Ferromagnetic resonance studies of (Ga,Mn)As with MnAs clusters

Th. Hartmann[a,*], M. Lampalzer[a], P.J. Klar[a], W. Stolz[a], W. Heimbrodt[a], H.-A. Krug von Nidda[b], A. Loidl[b], L. Svistov[c]

[a] *Department of Physics and Material Sciences Center, Philipps-University of Marburg, Renthof 5, 35032 Marburg, Germany*
[b] *Experimentalphysik V, EKM, Universität Augsburg, D-86135 Augsburg, Germany*
[c] *Kapitza-Institute for Physical Problems, 117334 Moscow, Russia*

Spin electronics and spin optoelectronics are key technologies of the future. They will provide new spin-dependent magneto-optical and magneto-electrical devices [1]. Possible candidates, which may serve as a spin aligner or spin injector material and are compatible with III–V semiconductor technology, are the ferromagnetic phases of the (Ga,Mn)As system. The $Ga_{1-x}Mn_xAs$ alloy is p-type and exhibits ferromagnetism. However, as the spin dephasing times for holes are much faster than for electrons and the observed Curie temperatures $T_C < 150$ K, the alloy is not suitable for room-temperature spin applications. Another phase of this material system is ferromagnetic MnAs clusters embedded in a paramagnetic $Ga_{1-x}Mn_xAs$ host matrix. This hybrid system allows n-type doping of the matrix and, in addition, exhibits ferromagnetism above room temperature due to the MnAs clusters. Therefore, it is of interest to investigate the magnetic behaviour of the clusters themselves and their interaction with the electrons of the host matrix. Here, we present systematic angular and temperature-dependent electron-spin-resonance (ESR) studies of $Ga_{1-x}Mn_xAs$ with MnAs clusters grown in situ by metal-organic vapour-phase epitaxy (MOVPE). De Boeck et al. [2] have reported previously on the formation of ferromagnetic MnAs nanoclusters (5–30 nm in diameter) due to post-growth

* Corresponding author. Tel.: +49-6421-2822118; fax: +49-6421-2827036.
E-mail address: thorsten.hartmann@physik.uni-marburg.de (Th. Hartmann).

thermal annealing of $Ga_{1-x}Mn_xAs$ alloys grown by low-temperature molecular beam epitaxy. Recently, such clusters have attracted much attention (e.g. Refs. [3–8]).

(Ga,Mn)As samples with and without MnAs clusters can be obtained by MOVPE [9–11]. In what follows, we focus on two 1 μm thick $Ga_{1-x}Mn_xAs$ samples with MnAs clusters grown on (0 0 1) and (1 1 1) GaAs substrates, respectively, at about 500°C and high Mn-fluxes. Under these conditions, MnAs clusters are formed in situ within a $Ga_{1-x}Mn_xAs$ host with $x \approx 0.1\%$. They are situated close to the surface and their size ranges between 50 and 200 nm. Despite the different crystal structures, the clusters are incorporated nearly dislocation-free into the $Ga_{1-x}Mn_xAs$ matrix [10]. ESR measurements were performed at X-band frequencies using a Bruker ELEXSYS E500 CW spectrometer equipped with a continuous-flow N_2 cryostat.

Ferromagnetic resonances are observed in the ESR spectra of both cluster samples even above room temperature. Due to the hexagonal symmetry of MnAs, the ESR signals show a strong angular dependence. Figs. 1a and 2a show a typical series of ESR spectra obtained at $T = 200$ K for the (1 1 1) and (0 0 1) samples, respectively. The samples were rotated with respect to H_{ext} as follows: the axis of rotation (perpendicular to H_{ext}) was chosen perpendicular to the GaAs [1 1 1] direction for the (1 1 1) sample and parallel to the GaAs [1 1 0] direction for the (0 0 1) sample. The magnetic resonance fields H_{Res} (symbols in Figs. 1b and 2b) can be derived by fitting the spectra with the first derivatives of asymmetric Lorentzian line shapes [12]. In the fit of the angular-dependent spectra of the (0 0 1) sample, we have only accounted for the strongest resonance. Two additional weaker resonances are also observed as discussed below. Assuming that the clusters are spherical uniaxial ferromagnets, the angular dependence of H_{Res} can be described by [13]

$$\frac{\omega_0^2}{\gamma^2} = [H_{Res}\cos(\vartheta_0 - \vartheta_H) + 2H_A\cos(2\vartheta_0)]H_{Res}\frac{\sin\vartheta_H}{\sin\vartheta_0}, \quad (1)$$

$$H_A\sin(2\vartheta_0) = H_{Res}\sin[2(\vartheta_H - \vartheta_0)], \quad (2)$$

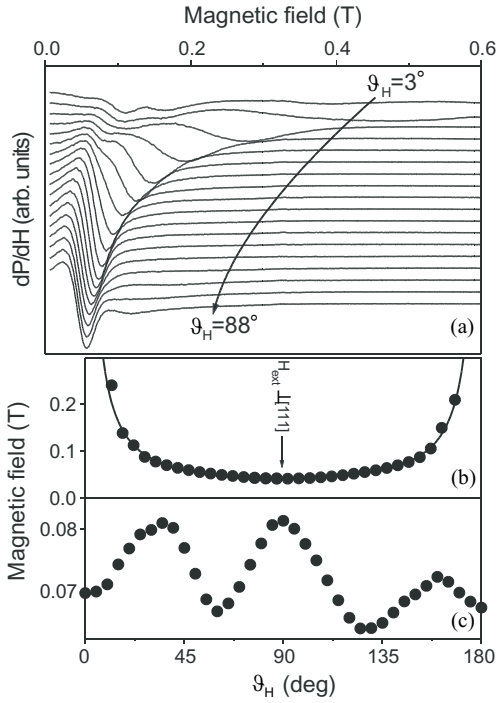

Fig. 1. Angular dependence of the ESR signal in samples on GaAs (1 1 1) substrate. The spectra in (a) are shifted for clarity. The angular steps are 5° in all graphs.

where H_A is the "anisotropy field" which is an empirical parameter. ϑ_H and ϑ_0 are the angles between the axis of anisotropy and H_{ext} and the magnetisation M_0, respectively. γ is the gyromagnetic ratio for a g-factor of 2. $\omega_0 = 2\pi\nu$, where $\nu = 9.35$ GHz is the microwave frequency. The model neglects demagnetisation effects and higher-order contributions to the anisotropy. The theoretical curves (solid lines in Figs. 1b and 2b) agree well with the experimental ones. Four important results are derived: (i) the crystallographic orientation of the clusters with respect to the matrix is the same for both samples, i.e. MnAs [0 0 0 1] ∥ GaAs [1 1 1]. This agrees with the crystal orientation reported for MnAs nanoclusters [8]. But it is different from those reported for MnAs thin films [14]; (ii) the easy axis of magnetisation is perpendicular to MnAs [0 0 0 1] for both samples, which is also common for MnAs bulk [15] and thin films [14]; (iii) at 200 K, the values of H_A are −1.3 and −0.58 T for the (1 1 1) and the (0 0 1) samples, respectively; (iv) all the

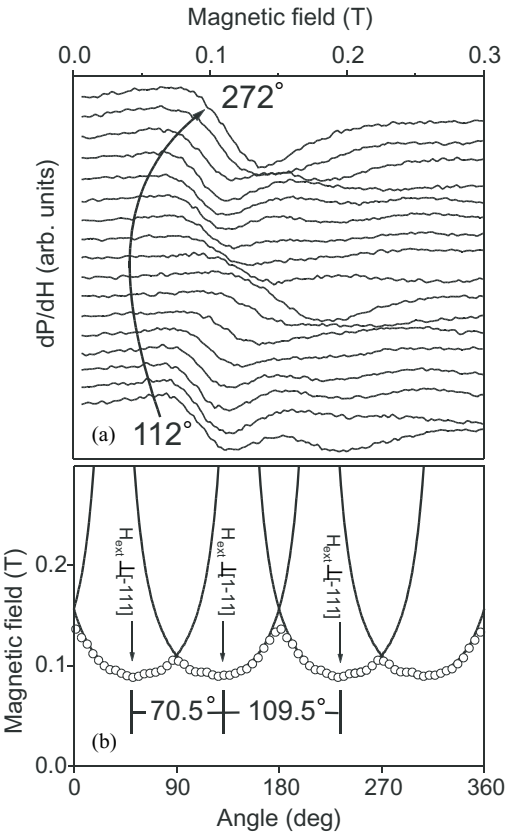

Fig. 2. Angular dependence of the ESR signal in samples on GaAs (0 0 1) substrate. The spectra in (a) are shifted for clarity. The angular steps are 10° and 5° in (a) and (b), respectively.

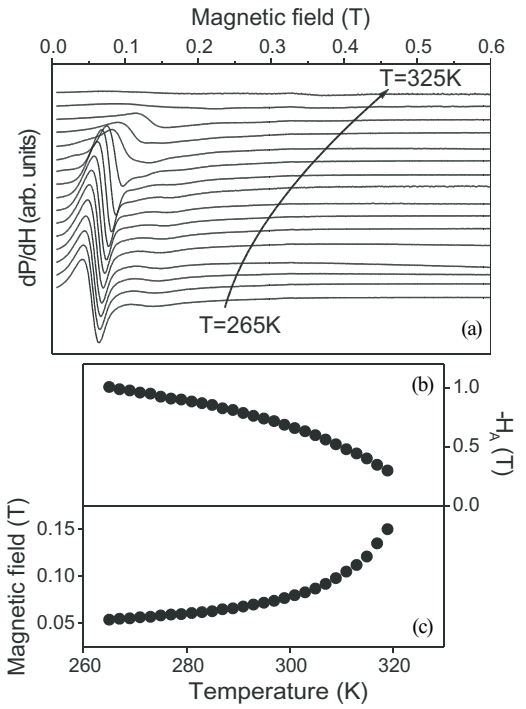

Fig. 3. Temperature dependence of the ESR signal in samples on GaAs (1 1 1) substrate. The spectra in (a) are shifted for clarity. The temperature steps are 4 K in (a) and 2 K in (b) and (c).

clusters in the (1 1 1) sample are oriented with the *c*-axis parallel to the growth direction. All magnetic moments will align in one direction in the plane of the layer for small H_{ext}. In contrast, the *c*-axis of the clusters in the (0 0 1) sample have four possible orientations along the equivalent GaAs [1 1 1] directions resulting in four, usually different directions of the magnetisation with respect to a small H_{ext}. This is reflected by the occurrence of more than one ferromagnetic resonance at most angles in Fig. 2b. Another angular dependence of H_{Res} for the (1 1 1) sample is shown in Fig. 1c, where the sample was rotated about the *c*-axis with H_{ext} perpendicular to the *c*-axis. An anisotropy with a 60° periodicity corresponding to the six-fold symmetry of the hexagonal plane of MnAs is observed. This in-plane anisotropy is at least by one order of magnitude smaller than the uniaxial anisotropy.

Figs. 3a and 4a show temperature-dependent ESR results. All measurements were performed with H_{ext} parallel to the easy axis of magnetisation (in the case of the (0 0 1) sample for one of the four possible orientations of the clusters). Figs. 3c and 4b depict the fitted resonance fields H_{Res}. Only one resonance is visible in the case of the (1 1 1) sample. The corresponding resonance field H_{Res} increases with temperature. Using Eq. (1) the anisotropy field H_A can be derived from H_{Res} at each temperature. H_A decreases as expected with decreasing ferromagnetism (Fig. 3b). The derived resonance fields H_{Res} for the (0 0 1) sample show a more complex temperature dependence (Fig. 4b). Three resonances are visible in this geometry. Two originate from the two cluster orientations with the *c*-axis in the plane of rotation, whereas the third originates from the remaining two degenerate cluster orientations with the *c*-axis out of the plane

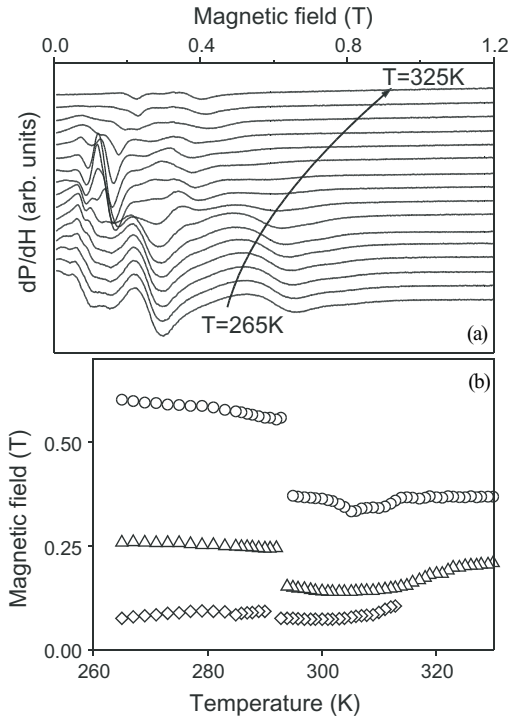

Fig. 4. Temperature dependence of the ESR signal in samples on GaAs (0 0 1) substrate. The spectra in (a) are shifted for clarity and the temperature steps are 4 K. In (b), the temperature steps are 2 K below 285 K and are 1 K in the vicinity of the phase transition.

of rotation. An abrupt reduction of H_{Res} of the three resonances occurs at about 290 K. Both samples remain ferromagnetic up to 330 K. The change of H_{Res} suggests that a structural phase transition might occur at 290 K. This would be in contrast to MnAs bulk material, where the magnetic phase transition coincides with a structural phase transition from hexagonal α-MnAs to orthorhombic β-MnAs at T_C [15]. Also, no change of signal is observed at 290 K for the (1 1 1) sample. Another possibility might be the decreasing anisotropy with increasing temperature. Below 290 K, the resonances at the higher fields originate from clusters, whose easy magnetisation axis are not parallel to H_{ext} (at least for $H_{ext} < 1.6$ T) due to the strong magnetic anisotropy. Above 290 K, the anisotropy is probably reduced such that all magnetic moments can align almost parallel to H_{ext}. Therefore, the required H_{ext} to reach the ferromagnetic resonances is abruptly reduced. Further experiments are needed to fully clarify this behaviour. The Curie temperature $T_C \approx 330$ K observed in both samples is slightly increased compared to $T_C \approx 318$ K of MnAs bulk material or thin films [15]. Possible explanations are the strain state or the single-domain character of the clusters, or the incorporation of a small fraction of Ga atoms into the clusters.

In conclusion, we have shown that (Ga,Mn)As-based hybrids consisting of a Mn-doped GaAs matrix with oriented MnAs clusters might be suitable for spin-electronic devices for operation above room temperature. From the point of view of the magnetic properties, the (1 1 1) samples offer advantages compared to the (0 0 1) samples. Further experiments to investigate possible applications are needed.

Funding by the DFG (SFB 383 and Graduate College "Optoelectronics of Mesoscopic Semiconductors") and by the BMBF (Contract No. 13N6917/0) is gratefully acknowledged.

References

[1] G.A. Prinz, Science 250 (1990) 1092.
[2] J. De Boeck, R. Oesterholt, H. Bender, A. Van Esch, C. Bruynseraede, C. Van Hoof, G. Borghs, J. Magn. Magn. Mater. 156 (1996) 148.
[3] H. Shimizu, M. Miyamura, M. Tanaka, Appl. Phys. Lett. 78 (2001) 1523.
[4] K. Takamura, F. Matsukura, Y. Ohno, H. Ohno, J. Appl. Phys. 89 (2001) 7024.
[5] H. Akinaga, S. Miyanishi, K. Tanaka, W. Van Roy, K. Onodera, Appl. Phys. Lett. 76 (2000) 97.
[6] K. Ando, A. Chiba, H. Tanoue, Appl. Phys. Lett. 73 (1998) 387.
[7] C. Chen, M. Cai, X. Wang, S. Xu, M. Zhang, X. Ding, Y. Sun, J. Appl. Phys. 87 (2000) 5636.
[8] P.J. Wellmann, J.M. Garcia, J.-L. Feng, P.M. Petroff, Appl. Phys. Lett. 71 (1997) 2532.
[9] Th. Hartmann, M. Lampalzer, W. Stolz, K. Megges, J. Lorberth, P.J. Klar, W. Heimbrodt, Thin Solid Films 364 (2000) 209.
[10] W. Heimbrodt, Th. Hartmann, P.J. Klar, M. Lampalzer, W. Stolz, K. Volz, A. Schaper, W. Treutmann, H.-A. Krug von Nidda, A. Loidl, T. Ruf, V.F. Sapega, Physica E 10 (2001) 175.
[11] M. Oestreich, J. Hübner, M. Bender, D. Hägele, W.W. Rühle, H. Kalt, Th. Hartmann, P. Klar, W. Heimbrodt, W. Stolz, Adv. Solid State Phys. 41 (2001) 173.

[12] V.A. Ivanshin, J. Deisenhofer, H.A. Krug von Nidda, A. Loidl, A.A. Mukhin, A.M. Balbashov, M.V. Eremin, Phys. Rev. B 61 (2000) 6213.
[13] A.G. Gurevich, G.A. Melkov, Magnetization Oscillations and Waves, CRC Press, Boca Raton, FL, 1996, ISBN/ISSN: 0849394600.
[14] L. Däweritz, Z.M. Wang, F. Schippan, A. Trampert, K.H. Ploog, Mater. Sci. Eng. B 75 (2000) 157.
[15] H. Katscher et al. (Eds.), Gmelin Handbook of Inorganic and Organometallic Chemistry Vol. 56 C9: Compounds of manganese with phosphorous, arsenic, antimony, 8th ed., Springer-Verlag, Berlin, 1983, ISBN: 3-540-93469-3.