Extremely strong domain-wall pinning and spontaneous demagnetization in MnAs(0001) films on GaAs(111)*B*

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We demonstrate a dramatic dependence of the magnetic properties of epitaxial layers on the substrate orientation. MnAs(0001) layers on GaAs(111)*B* contain magnetic moments that cannot be aligned by a magnetic field of 140 kOe, and produce a hysteretic behavior over the entire field range. Depinned magnetic moments demagnetize spontaneously at weak fields. These peculiar properties are related to an antiferromagnetic coupling that emerges at low temperatures, whose origin we attribute to antiphase boundaries. Despite the tiny fraction of the anomalous moments, they give rise to large magnetoresistance effects.

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I. INTRODUCTION

The interaction between crystalline imperfections such as dislocations and the atomic magnetic moments determines structure-sensitive magnetic properties, including coercive field H_c and high-field susceptibility. The dislocations exert a force on domain walls (DWs) and hinder the DW movement, which is a typical mechanism for the magnetization reversal. The relation is manifested in the fact that H_c increases with the square root of the density of dislocations.[1] The maximum value of H_c of the known bulk materials is about 10 kOe, found for SmCo and FePt. A very large saturation field is often found in material systems containing antiphase boundaries (APBs),[2] which are a natural growth defect in epitaxial layers that occur when the rotational and translational symmetries differ between the layers and the substrates, providing a clear evidence that the substrates can dramatically change the material properties. Understanding the nature of APB is important as they are a crucial mechanism for colossal magnetoresistance (MR) effect. The colossal MR can be exploited for read and/or write heads, high-density magnetic storage, and spintronic applications.

In this paper, we investigate the transport properties in MnAs layers grown on GaAs(111)*B*. MnAs is a prospective material for spintronics as it is ferromagnetic at room temperature and can be grown epitaxially on GaAs.[3] The layers grown on the rather unconventionally oriented substrates exhibit remarkably large MR, which is reminiscent of the colossal MR effect. We detect a magnetization component that is not only unsaturated at a magnetic field of 140 kOe, but also produces a hysteretic field dependence over the entire range. We also uncover spontaneous magnetic disordering at weak fields. While the low-temperature magnetic order in bulk MnAs is ferromagnetic, antiferromagnetic-like coupling is identified to develop at temperatures below one-third of the Curie temperature T_C. These peculiar magnetic properties do not take place in MnAs layers grown on GaAs substrates oriented in the typical (001) direction. Dramatic influences of the substrate orientation on the magnetic properties of epitaxial layers are, therefore, demonstrated. We explain the unusual properties in terms of the antiferromagnetism produced by APB. Although it can be easily understood that MnAs/GaAs structures can generate APB as we show below, they have received no attention so far in the analysis of the material properties of the MnAs layers. It is highlighted that MnAs on GaAs provides a different type of APB in comparison to, for instance, Fe_3O_4 on MgO, i.e., they are formed by the sublattice of a nonmagnetic element.

II. EXPERIMENT

Three MnAs layers were grown on GaAs(111)*B* substrates by molecular-beam epitaxy.[3] Qualitatively the same behavior was observed in all the layers, and so we present below only the results obtained from a 50-nm-thick layer grown at a temperature of 250 °C with an As_4:Mn beam equivalent pressure ratio of 117. The growth rate was 20 nm/h. The other two layers having a thickness of 50 and 110 nm were grown at a rate of 200 nm/h. Hexagonal MnAs layers grew on the substrates with the *c* axis being oriented along the growth direction, see Fig. 1(a). The $[\bar{2}110]$ and $[01\bar{1}0]$ directions of MnAs were parallel to the $[\bar{1}10]$ and $[11\bar{2}]$ directions of GaAs, respectively.

Due to the discontinuous change in the lattice constant at the phase transition at T_C between the low-temperature α phase and the high-temperature β phase, α-MnAs layers are

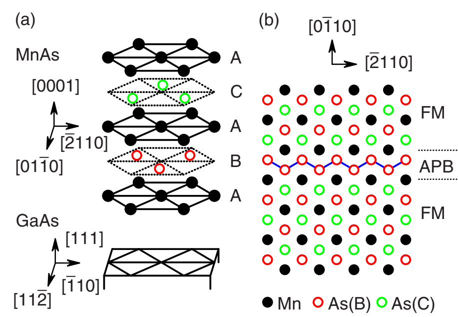

FIG. 1. (Color online) (a) Crystal structure and orientation relationship for a MnAs(0001) layer grown on a GaAs(111)*B* substrate. Mn and As atoms are displayed by the filled and open circles, respectively. The stacking in the growth direction of the hexagonal MnAs is described by the sequence …*ABACA*…. (b) An APB viewed in the (0001) direction. Two ferromagnetic (FM) regions are separated by the APB.

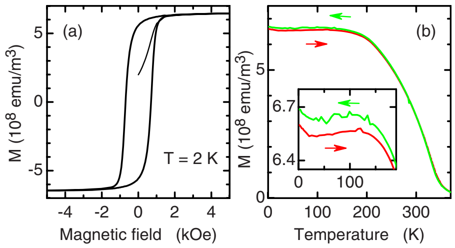

FIG. 2. (Color online) (a) Hysteresis in the reversal of magnetization M at temperature T=2 K. The thin solid curve shows the initial behavior obtained from the zero-field-cooled state. (b) Temperature dependence of magnetization M. After a zero-field cooling, the temperature was increased from 2 to 370 K and then decreased to 2 K, as indicated by the arrows, at a magnetic field of 45 kOe. The low-T part is shown in the inset with a magnified scale. The diamagnetic contribution of the GaAs substrate has been subtracted.

compressively stressed by the substrates.[3] The stress in the layers grown on GaAs(111)B substrates is much larger than that in the layers grown on GaAs(001) substrates, evidenced by the fact that the stress-induced coexistence of the α and β phases[4] ceases for temperatures lower than about 290 K in the MnAs(1$\bar{1}$00)/GaAs(001) system, whereas it persists to temperatures as low as about 160 K in the MnAs(0001)/GaAs(111)B system.[5]

The magnetic hard axis of bulk MnAs is along the c axis, and the C plane is the magnetic easy plane. The magnetization properties of the MnAs layers evaluated using a superconducting quantum interference device (SQUID) magnetometer were practically identical between the [$\bar{2}$110] and [01$\bar{1}$0] directions, indicating negligible magnetic anisotropy produced by the stress from the substrates.[6] We show in Fig. 2(a) the magnetization curve at a temperature of T=2 K. Note that the diamagnetic contribution of the GaAs substrate has been subtracted here.[7]

Hall-bar structures having a channel width of about 30 μm and a distance between voltage leads of about 400 μm were prepared from the epilayers using the lithography technique and Ar ion milling.[8] The resistivity of the MnAs channels was measured using four-probe lock-in technique at T=0.3 K. The excitation current I was 1–10 μA.

III. RESULTS

A. Magnetoresistance

Figure 3(a) shows the dependence of the longitudinal resistivity on an in-plane magnetic field H when the field was applied along the direction of the current flow ($H \parallel I$). The inset shows the MR when the field is orthogonal to the current ($H \perp I$), obtained from a different device. The blue (dark gray) curves are the MR in the first magnetic-field sweep when the devices were cooled from room temperature in the absence of an external field. After the application of the maximum field (140 kOe), the MR was drastically altered to the behavior shown by the red (medium gray) and green (light gray) curves. (The direction and sequence of the magnetic-field sweeps are indicated by the numbered arrows.) The layer is roughly demagnetized in the initial state realized by zero-field cooling (ZFC), and is divided into a large number of magnetic domains. The orientation of the spontaneous magnetization in the domains is random as a consequence of the magnetic isotropy.[3,9] The magnetic field magnetizes the layer by removing DWs. The resultant suppression of the magnetic disorder scattering is responsible for the large decrease in the zero-field resistivity ρ_0. The red (medium gray) and green (light gray) curves were approximately reproduced for subsequent field sweeps between −140 and 140 kOe. To be precise, the repeated field sweeps further reduced the resistivity. The reduction is responsible for the slight asymmetry between the two curves. Remarkably, despite the magnetic field of 140 kOe, hysteretic behavior persists for the entire field range, i.e., the magnetization in the layer is not saturated. As evident in Fig. 2(a), the majority of the magnetic moments flip cooperatively at an ordinary value of H_c. The unsaturated MR is, therefore, produced by a small amount of magnetic moments, a few percent of the total magnetic moments as we show below. Before we identify the anomalous magnetization component, we examine the characteristics of the strong pinning in detail, and reveal another unusual feature of magnetic moments in the MnAs/GaAs(111)B system.

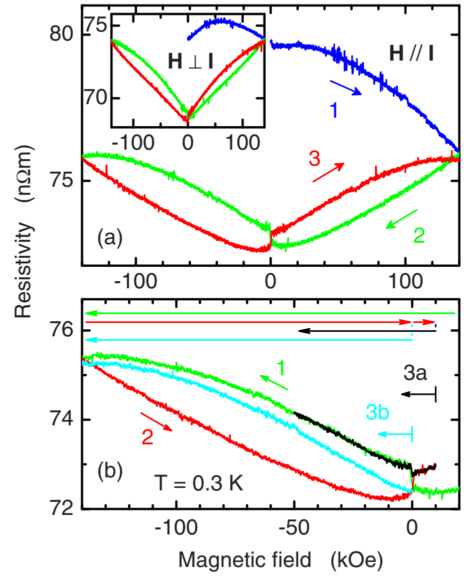

FIG. 3. (Color online) (a) Resistivity at temperature T=0.3 K when an in-plane magnetic field H is parallel to the current I. The numbered arrows indicate the direction and sequence of the field sweeps. The curves colored in blue (dark gray) were taken by the first magnetic-field sweep after a zero-field cooling of the devices. The curves colored in green (light gray) and red (medium gray) show the field dependence obtained in the subsequent sweeps. For the curves in the inset, H and I are orthogonal to each other. (b) Resistivity when the field sweep was reversed to the negative field direction at H=10 kOe (black curve) or 0 kOe [cyan (lighter gray, central) curve]. In addition to the numbered arrows, the sequence of the field sweeps is indicated at the top of the panel. Prior to acquisition of these curves, the field was swept to −140 kOe [green (light gray, upper) curve] and reversed to the positive field direction [red (medium gray, bottom) curve].

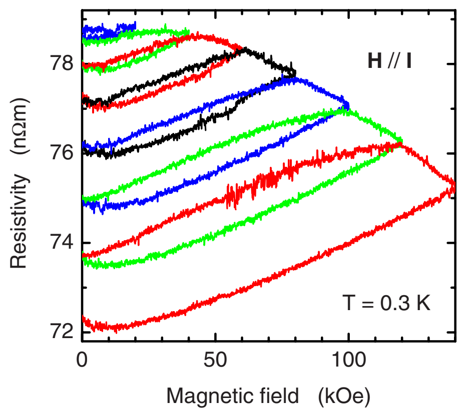

FIG. 4. (Color online) Resistivity at temperature 0.3 K when an in-plane magnetic field H parallel to the current I was repeatedly varied from zero to a maximum magnetic field H_{max} and then back to zero. For the series of measurement runs following a zero-field cooling, H_{max} was increased from 20 to 140 kOe at steps of 20 kOe.

The decrease of ρ_0 evidences permanent partial ordering of magnetic moments imprinted by the magnetic field. In Fig. 4, a series of MR curves were taken following a ZFC of samples by sweeping the magnetic field repeatedly between zero and a maximum magnetic field H_{max}. The value of H_{max} was increased at steps of 20 kOe. As we plot in Fig. 5, ρ_0 decreases gradually with increasing H_{max}. The absence of notable dependence on the angle θ between the field and the current indicates that the conventional anisotropic magnetoresistance effect does not play a role. The decrease in ρ_0 gets stronger when the MR turns negative in high magnetic fields. No tendency of magnetization saturation is observed in Figs. 3 and 4. Note, in particular, the large difference in the MR between $H_{max}=120$ and 140 kOe. The magnetic field required to fully magnetize the MnAs layer is, thus, suggested to be much higher than 140 kOe.

As shown in Fig. 4, the resistivity increases with magnetic field until the field reaches the maximum value of H_{max} in the preceding runs. If the magnetic field is further increased, the MR is suddenly governed by the negative field dependence that characterizes the "virgin" sweep. The abrupt change in the field dependence can be understood if the DWs defined by the magnetic domains having the initial random magnetization direction, which we refer to as initial DWs, are inactive and contribute to the transport properties only as a field-independent resistivity increase. The initial DWs are depinned when the force from the field of H_{max} exceeds their pinning forces, leading to the high-field negative MR.

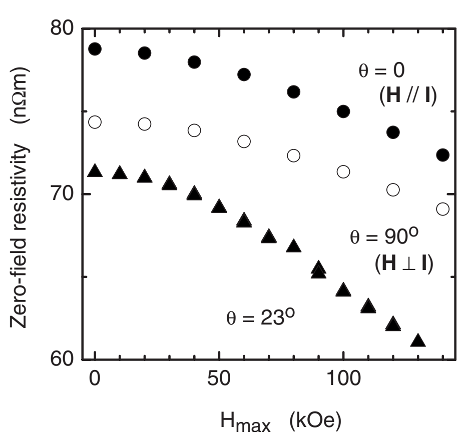

FIG. 5. Variation of the zero-field resistivity ρ_0 with H_{max}. The data were acquired from three devices having an angle θ between H and I of 0 (filled circles), 23° (triangles), and 90° (open circles).

Regarding the ordinary magnetization component, we find a jump in the resistivity at $H_c \approx 0.6$ kOe (Fig. 3). The flip of magnetization at H_c, from antiparallel to parallel orientation with respect to H, takes place by means of a propagation of DWs. (These DWs, which we refer to as regular DWs, replace the initial random DWs once the layer has been magnetized.) The pinning centers that have already released the initial DWs capture the regular DWs, and so the resistivity increases abruptly. The pinned regular DWs are released during the increase of the field to H_{max}. Thus, the resistivity for decreasing field is smaller than that for increasing field. Although the MR for $|H| < H_{max}$ is determined by the regular DWs, the hysteresis closes only at $H=H_{max}$. The fields required to depin the initial and regular DWs are, hence, suggested to be similar. This can be interpreted to mean that not the type of the DWs which is pinned, but the presence of anomalous pinning centers is essential for the high-field hysteresis. When the initial DWs are pinned, the associated magnetic moments are randomly oriented, resulting in a large scattering of conduction carriers. In contrast, the magnetic moments in the domains forming the regular DWs are parallel or antiparallel to the external magnetic field, giving rise to only a small magnetoresistance.

We now confirm that a single magnetization reversal at H_c generates the maximally pinned configuration of DWs for a given value of H_{max}. For the black curve in Fig. 3(b), the field sweep was reversed to the negative direction at $H = 10$ kOe ($\gg H_c$). (The sequence of the magnetic-field sweeps is indicated at the top of the panel.) The MR curve in the sweep down is indistinguishable from that in the sweep up [red (medium gray, bottom) curve] for $H_c < H < 10$ kOe, i.e., the field of 10 kOe is hardly enough to free the pinned regular DWs. After the magnetization flip of the ordinary component at $-H_c$, the MR curve is again identical to that of the high resistivity state [green (light gray, upper) curve]. Note the sharp increase in the resistivity, while H varies from 0 to $-H_c$. The resistivity step occurs when the magnetic field is swept around zero in a range stronger than $\pm H_c$. The repeated weak-field sweeps increase the number of pinned regular DWs by subjecting the pinning centers to DW movements. Thermal activation processes can also introduce DWs into the magnetized state. The time dependence of ρ_0 with $H_{max} = 140$ kOe, shown in Fig. 6, reveals the typical logarithmic relaxation.

In the course of a field sweep down from H_{max} in Fig. 4, the resistivity develops a minimum at $H \approx \pm 10$ kOe before reaching zero field. This suggests that some magnetic moments deviate away from the dominant magnetization direction around $H=0$ as the external field can no longer force them to be aligned. This interpretation is supported by the behavior shown by the cyan (lighter gray, central) curve in

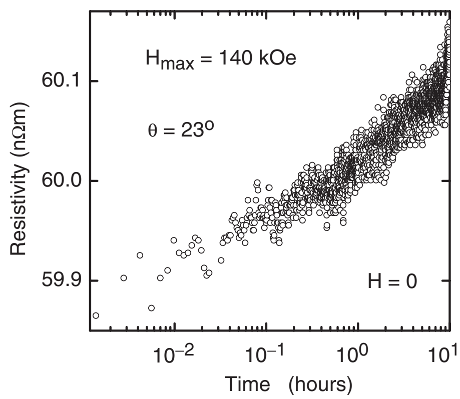

FIG. 6. Time dependence of resistivity at $H=0$. The sample was magnetized with $H_{max}=140$ kOe prior to the measurement.

Fig. 3(b). Here, the field sweep was reversed at $H=0$ to the negative direction. In spite of the absence of the magnetization reversal of the ordinary component, the magnetic order is reduced to merely an intermediate degree, evidenced by the resistivity which is now in between the values corresponding to the maximally and minimally ordered configurations. That is, the ferromagnetic layer undergoes spontaneous disordering due to partial demagnetization.

B. Magnetization

The strong DW pinning and the spontaneous demagnetization are accompanied by a low-temperature anomaly of magnetization. In Fig. 2(b), we show the temperature dependence of the magnetization measured on a SQUID magnetometer. The sample was subjected to a ZFC from room temperature to 2 K. A field of 45 kOe was then turned on. The ZFC and field-cooling (FC) curves were taken in the subsequent up and down temperature sweeps between 2 and 370 K, respectively. Surprisingly, the magnetization decreases for decreasing temperature below 110 K. Below 20 K, the ordinary temperature dependence reemerges. Although the low-temperature magnetization is larger in the FC than in the ZFC, the FC does not qualitatively alter the temperature dependence. Note that the magnetization does not fall to zero in Fig. 2(b) at T_C of MnAs. This feature is nothing but a consequence of the magnetic-field-induced phase transition from the nonmagnetic β phase to the ferromagnetic α phase.[10] In the absence of the external field, the magnetization vanishes at a temperature of about 320 K.

Although the magnetic coupling in bulk MnAs is solely ferromagnetic, an antiferromagnetic coupling is suggested to develop below 110 K in the epitaxial layers, thereby reducing the net magnetization. A ferromagnet surrounded by an antiferromagnetic layer is known to exhibit an exchange-biasing effect.[11] No evidence of the effect was found, however, in the magnetization curve with FC. The absence is not unexpected as the fraction of the antiferromagnetic component is estimated to be merely 1% from the magnitude of the anomalous temperature dependence. Given the threefold-symmetric stress in the MnAs layers imposed by the substrates, spin-glass-like behavior due to frustration on a triangular lattice may become relevant if an antiferromagnetic coupling is introduced into the MnAs/GaAs(111)B system. We point out that the coexistence of a magnetization flip at H_c and an unsaturated hysteretic component was observed in a spin-glass system of CuMn.[12]

IV. ANTIPHASE BOUNDARIES

We attribute the antiferromagnetic coupling to APB. A fairly randomized distribution of zero-field magnetic moments as well as a large value of H_c are typical features originating from APB.[13] When nucleated islands grow on a surface, neighboring terraces may coalesce with unmatched sublattices. In the case of MnAs on GaAs(111)B, the stacking sequence along the growth direction is described as …$ABACA$…, as illustrated in Fig. 1(a). The As sublattice can, therefore, form APB. In the example illustrated in Fig. 1(b), the APB consists of a zigzag chain of As atoms, as indicated by the solid line. The ferromagnetic order in the Mn hexagonal plane is considered to be mediated by the Mn–As–Mn bonds.[14] Therefore, the modified configuration of the As atoms in the vicinity of two neighboring Mn atoms could alter the magnetic coupling, despite the unchanged Mn sublattice. MnAs on GaAs is, hence, unconventional if the APB associated with the sublattice of a nonmagnetic element is indeed responsible for altering the magnetic properties. Identifying the origin of the antiferromagnetic coupling will prove the mechanism of the ferromagnetic coupling in α-MnAs and, in turn, gives insight into the controversial magnetic state in β-MnAs.[4]

We emphasize that MnAs layers grown on GaAs(001) do not exhibit such unusual magnetic properties,[15] i.e., the anomalous properties possess a remarkable dependence on the crystal orientation relationship. For the GaAs(001) substrates, the MnAs layers are $(1\bar{1}00)$ oriented, i.e., the c axis lies in the surface plane. In principle, APB can be present in MnAs layers irrespective of the substrate orientation. As the APB is normal to the c axis for the (001)-oriented substrates,[15] only the APB that extends parallel to the c axis is indicated to produce the anomalous magnetic interaction.[16]

If the magnetic order at the APB is altered to be antiferromagnetic, the adjacent ferromagnetic domains can be exchange decoupled. The decoupling can account for the spontaneous demagnetization when H approaches zero. The magnetization in the decoupled domains is aligned along a local magnetic easy axis at $H=0$, which is not necessarily along the direction of the global magnetization. The change in the temperature dependence of magnetization below 20 K may originate from a competition between the ferromagnetic coupling in MnAs and the antiferromagnetic coupling in APB. A ferromagnetic order is reestablished across APB if the exchange interaction regains the dominance at low temperatures, i.e., the antiferromagnetic order at the APB may be marginal as it will not originate from direct Mn–Mn bonds.

The spin-dependent carrier transfer between exchange-decoupled domains gives rise to a MR when the orientation of their magnetization is misaligned.[17] The scattering at APB yields a broad peak in the MR at H_c, as the decoupled domains flip the magnetization independently at their own char-

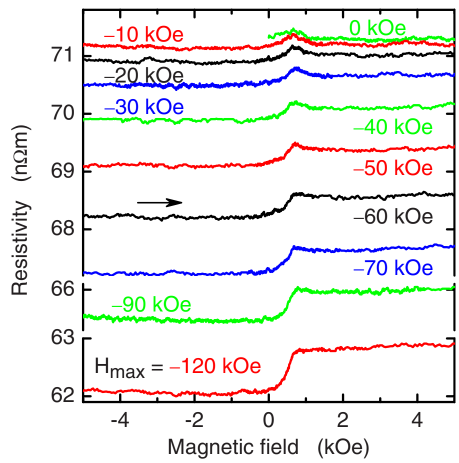

FIG. 7. (Color online) Magnetic-field dependence of resistivity at temperature 0.3 K. The series of curves was taken following a zero-field cooling. The magnetic field was swept to the positive direction, while the maximum field H_{max} having negative polarity was varied from 0 to −120 kOe, as labeled in the panel, in the order displayed from top to bottom.

acteristic fields distributed around H_c. In fact, we find such a MR property when H_{max} is small (Fig. 7). Here, the measurements were carried out in a fashion similar to those in Fig. 4. What is different is that, after setting the field to H_{max} at negative polarity, the field was swept through zero to beyond $+H_c$. With increasing $|H_{max}|$, we find (i) a transformation of the resistivity change at H_c from a broad peak to a steplike increase and (ii) an increase in the magnitude of the change. The former is plausibly due to the diminishing randomness for large $|H_{max}|$, as the random magnetic domains in the initial state are incorporated into a single regular magnetic domain. The latter is a consequence that more regular DWs can be trapped during the DW movement as there are more pinning centers that have discarded the initial DWs. The large difference in the magnitudes of the decrease of ρ_0 and the resistivity jump suggest that the regular DWs are less efficient than the initial DWs in either scattering the conduction carriers or being captured by the pinning centers.

V. CONCLUSIONS AND REMARKS

In conclusion, we have demonstrated that the low-temperature magnetotransport properties in MnAs(0001) layers grown on GaAs(111)B substrates are characterized by a hysteresis that spans the entire range of no less than 140 kOe. The enormously strong DW pinning occurs when the magnetization indicates the emergence of an antiferromagnetic-like order at temperatures lower than about one-third of the Curie temperature. We have proposed a model that attributes these unusual properties to the antiphase boundaries formed during the epitaxial growth of the layers.

Let us finally make several remarks. First, while it is likely that the pinning of DWs at APB leads to the unsaturated magnetization in high fields, the network of misfit dislocations at the MnAs/GaAs heterointerface[3] may also act as the pinning centers. These ordinary pinning centers alone, however, cannot be responsible for the anomalous magnetic properties as they are completely absent when the substrate is (001) oriented.[15] Second, the emergence of the anomalous phenomena depends on subtle differences in the growth conditions. Apart from a possible exception,[18] the temperature dependence of the magnetization in the MnAs/GaAs(111)B system reported by other groups[6,19] did not show the signature of the antiferromagnetic coupling. Nevertheless, considering that a tiny amount of the anomalous magnetic moments causes the large MR effects, the influences would be gigantic if the density of the anomalous moments can be increased. As we have utilized, the extraordinary sensitivity of the MR enables us to pick up subtle magnetic rearrangements that the conventional magnetization characterization fails to detect. The dramatic dependence of the MR phenomena on the substrate orientation can be employed to test the models of the colossal MR effect.

ACKNOWLEDGMENTS

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