Proceedings of the European Conference Physics of Magnetism, Poznań 2014

Magnetic Anisotropy in (Ge,Mn)Te Layers

W. Knoff[a,*], A. Łusakowski[a], A. Wołoś[a,b] and T. Story[a]

[a]Institute of Physics, Polish Academy of Sciences, al. Lotników 32/46, 02-668 Warszawa, Poland

[b]Institute of Experimental Physics, Faculty of Physics, University of Warsaw, Pasteura 5, 02-093 Warszawa, Poland

Ferromagnetic resonance study of magnetic anisotropy is presented for thin layers of IV–VI diluted magnetic semiconductor (Ge,Mn)Te with Mn content of 12 and 21 at.% grown by molecular beam epitaxy on BaF_2 (111) substrates. The layers with low Mn content grow in the rhombohedral crystal structure and exhibit perpendicular magnetic anisotropy whereas the layers with Mn content higher than about 20 at.% are of cubic (rock-salt) structure and show regular easy-plane type magnetic anisotropy. The quantitative analysis of the angular dependence of the ferromagnetic resonant field is performed taking into account the magnetic energy contributions due to rhombohedral distortion (axial term along the (111) growth direction of the layer) and the crystal field terms allowed for ferromagnetic systems of rhombohedral symmetry.

DOI: 10.12693/APhysPolA.127.404

PACS: 75.50.Pp, 75.30.Gw, 76.50.+g

1. Introduction

(Ge,Mn)Te is a IV–VI diluted magnetic (semimagnetic) semiconductor exhibiting carrier-induced ferromagnetic transition driven by the Ruderman–Kittel–Kasuya–Yosida (RKKY) indirect exchange interaction between well localized magnetic moments of Mn^{2+} ions via conducting holes [1, 2]. Depending on Mn content x and very high carrier concentration $p = 10^{20} - 10^{21}$ cm^{-3} the ferromagnetic Curie temperature T_C in p-$Ge_{1-x}Mn_xTe$ was found up to T_C =150 K for bulk polycrystals [3] and up to $T_C = 190$ K for thin epitaxial layers [4–7].

GeTe host crystal is known to undergo structural transition in which the high-temperature cubic (rock-salt) structure becomes distorted along cube's (111) diagonal to form at lower temperatures the crystal structure of rhombohedral symmetry. This transition is accompanied by the mutual shift of cation and anion sublattices supporting the ferroelectric state [8]. This structural transition takes place in GeTe well above room temperature at $T = 670$ K and can be controlled by the incorporation of Mn ions in $Ge_{1-x}Mn_xTe$. At room temperature the structural transition is observed for $x = 0.18$ in bulk polycrystals [3] and for $x = 0.2$–0.3 in crystalline thin layers grown by various methods [4–7,9–11]. The co-existence of ferromagnetism and ferroelectricity in single-phase $Ge_{1-x}Mn_xTe$ crystals gives opportunity to study their unique multiferroic properties. The coupling between structural and magnetic properties is expected to influence the magnetic properties of the layers, in particular resulting in perpendicular magnetic anisotropy observed in early magnetometric studies [4–7]. Very recently, magnetization and ferromagnetic resonance (FMR) measurements as well as phenomenological quantitative analysis of this effect and its relation to ferroelectric distortion was presented for 0.5 μm-thick $Ge_{1-x}Mn_xTe$ layers [9]. (Ge,Mn)Te is also known to undergo rapid crystalline-to-amorphous structural transition in a micro scale with metal-insulator and ferromagnet–paramagnet transition (magnetic phase change material) [10].

In this work, we apply ferromagnetic resonance method to in detail experimentally study the magnetic anisotropy in $Ge_{1-x}Mn_xTe$ layers ($x = 0.12$, $x = 0.21$) for which the X-ray diffraction technique identified the global crystal structure symmetry as rhombohedral ($x = 0.12$) or cubic ($x = 0.21$). We also examine the symmetry-allowed contributions to magnetic anisotropy energy as well as present phenomenological description of the angular dependence of the FMR resonance field.

2. Results and discussion

$Ge_{1-x}Mn_xTe$ layers with the thickness of about 1 μm were grown by molecular beam epitaxy (MBE) technique on insulating BaF_2 (111) substrates at $T = 420\,^{\circ}$C using GeTe, Mn, and Te_2 molecular sources. The growth of the layers was monitored *in situ* by reflection high-energy electron diffraction (RHEED) technique that revealed well defined streaky pattern characteristic for two-dimensional mode of layer growth. The Mn content in the samples studied ($x = 0.12$ and $x = 0.21$) was determined by energy-dispersive X-ray fluorescence analysis. Structural properties of $Ge_{1-x}Mn_xTe$ layers were examined by X-ray diffraction (XRD) method at room temperature. The layer with Mn content $x = 0.12$ revealed the (111)-oriented rhombohedral structure. In the $Ge_{1-x}Mn_xTe$ layer with Mn content $x = 0.21$ the cubic (rock-salt) structure was found. The relatively low value of full width at half maximum (FWHM) of about 600 arcsec derived from rocking curve of X-ray diffraction peaks observed in the layers proves their good crystalline quality.

Magnetic characterization of the layers was performed by superconducting magnetometry (SQUID) technique. Ferromagnetic Curie temperature T_C was found in

*corresponding author; e-mail: knoff@ifpan.edu.pl

the range $T_C = 30-40$ K from the analysis of the temperature dependence of magnetization $M(T)$. The Curie temperature of (Ge,Mn)Te crystals is known to vary depending on both Mn content and carrier concentration (controlled by crystal nonstoichiometry) [1–7, 9–11]. Magnetic hysteresis loop measurements were performed for external magnetic fields up to $H = 1$ kOe applied both perpendicular and parallel to the layer. The rhombohedral $Ge_{0.88}Mn_{0.12}Te$ layer revealed higher magnetization for the external field applied perpendicularly to the layer plane whereas in $Ge_{0.79}Mn_{0.21}Te$ layer higher magnetization was found for external field applied in the layer plane. In accordance with previous magnetic studies of (Ge,Mn)Te layers [4–7, 9–11] the rapid saturation of magnetization hysteresis loops observed experimentally already for fields of 300–1000 Oe corresponds to only about 1/3 of the total magnetization expected for a given Mn content. As experimentally shown the full magnetic saturation in (Ge,Mn)Te layers can be approached only at much higher fields of about 7 T, with the $M(H,T)$ dependence characteristic for the presence of antiferromagnetically coupled Mn clusters [11].

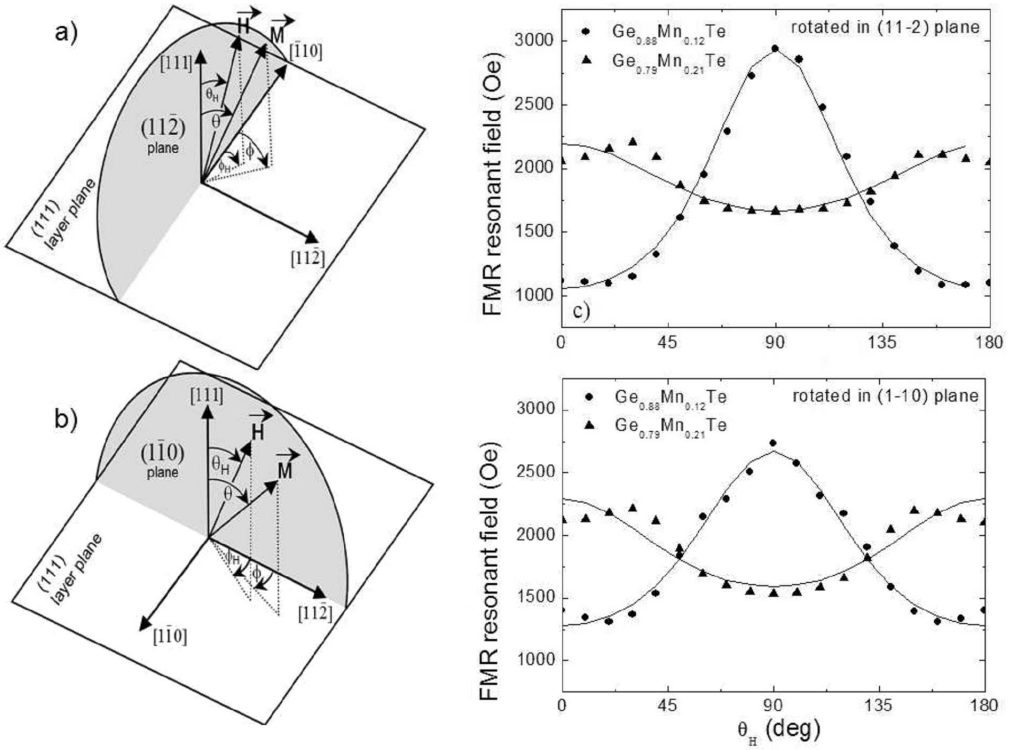

Fig. 1. The angular dependence of the FMR resonant field for two indicated rotation planes in $Ge_{1-x}Mn_xTe$ layers with Mn content $x = 0.12$ (a) and $x = 0.21$ (b). Solid lines show theoretical calculations based on the analysis of magnetic anisotropy energy (see Table).

The full angular dependence of magnetic anisotropy in (Ge,Mn)Te layers was studied by the FMR technique with Bruker X-band spectrometer operating at the microwave frequency of $\nu = 9.4$ GHz. This device is equipped with a continuous flow helium cryostat covering the temperature range $T = 3-300$ K and a goniometer permitting the control of the polar angle of applied magnetic field θ_H with the accuracy of about 1°.

The angular dependence of the FMR resonance field found in $Ge_{1-x}Mn_xTe$ samples with Mn content $x = 0.12$ and $x = 0.21$ is presented in Fig. 1. During the measurements the samples were rotated by 360 degrees (with a step of 10°) from [111] direction ($\theta_H = 0°$, normal to the layer) either to [1–10] direction or to [11–2] direction in the (111) layer plane (schematically shown in Fig. 1). The angle $\theta_H = 90°$ corresponds to magnetic field applied in the plane of the layer. Maxima and minima of the FMR resonance field clearly observed in both samples identify the hard and the easy axis of magnetization. From the FMR measurements of $Ge_{0.88}Mn_{0.12}Te$ layer the perpendicular magnetic anisotropy was found with easy magnetization axis for $\theta_H = 0°$. In contrast, for $Ge_{0.79}Mn_{0.21}Te$ layer this is the hard axis of magnetization. These observations qualitatively agree with the magnetization measurements reported in literature [4–7, 9, 11] as well as performed for the layers studied in this work.

In order to analyse quantitatively the experimental data we introduce a coordinate system with the z axis along [111] direction of the crystal (the growth direction of the layer). If the crystal has a cubic structure the x and y axes of coordinate system are along [11–2]

and [–110] directions, respectively. In this coordinate system the magnetic anisotropy free energy may be written in a form

$$\frac{F}{M} = -H\left[\cos\theta_H\cos\theta + \sin\theta_H\sin\theta\cos\left(\varphi-\varphi_H\right)\right] + a\cos^2\theta + b\cos^4\theta + c\sin^3\theta\cos\theta\cos 3\varphi, \quad (1)$$

where

$$a{=}2\pi M - \frac{1}{2}K_{4c}{+}K_2,\ b{=}\frac{7}{12}K_{4c}{+}\Delta K_4,\ c{=}\frac{\sqrt{2}}{3}K_{4c}.$$

The $2\pi M$ term results from demagnetization field for the ferromagnetic layer, the constant K_{4c} describes the cubic magnetic anisotropy of the crystal, finally K_2 and ΔK_4 take into account the possible deviations from the perfect cubic symmetry of the crystal (rhombohedral symmetry case). The direction of external magnetic field is described by the angles θ_H and φ_H whereas the direction of magnetization is defined by θ and φ. For a given θ_H and φ_H the resonant field is found from the equation

$$\left(\frac{\hbar\omega}{g\mu_{\mathrm{B}}}\right)^2 = \frac{1}{M^2\sin^2\theta}\left(F_{\theta\theta}F_{\phi\phi} - F_{\theta\phi}^2\right), \quad (2)$$

where ω is the resonance frequency, M is the magnetization saturation, $F_{\theta\theta}$, $F_{\varphi\varphi}$ and $F_{\theta\varphi}$ denotes second derivatives with respect to polar θ and planar φ angle calculated for θ and φ corresponding to the equilibrium direction of magnetization ($\partial F/\partial\theta = 0$, $\partial F/\partial\varphi = 0$).

TABLE

Parameters used in the analysis of the magnetic anisotropy of the FMR data presented in Fig. 1.

Mn content	rotation plane	K_2 [Oe]	$2\pi M$ [Oe]	g_{eff}
$x = 0.12$ (RH)	$(1\bar{1}0)$	–692	+242	3.08
	$(11\bar{2})$	–842	+242	2.97
$x = 0.21$ (RS)	$(1\bar{1}0)$	33	+207	3.70
	$(11\bar{2})$	–27	+207	3.65

The calculated angular dependence of the FMR resonance field is presented in Fig. 1 (solid lines) for two (Ge,Mn)Te layers and two rotation planes with the fitting parameters listed in Table. One can notice that the effect of perpendicular magnetic anisotropy in the rhombohedral layer with $x = 0.12$ is described very well just by effective uniaxial $K_2^{\mathrm{eff}} = K_2 + 2\pi M$ energy term with negligible magneto-crystalline K_4-type energy contribution. In contrast to other IV–VI DMS materials, like (Sn,Mn)Te or (Pb,Sn,Mn)Te [1, 12], the effective g-factor found experimentally in (Ge,Mn)Te layers in ferromagnetic phase is surprisingly large (also found in recent related work [9]) whereas the EPR measurements at high temperatures show regular value close to $g = 2.0$ as expected for Mn^{2+} ions. This effect apears to be correlated with magnetization deficiency mentioned above and is likely to result from the coupling between, presumably coexisting in (Ge,Mn)Te layers, ferromagnetically ordered Mn spins in conducting hole-rich regions and antiferromagnetically coupled Mn clusters in carrier-poor regions [2, 9].

The microscopic origin of this uniaxial term is related to spin–orbit dependent energy of p-(Ge,Mn)Te valence band electrons as is analysed elsewhere by density functional theoretical methods [13].

3. Conclusions

In conclusion, we applied ferromagnetic resonance technique to study the influence of crystal lattice distortion (cubic vs. rhombohedral) on the magnetic anisotropy in thin epitaxial layers of $Ge_{1-x}Mn_xTe$ ($x = 0.12$ and $x = 0.21$) deposited on BaF_2 (111) substrates. The qualitative change from perpendicular magnetic anisotropy in rhombohedral layer to regular anisotropy with in-plane easy axis in cubic layer was experimentally observed and phenomenologically, quantitatively described in terms of distortion-induced uniaxial anisotropy term along the layer growth direction.

Acknowledgments

This work was supported by the research project UMO 2011/01/B/ST3/02486 of Polish National Science Center (NCN).

References

[1] T. Story, *Acta Phys. Pol. A* **91**, 173 (1997).

[2] T. Dietl, H. Ohno, *Rev. Mod. Phys.* **86**, 187 (2014).

[3] R.W. Cochrane, M. Plischke, J.O. Ström-Olsen, *Phys. Rev. B* **9**, 3013 (1974).

[4] Y. Fukuma, H. Asada, S. Miyawaki, T. Koyanagi, S. Senba, K. Goto, H. Sato, *Appl. Phys. Lett.* **93**, 252502 (2008).

[5] W.Q. Chen, K.L. Teo, M.B.A. Jalil, Y.F. Liew, T.C. Chong, *Thin Solid Films* **505**, 145 (2006).

[6] W. Knoff, P. Dziawa, V. Osinniy, B. Taliashvili, V. Domuchowski, E. Łusakowska, K. Świątek, T. Story, *Mater. Sci.-Poland* **25**, 295 (2007).

[7] R.T. Lechner, G. Springholz, M. Hassan, H. Groiss, R. Kirchschlager, J. Stangl, N. Hrauda, G. Bauer, *Appl. Phys. Lett.* **97**, 023101 (2010).

[8] W. Jantsch, *Dielectric Properties and Soft Modes in Semiconducting (Pb,Sn,Ge)Te, Springer Tracts in Modern Physics*, Vol. 99, Springer, Berlin 1983.

[9] H. Przybylińska, G. Springholz, R.T. Lechner, M. Hassan, M. Wegscheider, W. Jantsch, G. Bauer, *Phys. Rev. Lett.* **112**, 047202 (2014).

[10] W. Knoff, K. Świątek, T. Andrearczyk, V. Domukhovski, P. Dziawa, L. Kowalczyk, E. Łusakowska, A. Šiušys, B. Taliashvili, J. Wróbel, T. Story, *Phys. Status Solidi B* **248**, 1605 (2011).

[11] W. Knoff, V. Domukhovski, K. Dybko, P. Dziawa, M. Górska, R. Jakieła, E. Łusakowska, A. Reszka, B. Taliashvili, T. Story, J.R. Anderson, C.R. Rotundu, *Acta Phys. Pol. A* **114**, 1159 (2008).

[12] T. Story, P.J.T. Eggenkamp, CW. Swüste, H.J.M. Swagten, W.J.M. de Jonge, A. Szczerbakow, *Phys. Rev. B* **47**, 227 (1993).

[13] A. Łusakowski, P. Bogusławski, *Acta Phys. Pol. A* **126**, 1177 (2014); arXiv:1502.01715v2.