 2 nm nonmagnetic Ag layer deposited in between the alloy layers as shown in figure 6. The reason for putting the 2 nm Ag in between is to test whether the observed thermal hysteresis is due to the exchange coupling between the alloy layers or not. The idea was that if 2 nm Ag is placed in between the layers it should kill the coupling and the results would be different (i.e. if there were no coupling between the alloy layers it would not be possible to form a thermal hysteresis of second kind at high external magnetic fields and the hysteresis loops would look like the schematic one shown in figure 3). However, the thermal hysteresis for the [$Co_{81}Gd_{19}$ 30 nm/Ag 2 nm/$Co_{83}Gd_{17}$ 30 nm] system is similar to the one without the 2 nm Ag spacer. Associated thermal hysteresis has occurred at considerably lower external magnetic fields due to weaker exchange coupling between the layers. For example, comparing figure 5 with figure 6 at 200 Oe the width of the bow-tie is 80 K for the binary system without the Ag spacer as shown in figure 5, whereas it is 10 K for the one with 2 nm Ag spacer as shown in figure 6. This observation of exchange coupling through a nonmagnetic spacer is in agreement with Skomski [39] that strongly reduced interface exchange may still yield substantial coupling. The mid-point ($\sim$200 K) of thermal hysteresis at 2.5 kOe in figure 6 matches with the average T_{comp} of the individual layers (150 and 250 K) combined as shown in figure 2. Here we demonstrate a nice example of coupling through a nonmagnetic spacer for our purposes; however, that is another big field of research by itself and is beyond the scope of this paper.

Observation of two different tunable thermal hystereses provides a profound way of looking into the thermo-magnetic properties of ECDLs, since the reversal of one of the layers has been adjusted with respect to the temperature dependence of the coercivity of the individual ferrimagnetic layers and the energy of the interface wall. (Compare the field values of maximum 0.4 kOe as shown in figure 2 and greater than 3 kOe as shown in figure 5. The difference between the field values of the order of several kOe between figures 2 and 5 is required to overcome the strong antiferromagnetic Co–Gd exchange coupling.) The hard layer reversal requirement and its consequences are directly shown for the first time here. Although the ECDL geometry we used has in-plane magnetization due to our measurement capabilities, the physics is the same when the magnetization is perpendicular to the plane as is the case in magneto-optical recording.

We anticipate that some form of thermal hysteresis may appear if a binary magnetic system exhibits strong temperature dependence in magnetization and/or decoupling in field hysteresis. In a recent paper [40] thermal hysteresis of an antiferromagnetic/ferromagnetic binary system was shown. In particular, thermal cycling may have dramatic effects even if there is a very small field or no external field at all, such that magnetic anisotropy of the system can stabilize the initial magnetic configuration in the entire temperature range (see figure 2(a) of [2]).

In conclusion, a new type of tunable magnetic thermal hysteresis is observed under high external fields for binary ECDL type structures. This is due to the change of the magnetic alignment (ferro to antiferro and back to ferro) more than once in a large temperature interval and subsequently the creation of interface domain walls between the alloy layers. Characteristic bow-tie thermal hysteresis is also observed under low external magnetic fields. Finally, both forms of thermal hysteresis are tunable with the change of external magnetic field.

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