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Distribution of type-*B* minority domains in a type-*A* MnAs thin layer on GaAs(001)

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We examine the statistics of the orientation of the magnetic moment in MnAs dots fabricated from an epitaxial film on GaAs(001). Magnetic-force microscopy reveals that the magnetic moment in about 6% of MnAs dots having a diameter of about 60 nm and a thickness of 37 nm be titled from the magnetic easy axis. The amount is consistent with the ratio of the type-*A* and type-*B* crystallographic directions of MnAs estimated from the magnetization curve of the film. The type-*B* minority MnAs is concluded to be distributed in the type-*A* matrix as islands with the lateral dimensions no more than 100 nm. © *2007 American Institute of Physics*.
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In growing hexagonal MnAs layers on the polar GaAs(001) surfaces, the epitaxy mainly takes place with two possible orientation relationships of the layers with respect to the substrates, as illustrated in Fig. 1.[1,2] In type-*A* structure, the MnAs layers are $(1\bar{1}00)$ oriented with their [0001] and $[11\bar{2}0]$ directions being parallel to the $[1\bar{1}0]$ and [110] directions of GaAs, respectively. In type-*B* structure, the $[11\bar{2}0]$ direction of MnAs is rotated in the surface plane by 90°. In this case, the *c* axis of MnAs either lies in plane, i.e., the surface orientation being $(1\bar{1}00)$, type B_0, or is tilted at an angle of about 30° from the surface, exhibiting the surface plane of $(1\bar{1}01)$, type B_1.[2] A preferential epitaxial orientation can be chosen during growth among these types by exploiting the sensitivity of the favorable orientation and azimuthal alignment to the stoichiometry-related surface structure.

The magnetic hard axis of bulk MnAs is along the [0001] direction and the (0001) plane is the magnetic easy plane. As a consequence, the magnetic easy axis in thin MnAs layers on GaAs(001) is oriented to be along the $[11\bar{2}0]$ direction of MnAs regardless of the type. The epitaxial relationships imply that the magnetic easy axes in the type-*A* and type-*B* domains are orthogonal to each other when they coexist on a GaAs substrate. For "spintronic" applications, it is thus important to avoid the coexistence of the two types. The magnetic properties, in addition, are known to differ between the type-*A* and type-*B* MnAs. Panguluri *et al.*[3] evaluated the spin polarization using the point-contact Andreev-reflection spectroscopy. The spin polarization was found to be slightly higher for the type-*A* orientation than for the type-*B* orientation. Chun *et al.*[4] reported an observation of the exchange-biasing effect in thick type-*A* layers, whereas the effect was absent in type-*B* layers. While the type-*A* crystallographic orientation is generally preferred,[2] a mixture of type-*B* minority domains may be inevitable in reality. Under such a circumstance, it is crucial to clarify how the minority domains are distributed in epitaxial layers.

In this paper, we deduce the distribution of the two types of MnAs by analyzing the magnetic properties of nanometer-scale ferromagnetic dots fabricated from an epitaxial layer. We show that, in a type-*A* layer, minority type-*B* domains are present as islands having lateral sizes as large as tens of nanometers.

We infer the planar distribution of the two types of MnAs by taking advantage of the fact that the magnetic moments in type-*A* and type-*B* MnAs are orthogonal to each other. That is, we map out the two-dimensional distribution of the magnetic easy axis in a layer. In order to determine the direction of local magnetic moments, we process the layer

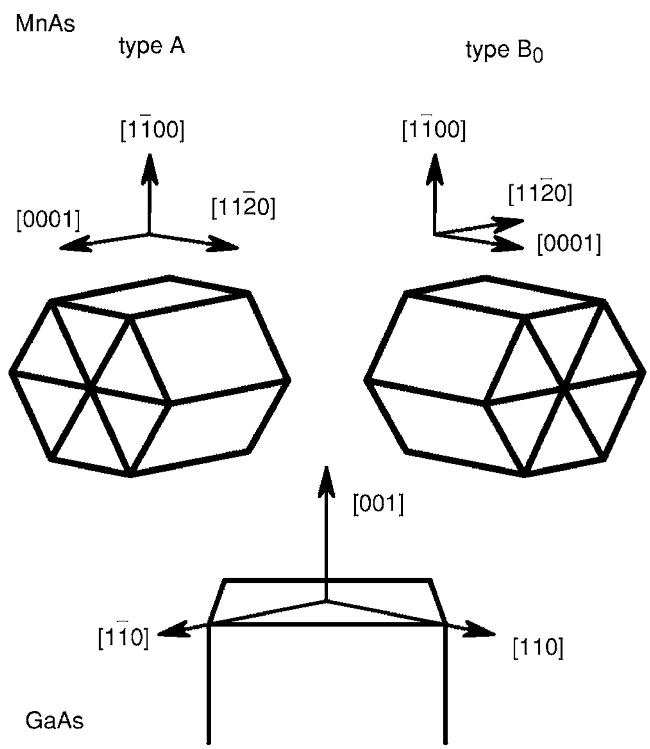

FIG. 1. Crystal orientation relationship for a MnAs layer grown on a GaAs(001) substrate. The *c* axis of hexagonal MnAs is along the $[1\bar{1}0]$ and [110] directions of GaAs for the type-*A* and type-B_0 orientations, respectively.

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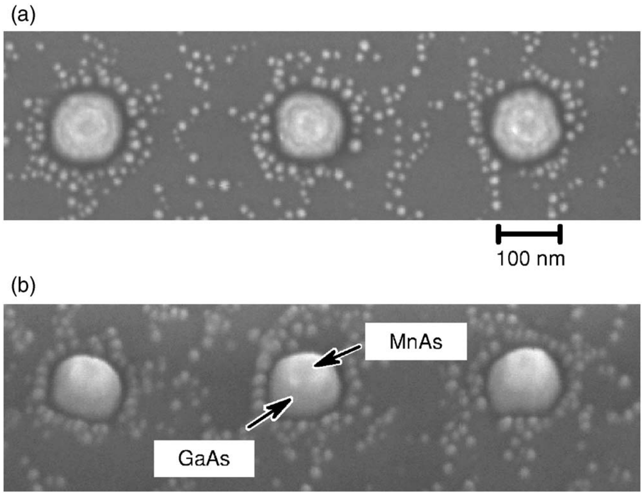

FIG. 2. Scanning-electron micrographs of MnAs dots on a GaAs substrate. Top and glancing-angle views are shown in (a) and (b), respectively. MnAs dots are placed on top of GaAs pillars.

into an array of nanometer-scale dots. Magnetic-force microscopy (MFM) is utilized to identify the direction of the magnetization of an individual dot.[5]

A MnAs layer having a thickness of 37 nm was grown by molecular-beam epitaxy on a GaAs(001) substrate at a growth temperature of 230 °C. The growth was carried out on an As-rich template that developed GaAs(001)-$c(4\times4)$ reconstruction. The As_4/Mn beam equivalent pressure ratio was set to be 250 and the growth rate was 15 nm/h. The MnAs layers grow with the type-*A* epitaxial orientation under these growth conditions.[6] Using electron-beam lithography and Ar ion milling, we fabricated MnAs dots having a diameter of about 60 nm from the epitaxial layer.[7] The dots were assembled in the form of a square array with a period of 350 nm. We show scanning-electron micrographs of the MnAs dots in Fig. 2. Note that the MnAs dots sit on top of GaAs pillars instead of being placed on a flat GaAs surface as the sputtering rate for GaAs is significantly larger than that for MnAs. We have taken magnetic-force micrographs of the MnAs dots at room temperature to determine the magnetic-domain structure in the dots.

We show a MFM image of the MnAs dots at a temperature of 25 °C in Fig. 3(a). The magnetic easy axis of MnAs is arranged to be in the vertical direction in this image. The MFM tip detects magnetic fields normal to the surface. Therefore, an in-plane magnetic moment in the dots gives rise to a combination of bright and dark contrasts in the sections where the stray magnetic fields of the magnetic moment are directed out of the surface and toward the surface, respectively. In the bottom three dots in Fig. 3(a), for instance, the magnetic moments are deduced to be oriented towards the top of the image, as we illustrate by the arrows in the atomic-force micrograph of the corresponding dots in Fig. 3(b).

The upper-right dot in Fig. 3 is invisible in the MFM image. The disappearance is a manifestation that the dot consists of nonmagnetic β-MnAs (the high-temperature phase above the Curie temperature T_C) rather than ferromagnetic α-MnAs (the low-temperature phase below T_C). Bulk MnAs

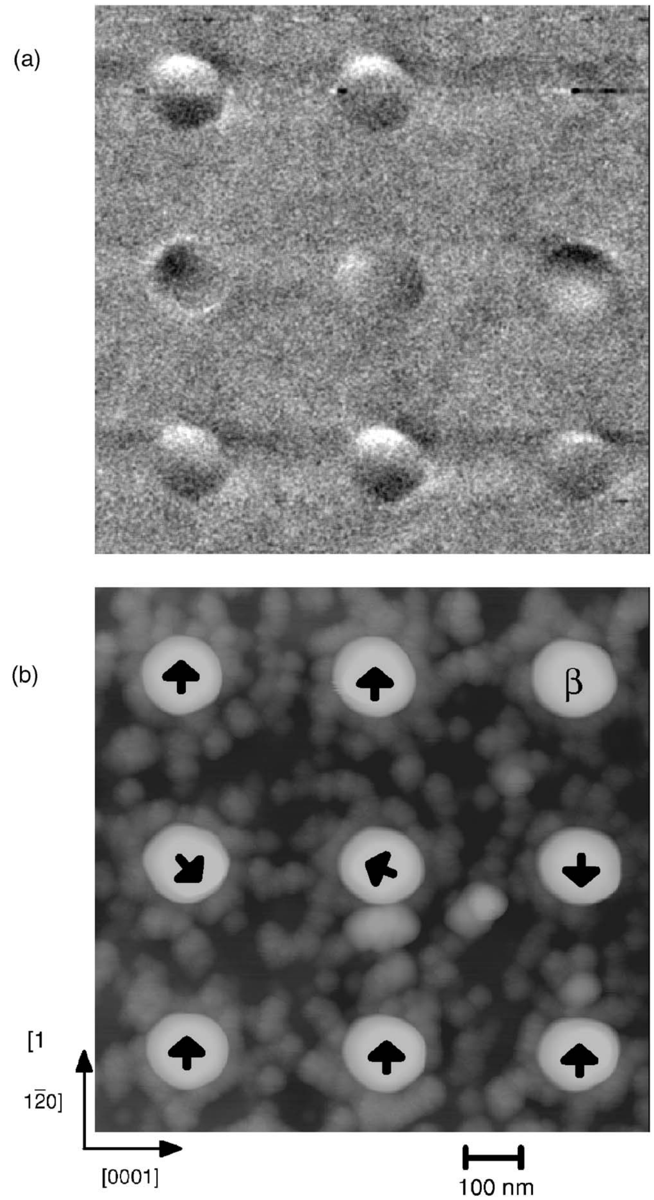

FIG. 3. (a) Magnetic- and (b) atomic-force micrographs of MnAs dots on a GaAs(001) substrate. The arrows in (b) indicate the direction of the magnetic moment in ferromagnetic dots. A nonmagnetic dot consisting of β-MnAs is indicated by "β." The magnetic easy axis of the MnAs layer is along the $[11\bar{2}0]$ direction.

is expected to be in the α phase at room temperature as $T_C \approx 40°$. However, as the phase transition at T_C is first order, the transition takes place only if a potential barrier separating the two phases is surmounted.[5] When nuclei of the stable phase are formed in a domain of the metastable phase through such a course, they grow larger and convert the phase of the entire dot. Due to the nucleation initiation of the first-order phase transition, an individual dot contains either α-MnAs or β-MnAs. However, the two kinds of dots generally coexist in a temperature range around T_C, as demonstrated in Fig. 3(a).[8] We note that the relaxation behavior of the phase transition was investigated in Ref. 9

The middle-left and center dots in Fig. 3(a) exhibit a magnetization which is not parallel to the expected magnetic easy axis. We emphasize that the deviation from the easy axis cannot be ascribed to the shape anisotropy. Due to a strong magnetocrystalline anisotropy, the shape-anisotropy effect is insignificant in MnAs. The magnetization in narrow wires was observed to remain transverse to the wire axis when they were stretched along the magnetic hard axis.[10] The atomic magnetic moments within a MnAs dot are rather strictly directed to be parallel to the magnetic easy axis.[5] The remanent magnetization in dots having a diameter of 100 nm was as large as 80% of the saturation magnetization.[11] That is, the tilting of the surface atomic magnetic moments to suppress the stray magnetic fields amounted at most to a mere 20% modification of the magnetization.

The strong uniaxial magnetocrystalline anisotropy also allows us to ignore the dipole-dipole interaction among the ferromagnetic dots[12] to alter the magnetization direction. The dipole fields are not sufficient to rotate the magnetization.[13] In Ref. 9, an enhancement in the time dependence of the remanent magnetization of an array of dots was observed instead of a decay. The anomalous behavior was attributed to the dipole-dipole interaction. The influence of the interaction was, however, much weaker than to dramatically tilt the magnetization direction to the degree we find in Fig. 3.

We have examined the magnetic properties of 89 MnAs dots using MFM. Eleven dots were invisible in MFM and thus indicated to consist of β-MnAs. Among the rest of 78 ferromagnetic dots, only five (6%) of them exhibited the inclined magnetization orientation. With respect to the 73 ordinary ferromagnetic dots, 40 and 33 dots pointed the magnetization upward and downward, respectively. The roughly the same number of dots for each magnetization direction indicates that the sample was in a demagnetized state. As multidomain structures did not emerge despite the demagnetization,[5] the dots are confirmed to be small enough to be the so-called nanomagnets. The nanomagnets contain only a single magnetic domain as the domain-wall energy exceeds the magnetostatic energy associated with the stray magnetic fields of magnetic domains.[14,15] The complete exclusion of domain walls in the dots assures that the tilted magnetization is unrelated to domain-wall phenomena.

We interpret the tilting as indicative of an unintentional inclusion of type-*B* MnAs in the anomalous dots. We can, furthermore, deduce the distribution of the type-*B* domains from the manner the anomalous dots spread in the array. Except for the two adjacent anomalous dots in Fig. 3(a), the other three anomalous dots were placed in the array separately in the surrounding of ordinary dots. The minority type-*B* MnAs domains thus exist in the type-*A* MnAs layer as islands with a typical size less than the period of the array (=350 nm). With increasing the fraction of type-*B* MnAs in an anomalous dot, the magnetization direction rotates from being along the nominal $[11\bar{2}0]$ direction to being along the nominal [0001] direction. The tilt angle θ thus allows us to determine the ratio of type-*A* and type-*B* MnAs within the dot, assuming that the magnetization is identical in the two types of MnAs, as

$$\tan\theta = f_B/f_A, \tag{1}$$

where f_A and f_B $(=1-f_A)$ are the fractions of the type-*A* and type-*B* MnAs, respectively. None of the MnAs dots exhibited

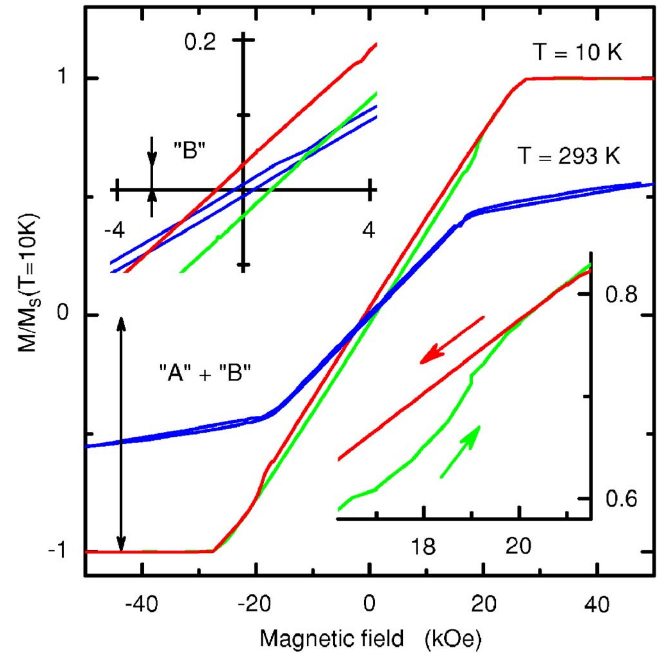

FIG. 4. (Color online) Magnetization M normalized to the saturation magnetization M_s at temperature $T=10$ K of the MnAs epitaxial layer on a GaAs(001) substrate, from which the dots were fabricated. An in-plane magnetic field (maximum strength 50 kOe) is applied along the magnetic easy axis of type-*A* MnAs. The sample temperature is $T=10$ and 293 K. The upper-left inset shows the low-field characteristics with expanded scales. The remanent magnetization is associated with the type-*B* MnAs, whereas both the type-*A* and type-*B* MnAs contribute to the saturation magnetization. The lower-right inset shows the flipping region of the hysteretic component. The arrows indicate the direction of the magnetic-field sweep.

the magnetization in the hard-axis direction, and so all the anomalous dots contained both type-*A* and type-*B* MnAs. The islands of type-*B* MnAs are hence unlikely to be much larger than the diameter of the dots (=60 nm). The lateral dimensions of the minority islands are, therefore, concluded to be less than on the order of 100 nm.

As evidence to support our speculation of a mixture of type-*B* MnAs in the type-*A* layer, we show the magnetization curve of the layer prior to the patterning in Fig. 4. The magnetization was measured using a superconducting-quantum-interference-device magnetometer. Here, an in-plane magnetic field was applied along the hard axis of type-*A* MnAs. In addition to the hard-axis behavior originating from type-*A* MnAs, a hysteresis gap is found to open for magnetic fields smaller than 20 kOe. (The low-field characteristics are shown with expanded scales in the upper-left inset of Fig. 4.) We attribute the hysteresis loop to the type-*B* MnAs contained in the layer, for which the magnetic field was applied along the easy axis. The magnitude of the hysteresis gap was almost independent of the magnetic field. We, therefore, estimate the fraction of the type-*B* MnAs to be about 3.5% from the magnitudes of the remanent magnetization (due to type-*B* MnAs) and the saturation magnetization (due to the whole MnAs). (Due to the field independence of the magnitude of the hysteresis gap, the suppression of the remanent magnetization from the saturation magnetization originating from the type-*B* MnAs can be ignored.) The tilt angle of the magnetic moment in the five anomalous dots was about 40°,

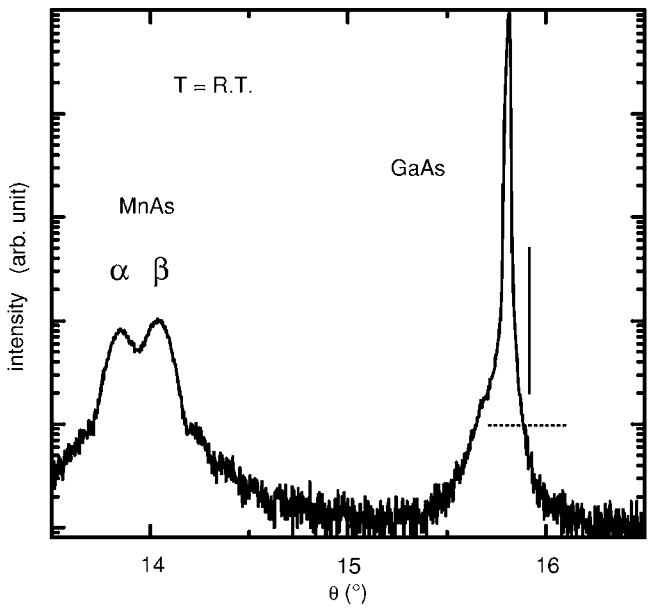

FIG. 5. X-ray diffraction curve of the unpatterned MnAs/GaAs structure at room temperature. The three peaks from left to right are associated with the $[1\bar{1}00]$ reflection of α-MnAs, [020] reflection of β-MnAs, and [002] reflection of GaAs. The peak position expected for the $[1\bar{1}01]$ reflection is indicated by the vertical bar. A prediction of the peak amplitude when the fraction of type-B_1 MnAs is 3.5% is shown by the horizontal dotted line.

42°, 48°, 53°, and 69°. Roughly a half of the MnAs content is thus suggested to be type B in about 6% of the dots. The fraction of the type-B MnAs deduced using the magnetization curve and the MFM images is, therefore, in quantitative agreement.

In Fig. 5, we show the x-ray diffraction curve of the MnAs layer. The three peaks are associated from left to right with the $[1\bar{1}00]$ reflection of α-MnAs, [020] reflection of β-MnAs, and [002] reflection of GaAs. The $[1\bar{1}01]$ reflection of α-MnAs is expected to give rise to a peak at the position of the vertical bar.[16] The horizontal dotted line indicates the peak amplitude anticipated from the magnitude of the $[1\bar{1}00]$ reflection of α-MnAs when the fraction of type-B_1 MnAs is 3.5%. Here, we have taken into account the difference in the structure factors of the $[1\bar{1}00]$ and $[1\bar{1}01]$ reflections. The absence of the peak corresponding to the $[1\bar{1}01]$ reflection indicates that the minority MnAs is of the type B_0 in our epitaxial layer.

Let us finally point out signatures of a magnetic interaction between the type-A and type-B regions. Despite the similar coercivity of type-A and type-B MnAs,[1] the coercive field for the hysteretic component in Fig. 4 is nearly as large as the magnetic field required to orient the magnetization along the magnetic hard axis. (The coercive field in our type-A layers is $\sim$0.8 kOe.[11]) In addition, the transition between the two branches of the hysteresis takes place gradually; see the lower-right inset of Fig. 4. These suggest a strong magnetic coupling between the islands of type-B MnAs and the matrix of type-A MnAs. The magnetic coupling may be utilized to achieve magnetologic operations.[17,18] Temperature dependence of these magnetic properties was weak. Around room temperature, however, the magnetic-field range for the hysteretic behavior expanded beyond 50 kOe. The coexistence of the α and β phases of MnAs in the epitaxial layer[2] is suggested to have a profound impact on the magnetic interaction.

In conclusion, we have investigated the distribution of type-B MnAs which was unintentionally contained in an epitaxial type-A MnAs layer on GaAs(001). The orientation of the magnetic moments in a square array of nanometer-scale dots prepared from the epitaxial layer was determined using magnetic-force microscopy. The deviation of the magnetic moment away from the nominal magnetic easy axis was employed to deduce the amount and distribution of type-B MnAs. We conclude that the minority domains are present as isolated islands whose lateral dimensions can be as large as 10–100 nm.

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