Materials Science-Poland, Vol. 26, No. 4, 2008

Ferromagnetic and structural properties of $Ge_{1-x}Mn_xTe$ epitaxial layers

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Ferromagnetic and structural properties of semimagnetic (diluted magnetic) monocrystalline $Ge_{1-x}Mn_xTe$ layers ($x = 0.04$ and $x = 0.19$) grown by molecular beam epitaxy on $BaF_2(111)$ substrates were experimentally studied by the SQUID magnetometry and by high resolution X-ray diffraction analysis. Based on the temperature dependence of the lattice parameter, the structural phase transition from the rock salt to the rhombohedral crystal lattice was experimentally found at $T = 675$ K in $Ge_{1-x}Mn_xTe$ ($x = 0.04$) layer. The ferromagnetic Curie temperature T_c in $Ge_{1-x}Mn_xTe$ layers was determined based on the measurements of temperature and magnetic field dependence of magnetization. In $Ge_{1-x}Mn_xTe$ with the highest Mn content studied ($x = 0.19$) the mean-field like analysis of magnetic properties showed the Curie temperature $T_c = 40$ K.

Key words: *semimagnetic layer; magnetic properties*

1. Introduction

Substitutional solid solutions $Ge_{1-x}Mn_xTe$ belong to the IV–VI group of semimagnetic (diluted magnetic) semiconductors in which ferromagnetic transition is induced by very high (typically 10^{21} cm^{-3}) concentration of conducting holes via the Ruderman–Kittel–Kasuya–Yosida (RKKY) mechanism [1, 2]. $Ge_{1-x}Mn_xTe$ is characterized by the highest, in this semiconductor group, ferromagnetic Curie temperature T_c (up to 150 K for $x = 0.4$–0.5). This properly is closely related to the relatively large p–d exchange integral J_{pd} between the spin of localized Mn ions (electron configuration 3d^5, $S = 5/2$) and the spin of quasi-free carriers ($J_{pd} = 0.6$–0.8 eV), and large density of states in the valence band [1, 2]. $Ge_{1-x}Mn_xTe$ crystallizes in the rock salt structure (high temperature phase) and undergoes a structural transition to the low temperature rhombohedral crystal structure. This structural transition results in a displacement of

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$Ge_{1-x}Mn_xTe$ crystal lattice along the [111] body diagonal with a change of lattice symmetry known to be responsible for ferroelectric properties of these crystals. In bulk GeTe crystals, this structural transition takes place well above room temperature at 700 K whereas in $Ge_{1-x}Mn_xTe$ polycrystals the incorporation of Mn ions decreases the temperature of the structural transition [1]. Therefore, by technologically changing the content of Mn ions and the concentration of charge carriers one can effectively control the critical temperature of both ferromagnetic and ferroelectric transitions in $Ge_{1-x}Mn_xTe$ [1, 2]. This control can be particularly effectively done in thin layers of $Ge_{1-x}Mn_xTe$ in which the Mn content and the layer stoichiometry can be easily changed in a broad range of parameters [3–7].

In this work, the structural and magnetic properties of monocrystalline $Ge_{1-x}Mn_xTe$ layers have been studied with Mn content of 4 and 19 at. % grown on $BaF_2(111)$ substrates. Particular emphasis of the work concerns the experimental determination of the temperature of the phase transition from the rock salt to rhombohedral crystal structure and the ferromagnetic Curie temperature.

2. Growth of layers and their structural and magnetic characterization

$Ge_{1-x}Mn_xTe$ layers were grown on freshly cleaved BaF_2 (111) substrates using a home-built molecular beam epitaxy (MBE) system equipped with effusion cells for GeTe, Mn and Te. The growth process of $Ge_{1-x}Mn_xTe$ layers was performed at various technological conditions (changing substrate temperature and excess Te flux) with the vacuum of 10^{-8} mbar during the growth. Growth process was controlled *in situ* by reflection high energy electron diffraction (RHEED) diffractometer. The characteristic streaky RHEED pattern was observed indicating a two-dimensional mode of growth of the layers. Unfortunately, contrary, e.g. to closely related PbTe layers grown in the same technological facility, no oscillations of the specular spot intensity of RHEED pattern were observed. Chemical composition of $Ge_{1-x}Mn_xTe$ layers was determined by the energy dispersive X-ray fluorescence analysis, revealing Mn content up to 19 at. %. X-ray diffraction measurements performed at room temperature confirmed good crystalline quality and (111) – oriented rhombohedral structure of all layers with the rocking curve width parameter in the range 200–600 arcsec. Good chemical homogeneity of the layers was confirmed by the secondary ion mass spectroscopy (SIMS). Typically, the thickness of the layers was about 0.5 μm.

For the determination of the structural transition temperature, a $Ge_{1-x}Mn_xTe$ layer with Mn content $x = 0.04$ was studied employing high resolution X-ray diffractometer (Philips X'pert) in a broad temperature range 300–700 K. The analysis of the temperature dependence of the lattice parameter presented in Fig. 1 showed that the rock salt to the rhombohedral transition in this layer takes place about 675 K.

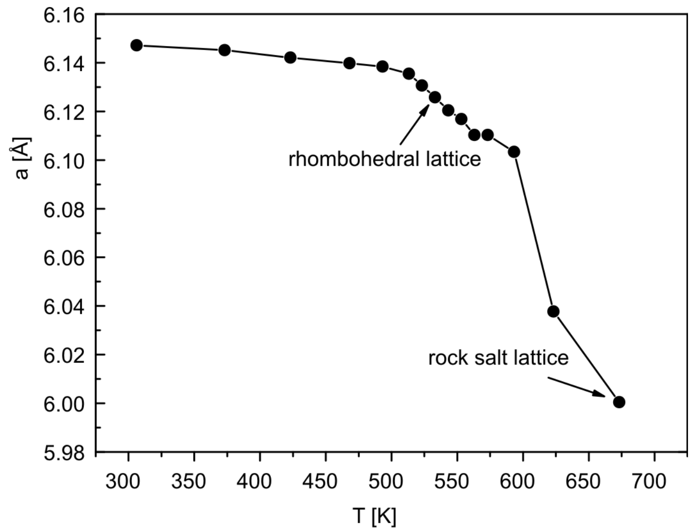

Fig. 1. Temperature dependence of the lattice parameter for $Ge_{1-x}Mn_xTe/BaF_2$ (111) layer with Mn content $x = 0.04$

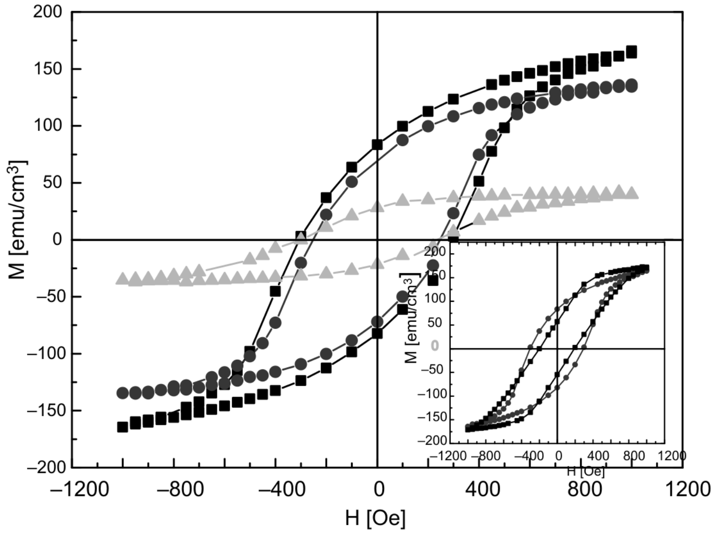

Fig. 2. Magnetic hysteresis loops of $Ge_{1-x}Mn_xTe$ ($x = 0.19$) layer at $T = 5$ K (squares), $T = 20$ K (circles) and $T = 40$ K (triangles). The inset presents the magnetization curves at $T = 5$ K for external magnetic field applied perpendicular (squares) or parallel (circles) to the plane of the layer

The temperature dependence of magnetization and the magnetic hysteresis loops of $Ge_{1-x}Mn_xTe/BaF_2(111)$ layers were recorded in the temperature range 5–150 K with a superconducting quantum interference device (SQUID) magnetometer. The external magnetic field up to 1 kOe was applied both in the layer plane (along the $[1\bar{1}0]$ crystal direction), and normally to the layer along the [111] crystal axis. In Figure 2, magnetic hysteresis loops are presented for a $Ge_{1-x}Mn_xTe$ layer with the highest Mn content studied, $x = 0.19$. This layer shows the ferromagnetic transition at ca. 40 K as evidenced by the temperature dependence of magnetization presented in Fig. 3.

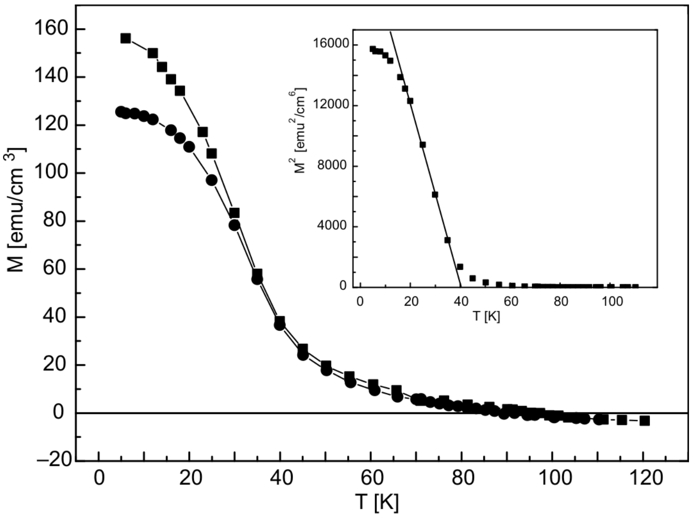

Fig. 3 Temperature dependence of magnetization for $Ge_{1-x}Mn_xTe$ layer with Mn content $x = 0.19$. The external magnetic field applied either perpendicular (squares) or parallel (circles) to the plane of the layer. The inset shows mean field model estimation of the Curie temperature from the $M^2(T)$ plot

We note that the temperature dependence of magnetization of this layer shows a mean-filed-like character (see the inset in Fig. 3) what is in contrast to previously studied layers revealing a very broad magnetic transition region [7]. In the inset in Fig. 2 as well as in Fig. 3, a comparison is presented of the temperature and magnetic field dependences of the magnetization of $Ge_{1-x}Mn_xTe$ layer measured with various orientations of magnetic field with respect to the layer (in-plane versus out-of-plane geometry).

3. Discussion and conclusions

Although the experimental data presented in Figs. 2 and 3 indicate a certain tendency of the $Ge_{1-x}Mn_xTe/BaF_2$ layer to exhibit normal to the plane easy axis, this effect is much smaller as compared to previously studied layers, for which a clear normal to the plane easy axis was experimentally found in both SQUID and ferromagnetic resonance studies [7]. As the Hall effect measurements of the layer with $x = 0.19$ indicated a very high carrier concentration (about 5×10^{21} cm^{-3}), it suggests that the variety of the magnetic field and temperature characteristics of magnetization observed in $Ge_{1-x}Mn_xTe/BaF_2$ layers is primarily governed by their electrical homogeneity and the concentration of carriers, with a negligible effect of possible irregularities in Mn ions distribution. The magnetization observed in the layer with $x = 0.19$ is close to saturation value expected for 5 Bohr magnetons per Mn^{2+} ion (160 emu/cm^3). However, experimentally we do not see a true magnetic saturation in fields available in our magnetometer (below 2 kOe).

In conclusion, we experimentally examined the crystal structure as well as the magnetic field and temperature dependences of magnetization of semimagnetic (diluted magnetic) semiconductor layers of $Ge_{1-x}Mn_xTe$ grown by MBE on $BaF_2(111)$ substrates. The structural transition from the rock salt to rhombohedral structure was analyzed based on the temperature dependence of lattice parameter. In the layer with the highest Mn content ($x = 0.19$) and carrier concentration of the order of 10^{21} cm^{-3} the ferromagnetic transition was observed at $T_c = 40$ K.

Acknowledgements

This work was supported by the research project 0992/T02/2007/32 of the Ministry of Science and Higher Education (Poland) granted for the period 2007-2010.

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Received 7 May 2007
Revised 30 May 2007