Hysteresis Loops in Diluted Antiferromagnetic Films

Zhongquan Mao¹⁺, Xiaozhi Zhan², Jiang Zhang¹ and Xi Chen¹

¹School of physics and optoelectronics, South China University of Technology, Guangzhou 510640, China
²Institute of High Energy Physics, Chinese Academy of Sciences (CAS), Beijing 100049, China
*Email: zhqmao@scut.edu.cn

Abstract. The hysteresis loops of diluted antiferromagnetic (DAF) thin films are studied by means of Monte Carlo simulations. The major hysteresis loops exhibit rich variety of shapes. A diagram of shape is demonstrated. Loop shift and coercivity enhancement are observed exclusively in field-cooling minor loops, which indicates that the effects are not the intrinsic properties of the DAF thin films. The numerical results may help to understand the stable mechanism of the uncompensated spins in the DAF layer and exchange bias systems.

1. Introduction

Magnetic hysteresis is caused by the delay of a system in responding to the magnetic field. It is an effect of non-equilibrium and commonly observed in ferromagnetic and ferrimagnetic materials. Interestingly many anomalous hysteresis loops are discovered in antiferromagnetic (AF) materials during the last two decades. In pure AF materials, butterfly hysteresis loops are reported in V₁₅ molecular complex [1] and molecular ferric wheel NaFe₆ [2]. Another way to obtained hysteresis loops in pure AF materials is to reduce their size in order to obtain surplus spins, as predicted by Néel [3]. In AF nanoparticles, measurable remanence and coercivity occur also due to either the thermal activating procession of spins [4, 5] or different reversal paths resulting from the finite size effects [6, 7]. For bulk and film AF materials, hysteresis phenomenon appears when the system becomes disordered by inducing random exchange bonds, non-magnetic defects or vacancies [8-10]. If the non-magnetic defect ions are diluted in sites of the magnetic ions, the AF system becomes a diluted AF (DAF) system, which can be mapped on the same class as the random field systems [10, 11]. While the relaxation and the critical phenomena in DAF systems are investigated intensively and well understood [11], the hysteresis phenomenon have received less consideration [12]. Perhaps the most-known knowledge about the hysteresis of DAF is that net remanence can be ascribed to the surplus spins within the frozen AF domains which were evidenced in the magnetization steps in the magnetization curves [10]. But the evolution of the hysteresis phenomenon with dilution in finite temperature remains unexplored.

On the other hand, nonmagnetic defect plays significant role in pinning the ferromagnetic thin film by the exchange bias (EB) effect [13]. It is thus assumed in the domains state model that the EB originates from the interfacial uncompensated spins (UCS) that caused by the non-magnetic defects well inside the DAF layer [14, 15]. An important problem is that in what circumstance the interfacial UCS can be stabilized under reversal field. Furthermore, it is also shown by Benitez et al that the two-dimensional (2D) DAF shells of the AF nanoparticles exhibit remarkable coercivity enhancement and loop shift [16, 17]. The authors attributed the phenomenon to the frozen net magnetization within the shell alone. Controversy arises because many other works suggest the origin of an EB effect coming
from the exchange coupling between the DAF shells and the AF cores [18-20]. This relates to the question that whether the loop shift and coercivity enhancement are intrinsic in a 2D DAF system or not. In this work, we present extensive calculation of the hysteresis properties for the DAF thin films and study their possible roles on the EB effect.

2. Numerical Model
The DAF thin films in the Monte Carlo simulations consisted of a simple-cubic lattice occupied with Heisenberg spins. The calculated size of the x-y plane was 100×100, and the extension in z direction is of four monolayers. Further increasing of the thickness does not influence the hysteresis properties substantially [21, 22]. Periodic boundary condition was applied in the x-y plane and free boundary condition was used along the z direction. By considering the nearest-neighbour interaction with an AF coupling $J_{AF}$, the Hamiltonian in unit of $J_{AF}$ is written as:

$$H = \sum_{<ij>} e_i e_j S_i \cdot S_j - h \cdot \sum_i e_i S_i - k \sum_i e_i S_i^2$$

where unit vectors $S_i$ denotes the spin orientation. The terms from left to right represent the reduced exchange energy, Zeeman energy and anisotropy energy, respectively. The field is applied along the +x direction, while the easy axis of the spin parallels to the x axis. We used $k = 0.1$, which corresponding roughly to a ratio of anisotropic energy ($5.2 \times 10^{-22}$ J/atom [23]) and exchange energy ($4.2 \times 10^{-21}$ J/link [24]) for CoO. The factor $e_i = 1$ or 0 depending on whether the $i$th site is occupied by a spin or not. Dilution $p$ denotes the probability of randomly occupied non-magnetic defects. The spin configurations were simulated by standard Metropolis algorithm. The spin was updated in a small variation around the initial spin [25]. Zero field cooling (ZFC) and field cooling (FC) loops were concerned. In a ZFC/FC experiment, the system was first demagnetized at high temperature and then cooled down without/with an external field. After that the magnetization curves were measured.

3. Results and Discussion
Figure 1 (a) shows the temperature effects for a low-diluting film. At high temperature the system demonstrates typical linear field-dependent magnetization. As the film was cooled to low temperature, an abrupt jump takes place at around $h \sim 1$, indicating a spin-flop transition which arises when the field is applied along the easy axis of the spins. Accompanying with the spin-flop transition is the emergence of the hysteresis behaviour. The curves show inversed ‘S’ shape hysteresis loops before an equilibrium field $h_{eq}$ (indicates by arrow in figure 1(a)), higher than which the films become equilibrium and the hysteresis phenomenon disappears. By adding more defects to the films, the spin-flop transition disappears gradually. Simultaneously the loops change from the inversed ‘S’ shape to the ‘S’ shape, as shown in figure 1(b). The $h_{eq}$ decreases monotonically. The corresponding changes of the remanence and coercivity with defects are displayed in figure 1(c). Both remanence and coercivity exhibit a non-monotonic variation with a maximum value. But the peak position of the coercivity is at much smaller dilution than that of the remanence.
Figure 1. (Colour online) Dilution and temperature dependent ZFC hysteresis behaviours of the DAF thin films. (a) Temperature dependence of the magnetization curves. (b) Evolution of hysteresis loops. (c) Dilution dependent coercivity and remanence. (d) Step-shape loops at low temperature. The inset of (d) shows dilution dependence of the step loops at $t = 0.01$. The arrow marks the position of the steps.

The shape transition and non-monotonic dependence of the remanence and coercivity could be understood in terms of the development of the AF domain structures. With few defects, the film maintains a long-range AF system, which makes the spin-flop transition rather remarkable. Since the inverted ‘S’ loops are accompanying with the spin-flop transition, the inverted ‘S’ loops dominate in low dilution region. As the defects increase, the film is divided into AF domains separating by walls or broken exchange coupling bonds [26]. In such domain state the broken coupling bonds cause random fields and spin frustrations, giving rise to the slow relaxation of the system [11, 26]. When the film is subjected to a circling field, hysteresis behaviour arises. The magnetizing of the system is composed primarily of the rotation and reversal of the domains. In this case, the spins within the same domain tend to rotate coherently. Consequently, the spin-flop transition vanishes and the loop behaves like ferromagnetic materials. The remanence in a DAF is attributed to the surplus magnetizations of the domains [14], it relates directly to the average size and the number of the domains. Both small number of large domains and large number of small domains will yield a small remanence; thus, a maximum remanence can be obtained in systems with optimal domain size and number at proper dilution. In the same scenario, the initial rapid increase of coercivity can be interpreted by the formation of domains, while the decrease of it could be a result of the reducing domain size.

When an intermediate diluting film is further cooled to very low temperature, steps emerge from the hysteresis loops, as displayed in figure 1 (d). Similar step-shape loops have been reported in other AF materials [19] and EB systems [27]. However, the mechanism of such step remains unclear. In the inset of figure 1(d), we show that the steps do not vary with dilution at extremely low temperature. Hence it may relate to the anisotropy of the spins. In order to gain an intuitive picture for the steps, spin configurations have been simulated. By comparing spin textures recorded at the two plateaus next to the step, we can find out the changing spins in the two states. Figure 2 shows the spin configurations of randomly selected area for the top layer of a $p = 0.6$ film. The other layers exhibit similar properties. To highlight the spins which change their orientations, different colours are used according to the rotating angles. It is clear that, by reducing the field through the step, the isolated spins flip to the opposite directions, while other spins within AF domains rotate with angles smaller than 30°. Therefore, the step is mainly caused by the reversal of isolated spins frozen at very low temperature.
Figure 2. (Colour online) Select-area spin configurations of the top monolayer of a \( p = 0.6 \) film at \( t = 0.01 \). Red colour in (b) highlights the spins with rotating angles larger than 90° with respect to its direction in (a), while yellow colour highlights the spins of rotating angles smaller than 90° but larger than 30°.

Figure 3. (Colour online) Loop shape diagram for DAF thin films.

Figure 3 summarizes the loop shapes in different dilution and temperature. As the defects increase, the spin-flop behaviour is suppressed, and finally disappears at an intermediated dilution. Meanwhile the hysteresis emerges from low dilution, and becomes strong at intermediated dilution before declining in heavily diluting films. The hysteresis region (yellow colour area) is therefore surrounded by the equilibrium region where neither remanence nor coercivity can be detected. The spin-flop region (blue shadow) overlaps partly with the hysteresis region. In the overlapping region the inversed ‘\( S \)’ loops are observed, while in the rest area the ‘\( S \)’ loops are found. The step loops (black shadow) begin at \( p \approx 0.4 \) where there are detectable percentage of isolated spins. The inversed ‘\( S \)’ loops persist to half diluting, which consists well with the experimental data on the Fe\(_{1-p}\)Zn\(_p\)F\(_2\) systems where inversed ‘\( S \)’ loops and ‘\( S \)’ loops are obtained in \( p = 0.52 \) and \( p = 0.75 \), respectively [28]. The inversed ‘\( S \)’ loops and ‘\( S \)’ loops have likewise been reported in many other defect-doping AF materials, such as dopant BiFeO\(_3\)[19]. Although the hysteresis loops of AF materials exhibit a wide variety of shapes, the inversed ‘\( S \)’ shape and the ‘\( S \)’ shape could be the generic shapes.
Figure 4. (Colour online) ZFC (black solid lines) and FC (red dash lines) hysteresis loops of the DAF thin films for (a) inversed ‘S’ loops, (b) ‘S’ loops and (c and d) step loops. The cooling field is $h = 2$

In figure 4 the question of possible loop shift and coercivity enhancement is tested with all the three types of loops. As can be see, both ZFC and FC curves are symmetric and superimposed with each other. Neither shift nor coercivity enhancement is evidenced. We have measured the FC loops in wide ranges of cooling field as well as cooling rate, and similar results are observed. It may argue that if the maximum sweeping field (MSF) is strong enough, all the spins can be dragged to the field direction. In this case, there would be no loop shift. In order to estimate this effect, different MSF have been applied during calculations. It is found that no loop shift (defined as ref [21]) or coercivity enhancement can be obtained as long as the MSF is higher than $h_{eq}$, as shown in the inset of figure 5. In another word, a major hysteresis loop measurement cannot produce loop shift and enlarged coercivity. On the contrary, if the MSF is smaller than the $h_{eq}$, a minor loop is obtained. In this case, the loop shift and coercivity enhancement can be found in the FC curves, as shown in figure 5. Therefore, we can conclude that the loop shift and coercivity enhancement are not intrinsic in the 2D DAF systems, but they occur in minor-loop magnetizing as in ferromagnetic systems.

Figure 5. (Colour online) ZFC and FC minor loops for a $p = 0.7$ DAF thin film. The inset shows the MSF dependence of the loop shift in FC curves. The cooling field is $h = 2$

The result provides an effective way to distinguish the origins of the loop shift or coercivity enhancement in the AF and DAF nanostructures. For example, if the loop shift and coercivity enhancement decreases with the increasing MSF, they may originate mainly from the minor-loop magnetizing of the DAF shell. That is the frozen metastable domains [16, 17]. Otherwise, they may originate from the EB effect due to the exchange coupling between the DAF shell and the AF core [18-20]. In the domains state model [13-15], the surplus spins within the AF domains make a
significant contribution to the UCS. But how these UCS maintains stable during magnetizing under the external field is not discussed. An important finding from figure 4 and figure 5 is that the vertical shift which directly correlates to the frozen UCS appears exclusively in the FC minor loops. Hence it reveals that the UCS can only be stable under external field smaller than the $h_{eq}$ of the DAF layer.

4. Conclusion
In summary, we have presented a numerical model to study the hysteresis loops of the DAF thin films. Major hysteresis loop and minor loop are observed when the maximum sweeping field is stronger and smaller than the equilibrium field, respectively. The full hysteresis loops are found to exhibit rich shapes. A loop-shape phase for the DAF films is demonstrated. We clarify that loop shift and coercivity enhancement cannot occur in major FC hysteresis loops. Instead, a minor-loop magnetizing yields distinct loop shift and enhanced coercivity, which indicates a possible mechanism of how the UCS stabilize within the AF pinning layer of EB systems.

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6. References
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