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Magnetic out-of-plane component in MnAs/GaAs(001)

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From highly sensitive superconducting quantum interference device magnetometry and magnetic force microscopy, we deduce a small out-of-plane magnetization component of MnAs/GaAs(001) films. Its temperature dependence is substantially different from the dominating in-plane magnetization, particularly as it is still detectable above the phase transition temperature of MnAs films. Our measurements indicate that the out-of-plane component is due to small isolated magnetic "grains" within the film. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1615682]

Ferromagnetic manganese arsenide films on GaAs are promising candidates for future spin-injection devices.[1] Nowadays high-quality epitaxial MnAs($\bar{1}100$) films are routinely grown by molecular beam epitaxy (MBE) on the two technologically leading semiconductor substrates, Si(001) and GaAs(001).[2–5] In the bulk, MnAs loses its long-range magnetic order abruptly at about 40 °C, when the ferromagnetic α-phase transforms into the paramagnetic orthorhombic β-phase by a first-order transition.[6,7] In thin films of MnAs($\bar{1}100$) on GaAs(001), on the other hand, the transition from α- to β-phase does not proceed abruptly. Instead, there is a broad temperature region from 10–40 °C, where both phases coexist[8] forming a self-organized stripe pattern of the two phases.[9,10] Figure 1(a) illustrates the striped pattern at 15 °C using magnetic force microscopy (MFM). The ferromagnetic α-phase can be identified by the zigzag-shaped magnetic contrast due to its domain pattern in the demagnetized state. At 41 °C, the striped pattern has disappeared [Fig. 1(b)], however, some residual magnetic contrast is locally detectable in the β-phase (marked by arrows). A detailed temperature dependent MFM analysis revealed that these dots serve as pinning centers for the formation of the ferromagnetic α-phase upon cooling.[11]

To study the origin of the magnetic contrast in more detail, we performed highly sensitive measurements by a superconducting quantum interference device (SQUID) magnetometer of the magnetic properties of MnAs/GaAs(001) in-plane along the *a* axis and out of plane. We recorded magnetic hysteresis loops evolving as a function of the temperature across the phase coexistence range. From the in-plane measurements, we can confirm the loss of long-range order at about 50 °C whereas the out-of-plane measurements reveal a small hysteresis loop which is present in the temperature range of 0–55 °C, thus indicating that a small fraction of the MnAs film exhibits a finite out-of-plane component of the magnetization.

As substrates, commercial epiready GaAs(001) wafers were coated by standard solid-source MBE (Ref. 12) with a 100-nm-thick GaAs(001) buffer layer after oxide removal. MnAs was deposited at a substrate temperature of 250 °C, at a As_4/Mn beam equivalent pressure ratio of 250, and at a rate of 20 nm/h. The epitaxial orientation with respect to the substrate is MnAs($\bar{1}100$)∥GaAs(001) and MnAs[0001]∥GaAs[$1\bar{1}0$], see Refs. 4 and 13. The easy axis of magnetization is MnAs[$11\bar{2}0$], i.e., the in-plane *a* axis, while the magnetically hardest direction is along the in-plane MnAs[0001] (*c*-axis).[5,14] This leads to the unusual situation that the out-of-plane anisotropy field ($\sim$1.0 T) is smaller than the in-plane one ($\sim$2.0 T).[5] The magnetic measurements were carried out using a commercial SQUID magnetometer with the external field applied in the film plane and out of plane. The temperature was varied from 0 °C to 60 °C and backward to exclude thermal hysteresis effects. Each hysteresis loop was measured at stabilized temperatures ($<\pm 0.05$ °C).

Figure 2 shows hysteresis loops of a 60 nm MnAs/GaAs(001) film at 0 °C and 50 °C recorded in plane along the *a* axis [Fig. 2(a)] and out of plane [Fig. 2(b)]. The in-plane measurements reveal an almost perfectly squarelike loop indicating that this direction is indeed the easy axis. Since the remanence is not reduced, the formation of domains can be excluded and the saturation value of the magnetization of 0.65 ± 0.02 MA/m is in good agreement with the magnetization of 0.67 MA/m reported for bulk α-MnAs at 10 °C.[15] Compared to 10 °C, the hysteresis at 50 °C is strongly reduced; note, that the data are enlarged by a factor of 50. Obviously, the system loses its long-range order and appears to be completely transformed into the paramagnetic

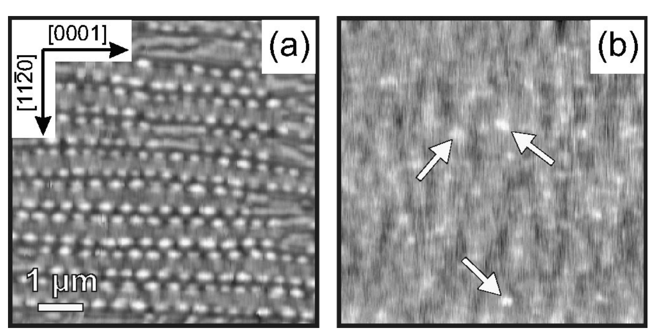

FIG. 1. MFM images of a 180 nm MnAs/GaAs(001) film (see Ref. 13) at (a) 15 °C and (b) 41 °C. (a) shows the coexistence of the ferromagnetic α- and the paramagnetic β-phase; the α-phase is demagnetized thus showing a zigzag-shaped domain pattern of the α-stripes. In image (b), some residual contrast of small isolated dots is visible (arrows).

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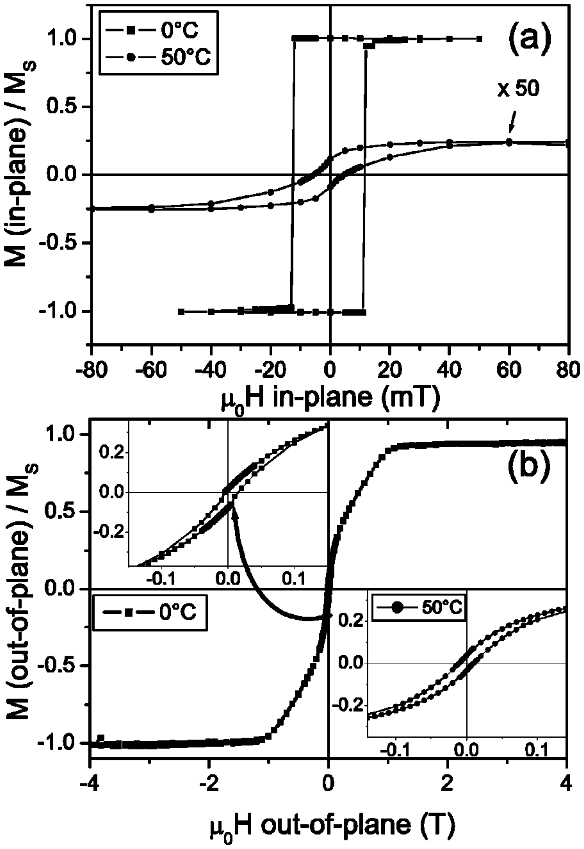

FIG. 2. Magnetic hysteresis measurements of a 60 nm MnAs/GaAs(001) film at 0 °C and 50 °C. The external field is applied (a) in the film plane along the *a* axis and (b) out-of-plane. Note, that the in-plane hysteresis at 50 °C is normalized to the saturation magnetization at $T=0$ °C ($M_S=0.65$ MA/m) and magnified by a factor of 50. The two insets of (b) enlarge the low-field range revealing clear out-of-plane hysteretic behavior.

β-phase. The whole temperature dependence of the in-plane remanent magnetization throughout the phase coexistence range from 0 °C to 60 °C is summarized in Fig. 3 (squares).

Figure 2(b) shows a hysteresis loop of the same sample traced along the out-of-plane direction from -4 to $+4$ T. The diamagnetic contribution of the substrate and the sample holder derived from the high-field data is subtracted, the sample holder alone showed pure diamagnetic behavior. In accordance with Ref. 5, we find hard-axis behavior with an anisotropy field of 1 T, containing both the shape and the crystalline contribution. The demagnetizing field (shape anisotropy) can be evaluated from the saturation magnetization yielding $\sim$0.5 T, the out-of-plane crystalline anisotropy field is, therefore, $\sim$0.5 T. The upper inset of Fig. 2 shows a magnification of the low-field region at 0 °C exhibiting both clear remanence and coercivity, thus revealing a small out-of-plane component of the magnetization. We want to point out that the loop is not squarelike indicating either that the out-of-plane direction is not the easy axis of the detected component or it originates from weakly coupled "grains" similar to the behavior of polycrystalline materials. Assuming that the out-of-plane contribution is due to regions which possess the same magnetization as the MnAs film, we derive a volume fraction of 2%–3% from the SQUID measurement (which essentially measures the product of magnetization and volume). We remark, that a small hysteresis can be recognized in previous out-of-plane magnetic measurements,[3,5,16] however, it was not addressed in these studies. To corroborate our findings, we performed analogous measurements at higher temperatures, e.g., at 50 °C shown in the lower inset of Fig. 2(b). Surprisingly, nearly all of the same hysteretic behavior as at 0 °C with essentially the same remanence and coercivity is found. This observation—contrary to older work, where no temperature dependent measurements have been performed out of plane—excludes that the signal at 0 °C is due to the misalignment of the sample with respect to the external field. In that case, the out-of-plane signal would exhibit the same temperature dependence as the in-plane component and should be drastically reduced compared to 0 °C. Therefore, we can also conclude that the out-of-plane signal is due to a magnetization aligned along or close to the surface normal. For similar reasons, the out-of-plane component can not be attributed to domain-wall formation or tilted magnetization contributions at furrows as discussed in Ref. 5.

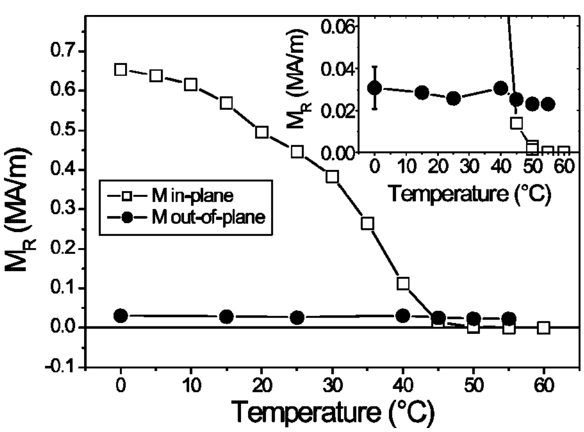

FIG. 3. Remanent magnetization, in and out of plane, derived from hysteresis measurements in the temperature range of 0–60 °C. The inset shows that the out-of-plane component is nonzero throughout the whole temperature range and it decreases only slightly while the in-plane component vanishes.

Figure 3 summarizes our SQUID measurements showing the in-plane (squares) and out-of-plane (circles) component of the remanent magnetization of 60 nm MnAs/GaAs(001). Whereas the in-plane component—which by far dominates at lower temperatures—gradually decreases until it vanishes around 50 °C, the out-of-plane component shows only a weak temperature dependence and is present even when the in-plane magnetization has already disappeared. The existence of an out-of-plane component of the magnetization has several implications. (i) The out-of-plane component does not follow the temperature dependence of the in-plane magnetization, therefore, it is decoupled from the structural and magnetic phase transistion. This points toward isolated regions within the film which are consistent with the MFM findings in Fig. 1(b). (ii) For being magnetized out-of-plane, these regions must have considerable crystalline anisotropy, otherwise, the demagnetizing field and the coupling to the remainder of the film would push their magnetization back to the film plane at lower temperatures. (iii) Since we detect an out-of-plane ferromagnetic signal above 55 °C (see inset of Fig. 3), which is significantly above the ordering temperature of bulk MnAs (40 °C), a structural configuration with a higher transition temperature compared to the bulk has to be present. This finding may turn out to be important for technical applications because, for room-temperature devices, the bulk transition temperature is quite low. (iv) Careful hard-axis magnetic measurements can serve as a highly sensitive probe for testing the structural homogeneity of MnAs films.

To unambiguously clarify the structural origin of the out-of-plane magnetiztion component further, careful investigations are necessary. From our magnetic measurements, we can only derive indirect conclusions. The finite crystalline anisotropy of the observed component requires crystallinity of these regions. Their easy axis should be close to out of plane and the rounded shape of the out-of-plane loop suggests that these regions are well separated and weakly coupled. The out-of-plane magnetization seems to be linked to the granular magnetic contrast recorded by MFM above the transition temperature. Recently, Iikawa *et al.*[17] observed a MnAs component in MnAs/GaAs(001) which is rotated by 30° along the *c* axis, thus exhibiting an *a* axis perpendicular to the substrate plane. It is not clear, however, why this structurally equivalent MnAs fraction should have a higher transition temperature. We therefore actually favor Mn-rich inclusions with differing magnetic properties as a possible explanation, which were found recently at the MnAs/GaAs interface.[18]

In conclusion, we presented experimental evidence for an out-of-plane component of the magnetization of MnAs films on GaAs(001) by temperature dependent SQUID measurements. This component seems to be linked with small dots of residual magnetic contrast in MFM images at elevated temperatures consistent with the observation of a rounded hysteresis loop. Magnetic separation of these grains is supported by their different temperature dependent magnetic behavior with respect to the dominating in-plane magnetization of MnAs/GaAs(001).

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