Competing magnetic interactions in MnAs studied via thin film domain pattern analysis

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Manganese arsenide is one of the few ferromagnetic metals that can be grown on semiconductor substrates as a thin film with high structural perfection. The coupled magnetic and structural phase transition around 40 °C leads to a variety of different phenomena such as the self-organized stripe formation on GaAs(001) substrates or the anisotropic lattice shrinkage. By investigating the domain pattern in the phase coexistence region we provide experimental evidence that the magnetic order is due to competing ferromagnetic double-exchange and antiferromagnetic direct exchange interactions. This scenario corroborates recent theoretical calculations and may explain the frequently observed angle of 38° in the domain pattern of epitaxial MnAs films.

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Manganese arsenide is a promising ferromagnetic material for spintronic applications based on spin injection[1] and for magnetologic devices.[2] High quality epitaxial films can be grown on semiconductors like GaAs(001),[3] GaAs(111),[4] GaAs(113),[5] and Si(001).[6] The ferromagnetic phase is metallic and shows a hexagonal NiAs($B8_1$) structure with alternating hexagonal planes of Mn and As atoms (α phase). At about 40 °C it transforms into the nonmagnetic, orthorhombic β phase (B31).[7–9] In the course of the phase transition, the magnetic order discontinuously breaks down and the lattice spacing within the hexagonal plane (a axis) shrinks abruptly, while the spacing along the c axis remains unchanged.[10] The contraction in volume amounts to ~2%. In MnAs films on GaAs(001), the phase transition no longer proceeds abruptly due to the epitaxial constraints. Instead, over a broad temperature range of 10–40 °C, a strain-stabilized two-dimensional pattern of coexisting α- and β-MnAs stripes along the c axis forms.[11,12] The crystallographic relationship of MnAs grown on GaAs(001) substrates is sketched in Fig. 1. Note that all Mn atoms along the a and the c axis are lying in the same plane whereas the As atoms are above that plane.

The magnetic properties of bulk MnAs have been subject to extensive experimental and theoretical studies. Among these, a phenomenological model for the coupled structural and magnetic α/β-phase transition was proposed that included three different types of magnetic interactions:[13] (i) a direct exchange interaction between the neighboring Mn atoms along the hexagonal c axis which favors ferromagnetic order, (ii) a Mn-As-Mn double exchange mechanism which is of ferromagnetic character as well, and (iii) an antiferromagnetic direct exchange between neighboring Mn atoms along the a axis. This model accounts for the fact that each Mn atom in MnAs has two different types of Mn neighbors: two along the c axis which are next neighbors (0.286 nm), and six within the hexagonal plane where the spacing is 0.37 nm. Recently, density functional theory (DFT) calculations revealed that in the β phase, the Mn planes are antiferromagnetically ordered but there exists no magnetic correlation along the c axis[14]—which would account for the fact that no long-range antiferromagnetic order has been found by neutron scattering experiments.[15] Measurements of the stress and the magnetostriction of epitaxially constrained MnAs-on-GaAs(001) during the phase transition suggest that the Mn-As-Mn interaction governs the ferromagnetic order of MnAs while the magnetic interactions along the c axis play at most a minor role.[16,17]

It is also noteworthy that β-MnAs was already assumed to be antiferromagnetic based on measurements of the anomalous inverse susceptibility. It becomes Curie-Weiss-like only in the γ phase, i.e., above 130 °C, exhibiting a characteristic kink at this phase transition.[7] Moreover, the extrapolation of the spontaneous magnetization of α-MnAs would lead to a Curie temperature of around 130 °C, where the orthorhombic β-MnAs transforms into the hexagonal γ-MnAs, which is without any doubt paramagnetic.[7]

Here we show that the complex domain pattern of MnAs/GaAs(001) in the striped $\alpha+\beta$ coexistence region may provide valuable information about the magnetic interactions in MnAs. From micromagnetic domain imaging of a number of samples with different film thicknesses[18,19] and at different temperatures[20] it is known that domain walls are not only directed perpendicular or parallel to the stripes but do occur under intermediate angles as well. The commonly observed angle of ~38° will be discussed assuming competing

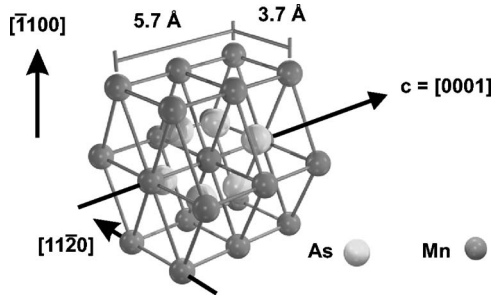

FIG. 1. NiAs-type crystal structure of α-MnAs.

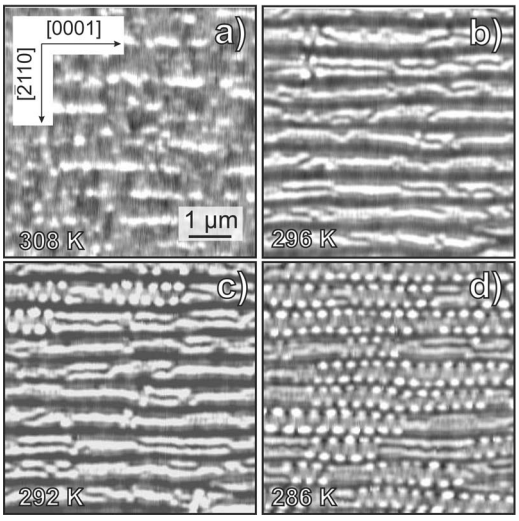

FIG. 2. MFM images of a 180-nm-thick MnAs layer on GaAs(001) in the $\alpha+\beta$ phase coexistence region at different temperatures. The scan size is 7×7 μm^2.

anisotropic magnetic interactions, especially an intrinsic antiferromagnetic coupling along the a axis in MnAs that is overcome by the Mn-As-Mn interaction in the ferromagnetic α phase.

The MnAs-on-GaAs(001) samples were grown by standard molecular beam epitaxy yielding MnAs$(\bar{1}100)\|$GaAs(001) and MnAs$[0001]$(c-axis)$\|$GaAs$[1\bar{1}0]$.[21] The shape anisotropy leads to a situation where the in-plane a axis and the c axis are the magnetic easy and hardest direction, respectively. The magnetic domains were imaged using temperature-dependent magnetic force microscopy (MFM). The MFM is sensitive to the component of the magnetic stray field which is oriented parallel to the tip axis. The ferromagnetic α stripes exhibit a large variety of magnetic domains, and some have been discussed in recent publications.[22–24] Figure 2 shows the evolving domain pattern while cooling down a 180 nm thick MnAs/GaAs(001) film from the pure β phase (at 323 K) in zero field. Close to the pure β phase, the α stripes are not yet contiguous (a). The nominal temperature of the MFM heat stage is 308 K. At around 296 K (b) the magnetic domains exhibit walls parallel to the stripes, i.e., the individual stripes have domains across their width. Another interesting domain geometry is also visible: the onset of domain walls which are not directed parallel or perpendicular to the stripes. At 292 K (c) the domain pattern typically shows only very few inclined domain walls and zig-zag-shaped domain patterns start to show up (the stripe forms domains along its length but not its width anymore) and finally they dominate the pattern at 286 K (d). These other types of domain patterns are discussed in greater detail in Ref. 22. Obviously, the occurrence of the tilted domain walls is restricted to a limited temperature interval below the α-β phase transition temperature. Furthermore, the occurrence of tilted domain walls is restricted to an interval of film thicknesses where the lower bound is given by the onset of dipolar coupling between the stripes and the upper bound by the formation of closure domains in depth. As the α-β-stripe period is a linear function

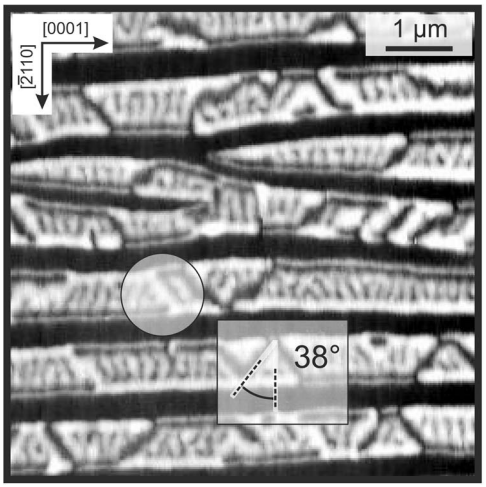

FIG. 3. MFM image of a 215-nm-thick MnAs layer on GaAs(001) in the $\alpha+\beta$ phase coexistence region at 293 K. The scan size is 7×7 μm^2.

of film thickness, the dipolar forces between the α stripes start to dominate the magnetic interaction for film thicknesses around 60 nm. This has recently been investigated by ferromagnetic resonance.[25] For example, in Fig. 2(d) a weak inter-stripe correlation of the zig-zag-shaped domain pattern is already visible. To illustrate the upper limit of the film thickness, Fig. 3 shows a 215-nm-thick MnAs film at 293 K where a large number of inclined domain wall is visible which coexist with E-shaped or comb-like domain patterns. These domain patterns indicate that the individual stripes start to form a domain pattern in depth.[24] In case of even thicker MnAs films, i.e., a larger α-β-stripe period and thus wider ferromagnetic stripes, the stripe minimizes its magnetic energy by forming zig-zag-shaped domain walls at angle of 45°.[26]

A closer inspection of a large variety of films of different thicknesses from 60 to 220 nm at different temperatures close to the phase transition reveals that a significant number (e.g., 0.3 per μm^2 at 36.5 °C for a 120-nm-thick film) of these tilted domain boundaries are directed under a fixed angle of $\sim$38(1)° to the a axis (the stripe width). This typical angle is indicated in Fig. 3 depicting a 215-nm-thick MnAs/GaAs(001) film at 293 K. In some cases, deviations from the 38° tilt angle are due to closure domains at the stripe edges, i.e., a combination of domain walls along the c axis and 38° domain walls (cf. circle in Fig. 3). Having investigated a large number of MnAs films that are all showing inclined 38° domain walls, we can exclude that the observed angle is neither due to the special stripe geometry nor due to the formation of closure domains. In addition we want to point out that 38° domain walls are also visible in MnAs films grown on GaAs(113)A as revealed by MFM,[18] however, this special geometry has not been discussed in detail so far.

The characteristic geometry of the inclined domain wall angle may provide a deeper insight into the different exchange mechanisms responsible for the ferromagnetic order of MnAs. Following the early phenomenological work,[13] we have to account for three different types of interaction for

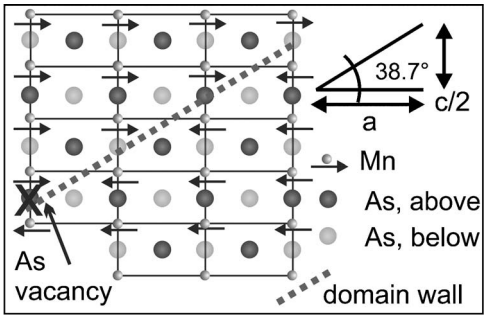

FIG. 4. Sketch of the MnAs film plane and the nucleation of the domain wall (see text). As atoms are situated either above or below the Mn plane. An As vacancy can nucleate a domain wall running along the Mn-As-Mn bond direction which is oriented under 38.7°. Note that the sketch only illustrates one quadrant, however, equivalent scenarios hold for all four quadrants.

which we assume different temperature dependencies. The following discussion neglects the direct exchange interaction along the *c*-axis since measurements of stress[16] and magnetostriction[17] revealed that this interaction plays at most a minor role for the magnetic ordering. This is, however, surprising by itself since the Mn atoms along the *c* axis are next neighbors. We assume two competing magnetic interactions: (i) a direct antiferromagnetic (AFM) coupling of the moments of neighboring Mn atoms along the *a* axis, which is in agreement with recent theoretical calculations,[14] and (ii) the Mn-As-Mn double exchange mechanism.[16] Obviously, in the α phase, (ii) is dominating over (i) since MnAs is ferromagnetic. Further, we assume a stronger temperature dependency for (ii) compared to (i), which is reasonable based on the two different underlying mechanisms (direct versus indirect exchange). Close to the phase transition, the indirect ferromagnetic exchange gets weaker than the direct antiferromagnetic exchange so that the AFM interaction between the Mn atoms along the *a* axis dominates. The fact that the lattice shrinks only along the *a* axis at the phase transition was held as an indication that the magnetic moments of the Mn atoms along the *a* axis align mostly antiparallel so that the AFM interaction along the *a* axis is then dominant. This lattice shrinkage due to the change from parallel to mostly antiparallel alignment of the Mn atoms was termed "exchange striction" in Ref. 13.

According to the above discussion we assume that an antiparallel alignment of the magnetic moments of the Mn atoms along the *a* axis is favorable and thus an antiparallel alignment of the magnetic moments across the stripe width (*a* axis). This is in agreement with the recent calculations showing that β-MnAs energetically favors an antiferromagnetic ordering within the hexagonal Mn plane.[14] However, if the moments of the Mn atoms along the *a* axis are aligned antiparallel, the Mn-As-Mn interaction forces the neighboring Mn moments in the *subsequent* hexagonal plane into a parallel alignment (see Fig. 4). If the competing magnetic interactions are of comparable strength, which is the case close to the phase transition, a domain wall may be nucleated by an As vacancy as illustrated in Fig. 4. The vacancy locally enables the AFM interaction to turn the magnetic moments of the two bottom-left neighboring Mn moments along the *a* axis antiparallel to each other. The other As atoms then orient all magnetic moments of the Mn atoms on the right hand side of the vacancy parallel, thus forming a magnetic domain wall. Such a domain wall is then aligned along the Mn-As-Mn bond because of the dominant Mn-As-Mn exchange interaction. Thus, it would run from one Mn atom in a given hexagonal plane to a Mn atom in the subsequent plane which is shifted by one *a*-lattice length as sketched in Fig. 4 by the dashed line. Taking the lattice parameters *a* and $c/2$ one can calculate that such a domain wall should be oriented under an angle of 38.7°, which is the Mn-As-Mn bond angle. Recently, the observed inclined domain wall angle has been seen in micromagnetic simulations. Using a phenomenologically motivated exchange stiffness tensor, tilted domain wall angles were found in the remanent state.[26] Note that the above scenario could be the underlying physical reason for the introduction of an anisotropic exchange tensor.

In order to prove that the Mn planes in MnAs are indeed antiferromagnetically coupled while there is no correlation along the *c* axis, further experiments are needed. The induced magnetic moment at the As site of $-0.23\mu_B$ (antiparallel to the one of Mn), measured by neutron scattering at 293 K,[27] supports that a Mn-As-Mn exchange mechanism plays a dominant role in the magnetic ordering. However, it would be desirable to figure out whether the induced moment changes significantly by going from the α to the β phase. To directly verify the antiparallel character of the Mn-Mn bond along the *a* axis it may be a viable way to perform angular- and temperature-dependent magnetic extended x-ray absorption fine structure (MEXAFS) measurements, which measure the spin-dependent local pair correlation function site-specifically. Alternatively, measurements of the x-ray magnetic linear dichroism (XMLD) on single crystal MnAs samples would demonstrate if there is a magnetically ordered axis along the *a* axis in the β phase. In that context it is worth noting that the early neutron diffraction measurements which ruled out long range antiferromagnetic ordering in the β phase were performed on polycrystalline MnAs.[15]

In conclusion, the tilted domain boundaries of MnAs/GaAs(001) observed with MFM are frequently oriented under an angle of 38° with respect to the *a* axis, i.e., the stripe width. This type of domain wall is seen in films of different thicknesses. These findings are well explained by the phenomenological model of Ref. 13, assuming an AFM interaction between Mn atoms along the *a* axis which is overruled by a ferromagnetic Mn-As-Mn double exchange mechanism. They corroborate recent DFT calculations showing that β-MnAs is likely to be an uncorrelated antiferromagnet.[14] The shrinkage of the lattice along the *a* axis during the phase transition accordingly is well explained by "exchange striction."

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