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Negative magnetic remanence in Co/Mn/Co grown on GaAs(001)

Y. Z. Wu,[1] G. S. Dong,[1] and X. F. Jin[1,2,*]
[1]Surface Physics Laboratory, Fudan University, Shanghai 200433, China
[2]Physics Department, Hong Kong University of Science and Technology, Hong Kong
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Co/wedged Mn/Co sandwiches were grown on GaAs(001) at room temperature. With the magneto-optical Kerr effect technique, magnetic hysteresis loops at different Mn thicknesses ($0 \leqslant d \leqslant 4$ nm) were measured and the negative remanence phenomenon was clearly observed when the magnetic interlayer coupling between the two Co layers was antiferromagnetic. The mechanism can be understood with the help of a well-defined model calculation, and the result indicates that such anomalous hysteresis loops are not frequently encountered because of the critical conditions required to realize them.

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I. INTRODUCTION

Hysteresis has been one of the most important enduring symbols of magnetism. It is the central feature of ferromagnetic materials below the Curie temperature. All applications, from electric motors to transformers and permanent magnets, from various types of electronic devices to magnetic recording, rely heavily on particular aspects of hysteresis. On the other hand, the comprehension of the physical mechanism responsible for hysteresis has attracted the attention of physicists for decades. It is a beautiful example of a physical problem of intriguing elegance and challenging complexity, that is at the same time the source of pervading technological progress.[1]

Hysteresis loops may take a variety of different forms. The two most important parameters to characterize them are the remanence M_r and the coercive field H_c. The remanence represents the magnetization obtained after applying a large magnetic field to a specimen and then removing it. It is the natural quantity expressing the fact that a ferromagnet can be spontaneously magnetized, even in the absence of the external actions. The coercive field is the magnetic field needed to bring the magnetization from the remanence value to zero. It measures the order of magnitude of the field that must be applied to a material in order to reverse its magnetization.

However, these well-established characters of ferromagnetic materials were first challenged by Esho in 1976.[2] It was found that the hysteresis loops for amorphous Gd-Co films showed a strange behavior, i.e., a negative remanence (or negative coercivity) behavior, which meant that the magnetization reversed its direction before the field reaching to zero. In fact, it was later realized that such an anomalous hysteresis behavior was not only found in this particular system but also observed in many other systems.[3–11] They can be divided into two categories: either amorphous films Gd-Co (Refs. 2–5) and (Ni, Fe)-SiO_2,[10] or multilayer systems [Co-O]/Cu, [Co-O]/Al,[7,9] [Ni-Cu]/[Ni-Co],[6] Pd/Ni,[1] and [Co-Pt]/[Gd-Pt].[8] Several mechanisms based on very different effects were proposed to explain this phenomenon, such as the inhomogenety effect,[2–5] the antiferromagnetic coupling effect,[7,8,9,11] and the dipolar-dipolar interaction effect.[10] Among these different mechanisms, the first and third are not so well defined and are difficult to check in experiments, while the second gives a more transparent physical picture which can further be verified.

It is noted that all systems previously explored were somehow too complicated to be well controlled or manipulated in experiments. In addition, all comparisons up to now between the proposed models and experiments were qualitative but not quantitative. Accordingly, we choose to work on a much simpler system, a Co/wedged Mn/Co sandwich structure on a GaAs(001) substrate. Using the magneto-optical Kerr effect (MOKE), a set of magnetic hysteresis curves is obtained along a wedged-shaped magnetic sandwich at different Mn thicknesses. Anomalous hysteresis curves with negative remanence are indeed clearly observed in the region where the two Co layers are antiferromagnetically coupled to each other. In addition, a theoretical simulation using the parameters obtained from experiments is carried out based on the Stoner-Wohlfarth model with uniaxial anisotropies. Based on the good agreement between the experimental and theoretical results, it is believed that the model proposed in Refs. 7 and 8 does contain the main physics to explain the strange magnetic behavior.

This paper is organized as following. In Sec. II we describe, first, the principles of how to design a well-defined structure to show the negative remanence hysteresis loops, then the sample preparation in experiment and a magnetic characterization by the longitudinal magneto-optical Kerr effect. The model calculation and its comparison with the experimental results are presented and discussed in Sec. III. A summary is given in Sec. IV.

II. EXPERIMENT

A qualitative picture can first be outlined for constructing a well-defined system to illustrate the main physics of the negative remanence phenomenon. It is trivial that a negative remanence hysteresis can never occur in a ferromagnetic material with a single domain. The same argument is also true for a ferromagnetic material with multidomains, if they are caused by magnetostatics, since no mechanisms can induce a reversal of the total magnetic moment before removing the magnetic field. However, a negative remanence hysteresis becomes possible in principle for a case where two magnetic materials with unequal magnetic moments ($M_1 > M_2$) are

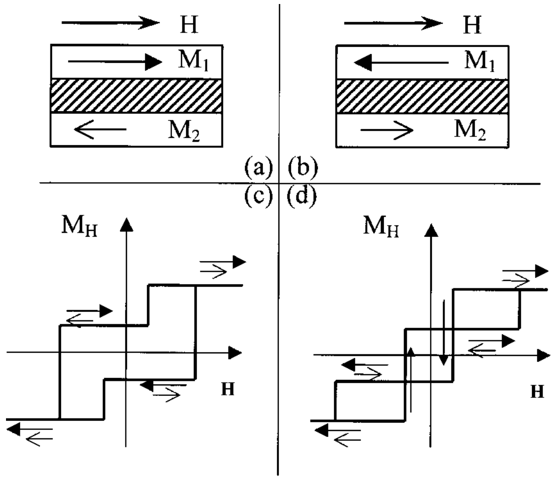

FIG. 1. Schematic hysteresis loops for an antiferromagnetically coupled sandwich with different uniaxial anisotropies. (a) and (c) correspond to $K_1 > K_2$; and (b) and (d) correspond to $K_1 < K_2$.

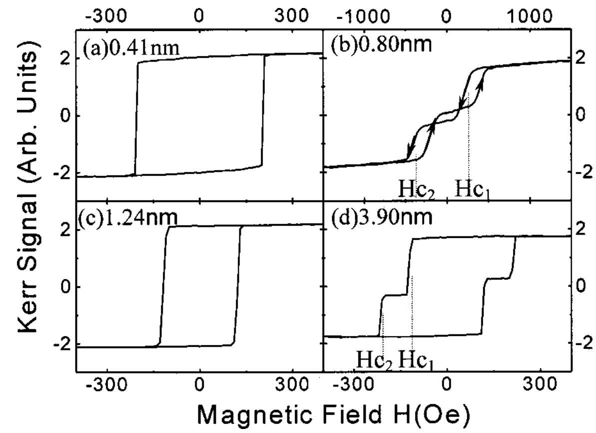

FIG. 2. Representative hysteresis loops measured by MOKE at different Mn thicknesses in a Co/wedged Mn/Co/GaAs(001) sandwich.

coupled antiferromagnetically, i.e., they tend to align their moments antiparallel to each other. In this case, one of the moments might be reversed (with the help of thermal or any other fluctuations) during the removing of a magnetic field to fulfill their antiferromagnetic coupling or to lower the total energy of the system. In fact, this idea can easily be realized by preparing a magnetic sandwich structure, as showed in Fig. 1(a) or 1(b). They consists of two ferromagnetic layers with M_1 and M_2 sandwiched by a nonmagnetic metal. When the thickness of the nonmagnetic metal is adjusted properly, it is known that the two magnetic layers can be coupled antiferromagnetically to each other.[12] In addition, if one now manipulates the sandwich system to display $M_1(>M_2)$ reserves first, as shown in Fig. 1(b) during the removing of magnetic field, then a negative remanence hysteresis loop should indeed be produced, as shown in Fig. 1(d). However, if it is M_2 that reverses first during the removed of the magnetic field as shown in Fig. 1(a), then a hysteresis loop with positive remanence is obtained as shown in Fig. 1(c). In other words, what is needed in this work is to construct a magnetic sandwich with one of the layers exhibiting higher and softer magnetic moments, and the other lower and harder moments. In fact we achieve this goal in the experiment by introducing the uniaxial magnetic anisotropies of K_1 and K_2 for M_1 and M_2 materials respectively, with their magnitudes satisfying $M_1 > M_2$ and $K_1 < K_2$.

The Co/Mn/Co sandwich structures were prepared on GaAs(001) in a molecular-beam-epitaxy (MBE) growth chamber with a base pressure lower than 3×10^{-8} Pa. The Te-doped GaAs(001) single-crystal wafers were treated using H_2SO_4:H_2O_2:H_2O=5:1:1 chemical etching before loading into the MBE system and flashing to 580 °C in the ultrahigh-vacuum system. A clean and ordered surface was achieved as described in Ref. 13. The film thicknesses of Co and Mn were determined *in situ* by a quartz-crystal monitor, and the depositions were kept at about 0.2 nm/min. It was noted earlier that the in-plane magnetic anisotropy of a Co film on GaAs(001) could have either uniaxial or fourfold symmetry depending on the growth temperature.[14] Accordingly, the Co/Mn wedge/Co sandwich structures were grown on GaAs(001) at room temperature. They was then capped on top by a Mn layer before being taken out of the chamber for MOKE measurements. The longitudinal MOKE signal was measured using the S-polarized light from a He-Ne laser. The beam was focused onto the sample surface by an optical lens, and the beam size was about 0.2 mm. The incident angle was about 20°, and the offset angle of the analysis polarizer about 2°. The linear background caused by the Farady effect through the optical lens was subtracted. In-plane magnetic uniaxial anisotropies with easy axis along the ⟨110⟩ direction were indeed confirmed by the longitudinal MOKE measurement for this sandwich structure. In fact, the two Co layers prepared this way yielded different magnetic in-plane anisotropies as expected, since they contact different materials on at least one side. A typical sample, to be discussed in the following, exhibiting the negative remanence hysteresis loops was prepared with detailed parameters: Mn(2 nm)/Co(2.5 nm)/wedged Mn(0–4 nm)/Co(3.5 nm)/GaAs(001). It should be pointed out that all the MOKE data were taken before any irreversible aging effects were realized.

Figure 2 shows four typical hysteresis loops measured along the easy axis of the wedge-shaped sandwich sample at different Mn thickness d_{Mn}. In the thin regime of $d_{Mn} < 0.58$ nm, the loops are rectangle shape, as shown in Fig. 2(a). As the Mn thickness increases, the magnetic saturation field also increases, so that the loops look somehow tilted. The most important thing happens when $0.70\,\text{nm} < d_{Mn} < 0.83$ nm, where the total effective magnetization changes sign before the field reaching zero; therefore, the loop indeed shows a negative remanence as in Fig. 2(b). However, as the Mn thickness further increases to $d_{Mn} > 0.95$ nm, the hysteresis loops again become rectangular, as shown in Fig. 2(c). In the thick regime of $d_{Mn} > 2$ nm, two steplike hysteresis loops start to appear, as shown in Fig. 2(d), which corresponds to the decoupling of the two Co layers. From Fig. 2(d) it can also be seen that the thicker Co layer (contacting the GaAs substrate) has a larger step but a smaller coercivity, which means that the magnetic anisotropy (K_1) of the thicker Co layer (M_1) is indeed weaker than that (K_2) of the thinner one (M_2). This is exactly what is expected

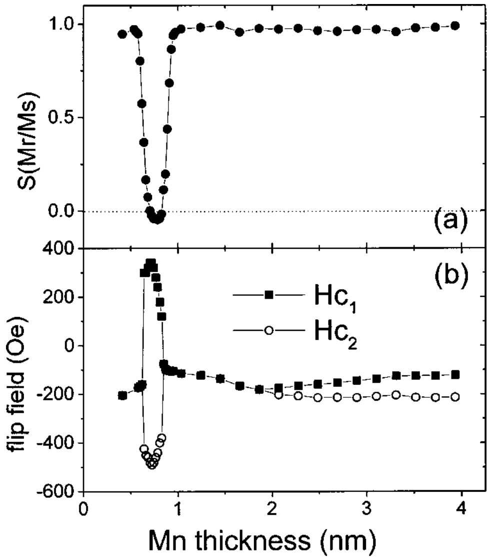

FIG. 3. (a) Mr/Ms ratio as a function of Mn thickness. (b) Flip fields Hc_1 and Hc_2 as a function of Mn thickness in Co/wedged Mn/Co/GaAs(001).

from our foregoing qualitative analysis.

Figure 3(a) shows the ratio of remnant magnetization over saturation magnetization, M_r/M_s as a function of Mn interlayer thickness d_{Mn}. It is seen that the ratio is close to unity in the thin regime, as expected from the ferromagnetic coupling between the two Co layers. Then it decreases significantly starting from $d_{\mathrm{Mn}}>0.59$ nm, corresponding to a turning of the antiferromagnetic layer coupling. It recovers again to unity when the Mn thickness further increases to $d_{\mathrm{Mn}}>0.95$ nm, corresponding to another ferromagnetic layer coupling state. Unlike the general oscillatory behavior of exchange coupling in magnetic sandwiches, such as Co/Cu/Co (Ref. 15), or Fe/Mn/Fe,[16] no further antiferromagnetic layer coupling states except the first are observed in the Co/Mn/Co system. Presumably this caused by the fact that the sandwich quality prepared in this experiment, such as the interface morphology or crystallographic structure, is not good enough to show any exchange coupling oscillations except the first one. The lack of oscillations was actually also obtained in a Co/Mn multilayer system, and speculated to be the interfacial roughness or pinhole effects.[17] It should be noted that, within the antiferromagnetic coupling region 0.59 nm$<d_{\mathrm{Mn}}<0.95$ nm, the M_r/M_s ratio becomes negative when 0.7 nm$<d_{\mathrm{Mn}}<0.83$ nm, as shown in Fig. 3(a) by the points below the dotted line.

In Fig. 3(b), the two magnetic flipping fields Hc_1 and Hc_2 of the two Co layers, as defined as in Fig. 2, are plotted as a function of Mn thickness, respectively. In the thin regime of $d_{\mathrm{Mn}}<0.58$ nm, M_1 and M_2 are strongly ferromagnetically coupled to each other, so that they flip simultaneously, leading to a single value for both Hc_1 and Hc_2. For 0.7 nm$<d_{\mathrm{Mn}}<0.83$ nm, because the thicker Co layer has a weaker uniaxial magnetic anisotropy, the antiferromagnetic exchange coupling causes its moment to flip first before the magnetic field reaches zero. Therefore Hc_1 is positive, and the remanence is negative. The two magnetic flipping fields merge together again as expected when the interlayer exchange coupling comes back to the ferromagnetic regime. However, it is noted that the two flipping fields Hc_1 and Hc_2 separate again after $d_{\mathrm{Mn}}>2$ nm. This separation is certainly not caused by the antiferromagnetic interlayer coupling, but instead is due to the decoupling of the two Co layers to become two independent magnetic layers, as shown in Fig. 2(d). In addition, it should also be noted that the existence of weak ferromagnetic interlayer coupling cannot be completely excluded in a hysteresis loop like Fig. 2(d). However, this contribution can be separated out after some careful analysis. It was found in experiment that the difference between Hc_1 and Hc_2 in Fig. 2(d) increases monotonically as a function of Mn thickness up to $d_{\mathrm{Mn}}\sim3.3$ nm, and is then saturated (not shown here). This indicates that the ferromagnetic layer contribution can be neglected after $d_{\mathrm{Mn}}>3.3$ nm, and the two Co layers become completely decoupled to each other.

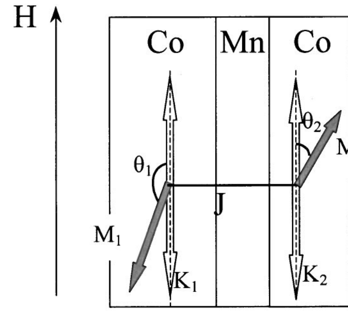

FIG. 4. Schematic model for the Co/Mn/Co sandwich. The symbols are defined in the text.

III. MODEL CALCULATION

In order to further verify the physical picture described above, we have carried out a model calculation using the parameters estimated from the experiment. As shown in Fig. 4, M_1 and M_2 represent the magnetic moments for the 3.5- and 2.5-nm-thick Co layers, respectively. From our experimental measurements, it is known that both Co layers show a uniaxial anisotropy and their easy axes are along the same direction as marked as K_1 and K_2. For simplicity, it is assumed that the magnetic hysteresis loops are dominated by the in-plane coherent rotation of magnetization. Thus the total energy per unit area for the whole system is expressed as

$$E=-M_1d_1H\cos(\theta_1-\theta_H)-M_2d_2H\cos(\theta_2-\theta_H)$$
$$+K_1d_1\sin^2\theta_1+K_2d_2\sin^2\theta_2-J\cos(\theta_1-\theta_2). \quad (1)$$

The first two terms represent the Zeeman contributions of the two FM layers; the next two terms represent the anisotropy energies of the two FM layers, and the last term is the interlayer coupling energy between the two FM layers. θ_1 and θ_2 are the angles between M_1, M_2 and their easy axis. θ_H is the angle of the applied magnetic field. In the calculation the applied field is always kept along the easy axis direction ($\theta_H=0$), as in the real experimental situation of Fig. 2.

Now we try to determine the values of K_1, K_2, M_1, and M_2 in our system. It is noted from Fig. 2(d) that the MOKE signal ratio between the two Co layers is about the same as that of their thickness ratio, indicating the fact that the magnetization values of the two Co layers are equal. It was reported that the magnetization value of Co film grown on GaAs at about 440 K is 1257 G (Ref. 20) or 1150 G.[21] The value is a little lower than the bulk value (1430 G), caused by the interface diffusion between Co and GaAs substrates. The interdiffusion is expected to be much weaker for samples grown at room temperature, so it is reasonably justified to assume that the two Co layers in our case have a bulk magnetization value. As discussed above, the two Co layers are decoupled for the thick Mn interlayer. For a single domain with uniaxial anisotropy, it is known from textbooks that the coercivity is $H_c = 2K_u/M$. Using the bulk magnetization value of Co and the film thickness (2.5 and 3.5 nm), as well as the H_c data from experiment, the magnetic uniaxial anisotropy constants K_1 and K_2 are estimated to be 8.72 $\times 10^3$ and 1.53×10^4 J/m^3 for thicker and thinner Co films, respectively. It is noted that the uniaxial anisotropy of Co/GaAs(001) was determined to be about 8×10^3 J/m^3 in Ref. 22, not too different from what we obtained here. Taking into account the fact that the magnetic anisotropy is generally quite sensitive to the sample preparations, the K values estimated above are presumably reasonable.

Now we have determined four coefficients among the five in the energy expression (1), except for the interlayer coupling strength J. In addition, given the applied magnetic field H (decreasing from large positive value), θ_1 and θ_2 can be determined by minimizing E of Eq. (1):

$$\frac{\partial E}{\partial \theta_1} = 0, \tag{2}$$

$$\frac{\partial E}{\partial \theta_2} = 0. \tag{3}$$

The net magnetization M per unit area at fixed H is obtained from

$$M_H = M_1 d_1 \cos\theta_1 + M_2 d_2 \cos\theta_2 . \tag{4}$$

By gradually changing H from a large positive value to a large negative value, we now obtain half a magnetic hysteresis loop for fixed J. The other half of hysteresis loop can easily be obtained by changing H the other way round. Taking J as an adjustable parameter, in Fig. 5 we calculate the magnetic phase diagram as a function of applied field H and interlayer coupling strength J. The inset shows five different magnetic phases of the system with four collinear states (1), (2), (4), and (5) and one canted state (3). It can be noted that this diagram can be divided into five regions according to different coupling strengths, as marked (a)–(e). The representative loops in the five regions are presented in Fig. 6. The negative remanence loop is realized in case Fig. 6(c). Correspondingly in region (c) of Fig. 5, it is clearly seen that the system changes from state (1) to state (4) then to state (5) when decreasing the field from a large positive value. It is the shadowed area (where M_1 is already reversed before H reaches zero) that gives the negative remanence. Therefore, in this model calculation using experimental parameters, we conclude that there exist the negative remanence loops realized in experiment. However, it can also be understood from this calculation that such abnormal loops are not so frequently observed in experiments, because so many parameters have to be well adjusted to have such a small shadowed area in the magnetic phase diagram. Figures 6(f) and 6(g) shows the experimental loops corresponding to regions (c)–(e) of Fig (5). Furthermore, it is noted that the two calculated loops [Figs. 6(a) and 6(b)] existing in regions (a) and (b) of Fig. 5 are not observed in our experiment. This is due to the fact that the AFM coupling strength in our Co/Mn/Co system cannot reach a value higher than those in region (c) of Fig. 5. However, such kinds of loops were indeed reported in some other systems.[8,18,19]

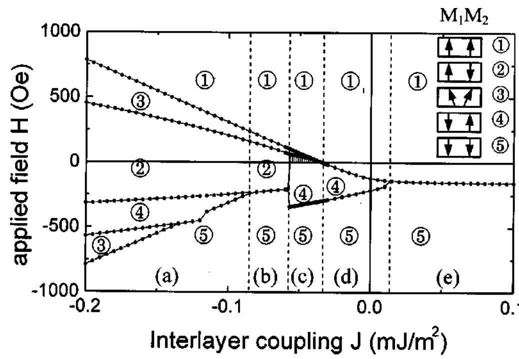

FIG. 5. Magnetic phase diagram as a function of applied field H (decreasing) and interlayer coupling strength J. The inset shows the four possible collinear magnetic states and a canted state.

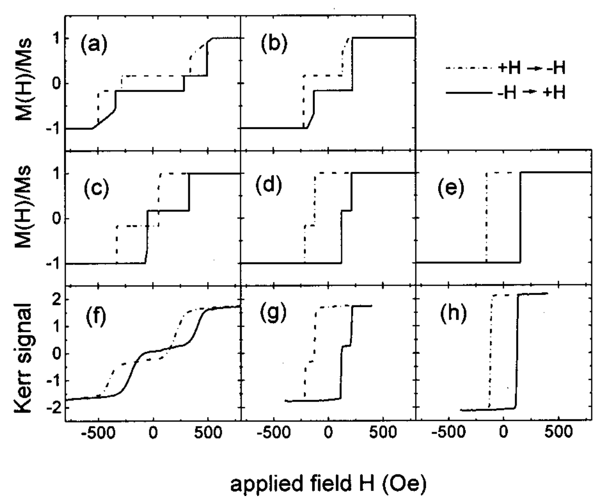

FIG. 6. Representative calculated hysteresis loops for regions (a)–(e) as shown in Fig. 5. The AFM interlayer coupling strength J for each case is (a) -0.15 mJ/m^2, (b) -0.075 mJ/m^2, (c) -0.05 mJ/m^2, (d) 0 mJ/m^2, and (e) 0.05 mJ/m^2, (f)–(h) experimental Kerr loops corresponding to regions (c)–(e) in Fig. 5. The dashed line represents field sweeping from $+$H to $-$H, while the solid line means field sweeping from $-$H to $+$H.

In addition, it is well known from textbooks that the area of a hysteresis loop $W_H=\oint HdM$ represents the energy dissipation per cycle, which must be positive no matter how strange its shape is, in order to satisfy the second law of thermodynamics. It is noted that the abnormal loops with negative remanence realized in our experiment have always one counterclockwise part and two clockwise parts, as shown in Fig. 2(b) or 6(c), but their total areas are always positive. Thus it is quite a puzzle to us to understand why some fully inverted loops could be previously observed [6,7,9–11], if they were not minor loops with aging effect.

Although hysteresis loops with negative remanence can in principle occur according to our model calculation, it is still interesting to check whether the J values in range (c) of Fig. 5 (between -0.058 and -0.034 mJ/m^2), which enable one to observe the negative remanence, can somehow be justified independently.

We now turn to the estimation of AFM coupling strength when the negative remanence is observed as in Fig. 2(b). It is noted from this curve that only M_2 flips when the field reaches Hc_2, corresponding to the transition from state (4) to state (5) in region (c) of Fig. 5. Accordingly, the coercivity Hc_2 can easily be deduced from Eq. (1) as

$$Hc_2=\frac{J}{M_2 d_2}+\frac{2K_2}{M_2}. \tag{5}$$

Using the parameters selected above and the experimental data for Hc_2, the coupling strength J can be estimated to be -0.1 mJ/m^2. Comparing with the previously calculated J values in Fig. 5(c) (between -0.058 and -0.034 mJ/m^2), this has the same order of magnitude but is a factor of 2 larger. Since it is known that the coercivity Hc calculated within the coherent rotation model is normally too simplified in describing the reality, this result obtained here should be regarded as reasonably good. In addition, comparing with the J values estimated in other sandwich systems, such as in a Co/Cu/Co sandwich (-0.16 mJ/m^2),[15] an Fe/Mn/Fe sandwich (-0.6 mJ/m^2),[16] and a Co/Mn multilayer (-0.032 mJ/m^2),[17] the J value we obtained here can also be regarded as reasonable and justified.

IV. CONCLUSIONS

By continually tuning the interface coupling in the Co/wedged-Mn/Co/GaAs(001) system, we observe the anomalous magnetic hysteresis loops with negative remanence when the two Co layers are antiferromagnetic coupled. Using the coherent rotation model, such abnormal hysteresis loops can be well explained. The AFM coupling strength of ~ -0.1 mJ/m^2 is estimated for the Co/Mn/Co sandwich.

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*Corresponding author. Email address: xfjin@fudan.ac.cn

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