Real-time direct observation of asymmetric magnetization reversal in exchange-biased single-layer systems

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We report asymmetric magnetization-reversal behavior in the exchange-biased MnAs film epitaxially grown on GaAs(001) substrate, composed of periodic stripes of ferromagnetic α-MnAs and antiferromagnetic β-MnAs phases existing *in a single layer*. A time-resolved domain observation reveals that the magnetization reverses with a sequence of discrete jumps of a sawtooth-type domain under an applied field parallel to the field-cooling direction, while the magnetization reversal takes place via a sudden single jump under an applied field antiparallel to the field-cooling direction. The asymmetric magnetization reversal in the MnAs system is explained by considering the nonuniform local exchange-biased field, induced by the local fluctuation of the volume ratio of the β phase.

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When a ferromagnet (FM) in contact with an antiferromagnet (AFM) is cooled through the Néel point of the AFM under an applied magnetic field, the hysteresis loop of the FM is shifted from the origin along the magnetic field-cooled axis by an amount known as the exchange-biased field H_E.[1] The exchange-bias effect occurs as the order of the AFM is established via field-cooling in the presence of the FM through the interfacial FM-AFM interaction. Since the AFM spins are generally stable while the FM is rotated, a shift of the hysteresis loop consequently occurs.[2] The exchange-bias effect has received considerable attention because of poor understanding of its microscopic origin as well as because of its technological applications to magnetic sensors, especially utilized in high-density magnetic recording[3] and magnetic random-access memory.[4]

One of the intriguing phenomena related to the exchange-bias effect is asymmetric magnetization reversal in both branches of a hysteresis loop, as recently observed in a variety of systems composed of a FM and an AFM such as NiFe/FeMn,[5] Fe/MnF_2 or FeF_2,[6] NiFe/NiO,[7] CoFe/IrMn,[8] etc. Also, the asymmetric reversal mode in exchange-biased systems[9,10] is theoretically predicted by adopting the domain-state model, which considers domains in an AFM. However, the exact mechanism and underlying physics of this asymmetric reversal are still unclear. So far, most of studies on asymmetric magnetization-reversal behavior have been devoted to the exchange-biased system of bilayered structure, having layer-by-layer contact between a FM and an AFM.[5–11] Here, we present a magnetization-reversal study of a different kind of exchange-biased system, i.e., field-cooled MnAs film,[12] where the exchange-bias effect exists *in the same single layer*, having a stripelike structure consisting of the FM and the AFM.

MnAs film grown on GaAs substrate is a promising system for future spin-injection devices.[13] In this film two structurally distinct phases (hexagonal α MnAs and orthorhombic β MnAs), in the form of self-organized periodic stripes of the two phases, are known to coexist via strain-stabilization.[14–18] The β-MnAs phase could be antiferromagnetic via the fine tuning of the strain profile induced by the adjacent α-MnAs and GaAs substrate. It should be noted that this MnAs system, having a FM α phase and an AFM β phase, provides us with an exchange-biased playground, where stripes of the FM and the AFM exist *in the same layer*, not like bilayer or multilayer FM/AFM systems. In this paper, we carry out a systematic microscopic observation of the domain dynamics during magnetization reversal in each branch of the exchange-biased hysteresis loop and report asymmetric magnetization reversal in this system irrespective of an applied field direction relative to the anisotropy axis of the sample and witnessed via real-time direct-domain observation.

In the present study MnAs film was epitaxially grown in a molecular-beam epitaxy (MBE) system on a GaAs(001) substrate. Before the growth of the MnAs, the substrate was heated to about 600 °C under the As_4 flux for thermal cleaning followed by deposition of a 350 nm undoped GaAs buffer layer. MnAs film with the thickness of about 50 nm was grown on the substrate at about 270 °C with the growth rate of about 0.5 Å/s. The reflection of the high-energy electron diffraction pattern from the film surface became strong and streaky with the annealing. The MnAs film was covered with a very thin (5 nm) GaAs layer at 220 °C in the MBE chamber. The epitaxial orientations of the MnAs film with respect to the GaAs(001) substrate were MnAs$(\bar{1}100)\|$GaAs(001), MnAs[0001]$\|$GaAs[$\bar{1}10$], and MnAs[$11\bar{2}0$]$\|$GaAs[110]. The torque and vibrating-sample magnetometer (VSM) measurements showed that the sample had an in-plane magnetic anisotropy with an easy axis along the MnAs[$11\bar{2}0$] and a hard axis along the MnAs [0001] in

the film plane. The out-of-plane direction of MnAs[$\bar{1}100$] was an intermediate axis.

To investigate the detailed microscopic domain dynamics, time-resolved magnetic domain patterns during the magnetization reversal were directly observed on the sample area of $80\times64\ \mu m^2$ using a magneto-optical microscope magnetometer (MOMM) with $\times500$ magnification capable of capturing domain images with an image-grabbing rate of 30 frames/sec in real time and a spatial resolution of 400 nm, via the longitudinal magneto-optical Kerr effect.[19] A heater capable of heating the sample maximum up to 80 °C was placed at the sample stage for the temperature-dependent domain observation and the Kerr hysteresis-loop measurement. Using the MOMM, we could carry out both real-time domain observation and the Kerr hysteresis loop measurement on the exact same area of the sample.

The strain-stabilized coexistence of the two MnAs phases (α MnAs and β MnAs) in the single-layer film was confirmed from the temperature-dependent x-ray diffraction (XRD) experiment, where the sample temperature was controlled by a radiative heating stage with an accuracy of within ±1 °C in the temperature range of 20–45 °C. We found that two distinguished peaks of α-MnAs and β-MnAs appeared at 20 °C and 45 °C, respectively, and the two peaks coexisted in the intermediate temperature range with the systematic variation of the relative intensities of the two peaks with temperature; the β-MnAs peak was increased with increasing temperature. Hence, two phases are believed to coexist in the temperature range of 25–40 °C. Considering the fact that the structure factors of both phases were almost equal, the ratio of the integrated intensities between the α-MnAs and β-MnAs peaks could be approximated as the ratio of the relative volume fractions of the two phases. To understand the influences of the AFM β-phase volume ratio on the magnetic properties of the FC sample, temperature-dependent hysteresis loops were measured, because the AFM β-phase volume ratio increased with increasing temperature. It was found that H_C increases, but H_E decreases with increasing temperature, i.e., with increasing of the AFM β-phase volume ratio. The magnetization-reversal study of the present work was carried out at the temperature of 26 °C controlled within ±1 °C, where the volume ratio between the α phase and the β phase was estimated to be approximately 9:1. The lateral period of the stripes in the MnAs sample with the thickness of 50 nm was expected to be about 350 nm from the relation between the period and the film thickness.[16,17] It should be mentioned that this estimated value was strongly dependent on temperature, magnetic history, etc.

For field cooling, a magnetic field of 45 Oe was applied along the easy axis of the sample with a temperature variation from 50 °C to 26 °C. Figure 1(a) shows a typical Kerr hysteresis loop of the field-cooled (FC) MnAs film together with that of the zero-field-cooled (ZFC) sample measured at 26 °C. It can be seen that the FC sample exhibits a shifted Kerr hysteresis loop, whereas the ZFC sample exhibits a symmetric loop. The loop shift clearly reveals the existence of exchange bias in the FC MnAs film. The exchange-biased field H_E was found to increase with the strength of the cooling field until its value saturated around the cooling field of 45 Oe. The FC MnAs film in Fig. 1(a) reveals that $H_E=19.9$ Oe, and the coercivity $H_C=79.7$ Oe were obtained using the relations of $H_E=(H_{C1}+H_{C2})/2$ and $H_C=(H_{C1}-H_{C2})/2$, where H_{C1} and H_{C2} are the coercivities in the increasing and decreasing field branches. In contrast, the ZFC sample exhibited a zero-exchange-biased field and a slightly smaller coercivity of 75.1 Oe.

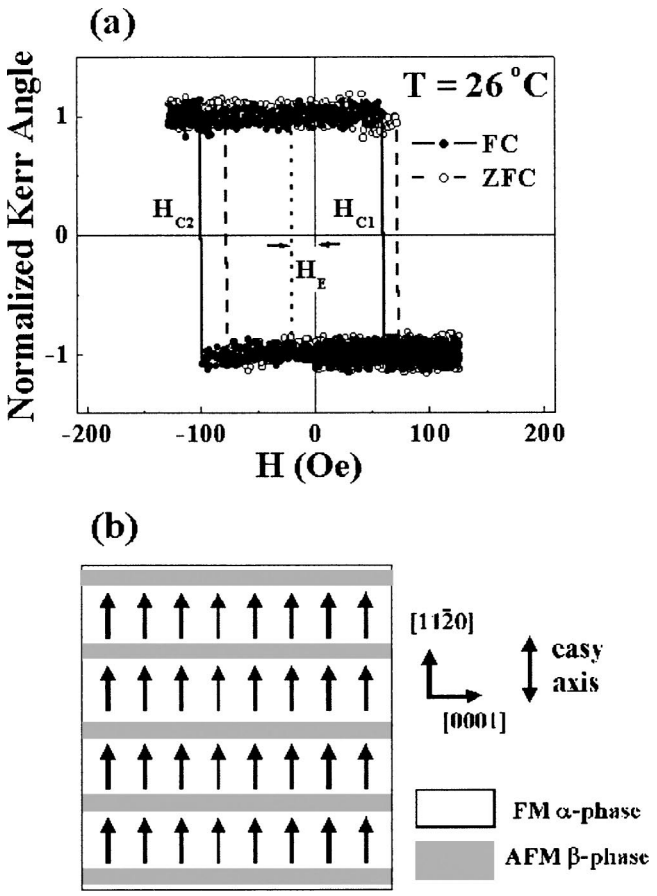

FIG. 1. (a) Typical Kerr hysteresis loops of the MnAs film at $T=26$ °C under the FC and ZFC conditions. Here, H_{C1} and H_{C2} denote the coercivities in the increasing and the decreasing field branches, respectively, and H_E is the exchange-biased field. (b) A schematic representation of the FC MnAs film with the striped patterns of the FM α phase and the AFM β phase.

The exchange-bias effect observed in the FC MnAs film implies an existence of the AFM state in this film. An interesting question is that of where the AFM state is located. The α phase is known to be ferromagnetic.[20] According to the magnetic phase diagram of the bulk MnAs, the β phase can be antiferromagnetic under pressures of 3–10 kbars.[21,22] Since the α phase and the β phase in MnAs film were strain stabilized, a large stress should exist due to the 7.5% and 6.5% lattice mismatches between the two phases and the GaAs substrate, respectively. Therefore, it is not surprising to imagine the antiferromagnetic state in the β phase. Hence, we believe that the FC MnAs film in the present study is an exchange-biased system having a FM α phase with a unidirectional anisotropy induced by the field-cooled AFM β phase.[11,12] Considering the stripelike structure of the film in

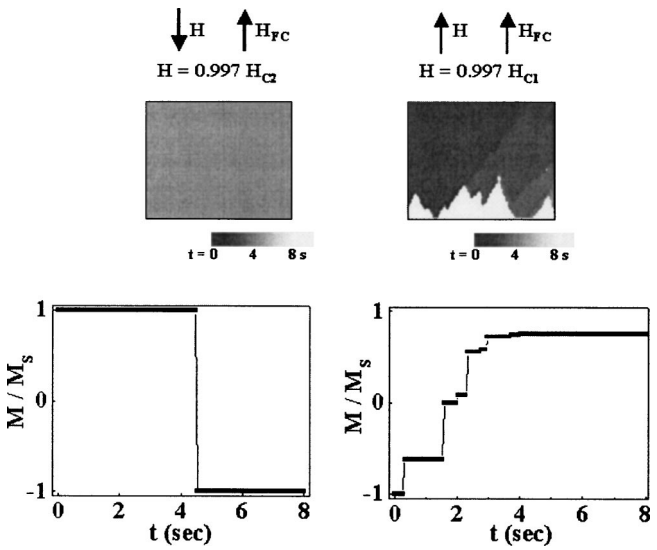

FIG. 2. The domain reversal patterns and magnetization-reversal curves in both field directions in the FC MnAs film at $T=26\,^{\circ}\mathrm{C}$.

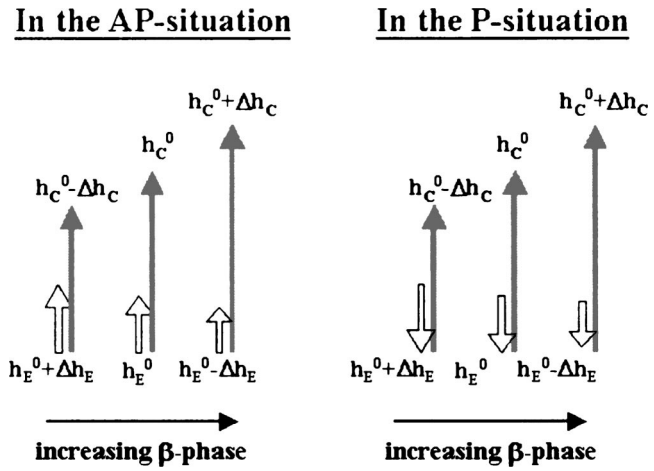

FIG. 3. A simple model for the explanation of the asymmetric magnetization reversal in the FC MnAs film.

which the α and β phases are intercalated periodically with each other as mentioned earlier, we envision the two-phase coexistence structure as schematically drawn in Fig. 1(b). Note that the easy axis along the MnAs$[1\bar{1}20]$ is perpendicular to the interface between two phases. A detailed investigation of the spin configuration in the AFM β phase is interesting and crucial to clarify whether the exchange bias in this system is ascribed to a perpendicular coupling at the FM/AFM interface,[23,24] which is beyond the scope of this study.

Strikingly, the asymmetric magnetization-reversal behavior is witnessed in the field-cooled MnAs film, dependent on the applied field orientation relative to the FC direction. Real-time domain evolution patterns under an applied field near the coercivities in both branches of the hysteresis loop in Fig. 1(a) are demonstrated in Fig. 2, along with the corresponding magnetization-reversal curves. The gray level from black to white indicates the elapsed time during 8, according to the gray palette at the bottom of the figure. The domain evolution patterns in Fig. 2 were obtained by taking the Kerr signals every 63 ms under a magnetic field either parallel or antiparallel to the field-cooled direction for an initially saturated sample. An applied field was kept at 99.7% of the coercive field in each branch for the direct comparison of magnetization-reversal behavior. As clearly seen in Fig. 2, magnetization reversal proceeds via sawtooth-type domain-wall (DW) propagation with a sequence of discrete jumps when an applied field is parallel (P) to the FC direction (P situation). Contrastingly, an abrupt magnetization reversal takes place with a sudden single jump under an applied field antiparallel (AP) to the FC direction (AP situation).

Two interpretations for the abrupt magnetization reversal observed in the AP situation are conceivable; either coherent rotation or fast domain-wall motion occurred beyond the time resolution of our experiment. To clarify this ambiguity, we have investigated the magnetization reversal on the sample area where the line defect existed. We observed that the domain wall was pinned at the line defect and that magnetization reversal was blocked without further processing to the other side of the line defect. If magnetization reversal were governed by coherent rotation, it would abruptly take place on the whole area of the sample around the line defect. Therefore, the magnetization reversals in both field directions are understood by DW propagation; fast domain-wall motion governs magnetization reversal in the AP situation, while slow domain-wall motion occurs in the P situation. So, it can be concluded that the exchange-bias effect on the magnetization reversal of the FC MnAs film only affects the domain-wall velocity of sawtooth-type domains rather than the magnetization-reversal mode itself. To experimentally confirm that the asymmetric magnetization-reversal behavior in the field-cooled MnAs film is indeed ascribed to the exchange-bias effect, we examined the magnetization-reversal behavior of the exactly identical area of the sample under the ZFC condition, and found that it exhibited no exchange-bias effect. The magnetization-reversal behavior was observed to be symmetric in both branches of a hysteresis loop for the ZFC sample. Thus, it is believed that the exchange-bias effect in the FC sample is truly a cause of the observed asymmetric magnetization.

To explain the asymmetric magnetization reversal observed in the FC sample, we propose a simple model, considering the nonuniform β-phase volume ratio. An atomic force microscopy study in the MnAs film shows that the β-phase volume ratio is distributed with fluctuation. Due to this nonuniformity, we can imagine that the local coercivity h_C and the local exchange-biased field h_E are varied in the sample, with the fluctuations of Δh_C and Δh_E from the average values of $h_C{}^0$ and $h_E{}^0$, respectively, as depicted in Fig. 3. In both field directions of the hysteresis loop, the local coercivities h_C, and the local exchange-biased fields h_E of the three representative local areas with increasing AFM β phases are schematically drawn, where h_C and h_E are denoted by the gray and white arrows, respectively. Since the magnetization reversal in the sample is governed by the DW motion, the coercivity is closely related with the energy barrier for reversal; a larger energy barrier is expected for a higher coercivity. So, the local energy barrier in the MnAs system is expected to increase with an increase in the β-phase volume ratio, because a higher local coercivity exists for a larger β-phase volume fraction.

For further discussion, we define the local energy barrier field as the applied field required to overcome the local energy barrier. The local energy barrier field h_B is then given by $h_B=(h_C \pm h_E)$, where the positive and negative signs correspond to the AP situation and the P situation, respectively. If the DW nucleates and moves at the minimal h_B, the effective energy-barrier field during the DW motion Δh_B would be the difference between the average and minimal energy-barrier fields. Hence, the effective energy-barrier field in the P situation is given by $\Delta h_B=\Delta h_C+\Delta h_E$, while in the AP situation it is given by $\Delta h_B=\Delta h_C-\Delta h_E$. Since the Δh_B in the P situation is large compared to the Δh_B in the AP situation, the magnetization reversal in the P situation results in a slow DW motion with a sequence of discrete jumps. Contrastingly, the magnetization reversal in the AP situation yields fast DW motion with a sudden single jump. So, the asymmetric magnetization reversal in the FC sample is ascribed to the difference in the effective energy barrier in both branches due to the local fluctuation of the exchange-biased field, induced by the nonuniform β-phase volume ratio. If the β-phase volume ratio were perfectly uniform, the asymmetric magnetization reversal in the FC sample would not be observed, because there is no local fluctuation of the exchange-biased field. Using this model, we can also explain the symmetric magnetization reversal observed in the ZFC sample. Since the exchange-biased field in the ZFC sample does not exist, the effective energy-barrier field Δh_B is the same in both branches. Therefore, one could expect the symmetric magnetization reversal, even though the local coercivity h_C fluctuates due to the nonuniform β-phase volume ratio.

Interestingly enough, the sawtooth domain in the FC MnAs film always propagates along the $[11\bar{2}0]$ easy-axis orientation and the sawtooth angle 2ϕ is nearly constant, irrespective of an applied field direction relative to the easy-axis orientation, as demonstrated in Figs. 4(a) and 4(b), respectively. These results are understandable, considering the fact that the anisotropy energy ($K_{\|}=5.7\times10^6$ erg/cc) is by an order of 2 larger than a typical Zeeman energy ($M_sH=5.0\times10^4$ erg/cc) in this film, and thus, the domain-reversal behavior naturally appears to be independent of an applied field orientation. If the magnetization directions of the two domains meet head-on, their separating domain walls generally develop a zigzag shape to reduce the magnetic charge density, and the detailed shape is determined by a minimization of the total energy consisting of the magnetostatic energy, the anisotropy energy, and the DW energy. For a theoretical estimation of the sawtooth angle 2ϕ we consider a simple sawtooth-type DW structure, in which the angle between the magnetization direction and the easy axis is assumed to be small, as revealed in our experiment. Then, considering the uniformly distributed magnetic charge, the magnetostatic energy per unit length perpendicular to the easy axis is approximately expressed by $8M_s^2D^2[3-2\ln(2B/s)]$, where $2D$ is the sample thickness, $2B$ is the sawtooth height, and $2s$ is the sample size. Also, the anisotropy energy per unit length is approximately given by

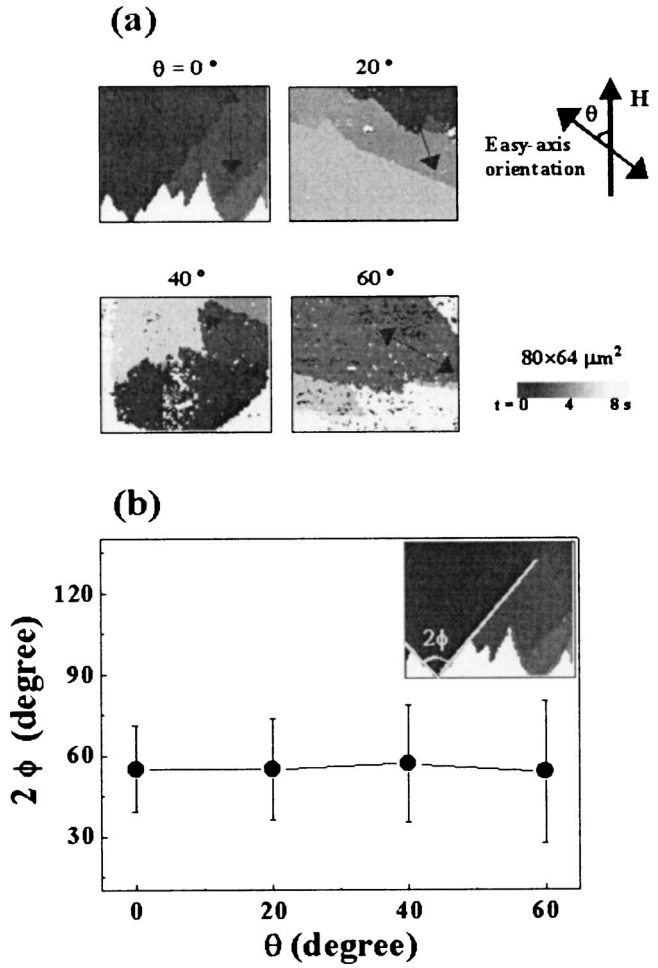

FIG. 4. (a) The domain reversal patterns and (b) the average values of the sawtooth angles with varying the applied field direction relative to the easy-axis orientation. The inset in (b) shows the definition of the sawtooth domain-wall angle 2ϕ.

$(4/3)DK_{\|}B\phi^2$ for a fanlike configuration. Since an anisotropy constant in the film plane, $K_{\|}$, is larger than one normal to the film plane, $K_{\perp}$, in our MnAs film it is reasonable to consider the Bloch wall,[18] and thus, the domain-wall energy per unit length can be given by $2\pi D(AK_{\perp})^{1/2}/\phi$, where A is the exchange stiffness. Then, the sawtooth angle 2ϕ is given by $2\phi=\pi(AK_{\perp})^{1/2}/8M_s^2D$ from energy minimization. For an estimation of the sawtooth angle in the MnAs film, we use the measured values of $K_{\perp}=1.7\times10^6$ erg/cc, $Ms=828$ emu/cc, and $D=2.5\times10^{-6}$ cm, and the estimated value of $A=3.5\times10^{-5}$ erg/cm, obtained using the relation of the Bloch domain-wall thickness, $\delta=\pi(A/K_{\perp})^{1/2}$, with a reported value of $\delta\approx80$ nm.[14] The sawtooth angle is estimated to be about 56.8°, which is well matched to the experimental value within the experimental error.

In conclusion, we directly observed the asymmetric magnetization-reversal behavior in the exchange-biased MnAs/GaAs(001) film consisting of the periodic stripes of the FM α phase and the AFM β phase placed *in a single*

layer. The asymmetric magnetization reversal in the MnAs system is ascribed to the nonuniform local exchange-biased field, basically induced by the local fluctuation of the volume ratio of the β phase.

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