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ELECTRICAL, MAGNETIC, AND STRUCTURAL PROPERTIES OF $Sn_{1-x}Mn_x$ Te LAYERS GROWN BY MOLECULAR BEAM EPITAXY

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Layers of $\operatorname{Sn}_{1-x}\operatorname{Mn}_x\operatorname{Te}(x \leq 0.1)$ with thickness $0.2-2 \ \mu m$ were grown by molecular beam epitaxy on BaF_2 substrates with a $0.01-1 \ \mu m$ thick SnTe buffer layer. Both SnTe and $\operatorname{Sn}_{1-x}\operatorname{Mn}_x\operatorname{Te}$ layers show metallic *p*-type conductivity with conducting hole concentrations (at T = 77 K) $p_{77} =$ $7 \times 10^{19} - 2 \times 10^{21}$ cm⁻³. The layers grown under the conditions of an extra Te flux have a high carrier concentration and exhibit ferromagnetic phase transition at $T_{\rm C} \leq 7$ K. The layers grown with no (or very low) additional Te flux show low carrier concentrations (below 10^{20} cm⁻³) and remain paramagnetic in the temperature range studied $T = 4.5 \div 70$ K.

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Bulk crystals of $Sn_{1-x}Mn_x$ Te semimagnetic semiconductors are known to exhibit ferromagnetic, spin glass or paramagnetic properties depending on the Mn content and on the concentration of carriers. The ferromagnetic Curie temperature $T_{\rm C}$ can be varied by changing the concentration of carriers governed by the number of native defects (metal vacancies) [1-3]. Our work constitutes a part of the project aiming at the creation of the new layered semiconducting ferromagnet-diamagnet epitaxial structures. Based on the known magnetic and electronic properties of bulk crystals of SnMnTe and SnTe we expect, in these structures, to realize a model of low-dimensional magnetic system with controlled both magnetic and electronic properties. In particular, by changing the concentration of carriers one can control both the magnetic properties of magnetic element of a structure (SnMnTe) and the electronic properties (such as the Fermi wave vector $k_{\rm F}$) of a non-magnetic (SnTe) layer. These possibilities are important, e.g., for the study of the mechanisms of the interlayer exchange coupling. In this work we discuss the molecular beam epitaxy (MBE) growth as well as the structural, transport, and magnetic properties of semi-bulk $Sn_{1-x}Mn_xTe/SnTe/BaF_2$ layers.

Layers of $\operatorname{Sn}_{1-x}\operatorname{Mn}_x\operatorname{Te}(x \leq 0.1)$ with thickness $0.2-2 \ \mu m$ were grown by MBE on cleaved (111) BaF₂ substrates with a $0.01-1 \ \mu m$ thick SnTe buffer layer.

The growth rate was about $0.4-0.8 \ \mu m/h$. The process of crystal growth was monitored *in situ* by reflection high energy electron diffraction (RHEED). Apart from SnTe and Mn effusion cells, additionally a Te effusion cell was used to modify the growth conditions. The crystal structure and the chemical composition of our Sn_{1-x}Mn_xTe layers were examined by X-ray diffraction, X-ray fluorescence analysis and electron microprobe analysis. The layers were found to grow in NaCl structure with (111) orientation. The halfwidth of the (222) X-ray rocking curve was typically about 300 arcsec, which is comparable with the parameters of thin layers of other magnetic semiconductors. Chemical homogeneity of layers, as seen on a sample composition profile along the diagonal of a layer (Fig. 1) was quite good; visible fluctuations correspond to the accuracy of measurements.



Fig. 1. A composition profile along the diagonal of a $Sn_{0.94}$ Mn_{0.06} Te layer as determined by electron microprobe analysis. Distance, measured in 0.2 mm units, is presented on horizontal axis. Atomic percents in relation to total composition are presented on vertical axes, left axis for Sn and Te, right axis for Mn.

Transport properties of $SnMnTe/SnTe/BaF_2$ structures were studied at temperatures T = 300 K and T = 77 K by standard dc method applying magnetic fields $B \leq 1.2$ T. Since both SnMnTe layer and SnTe buffer exhibit semi-metallic electric properties, the analysis of conductivity and Hall effect measurements was performed applying theoretical expressions previously used in the measurements of electrical parameters of silicon layers grown on doped Si substrates [4]. In such a structure the measured (effective) conductivity and Hall constant depend on the conductivities, carrier concentrations and thicknesses of both constituent layers. The analysis of the electrical properties of the SnMnTe layer is quite straightforward in the case when the buffer layer is much thinner (or much less conducting) as compared to the SnMnTe layer. In the other limit, a large correction factor has to be introduced. This factor depends on the conductivity and mobility ratio for the SnMnTe and the buffer layer. Since these parameters are usually not well known, the resulting electrical parameters provide only an order of magnitude estimates. The results of our measurements carried out on SnMnTe and SnTe layers as well as on SnTe and Sn_{0.91}Mn_{0.09}Te bulk crystals are presented in Fig. 2. Literature data for bulk Sn_{1-x}Mn_xTe crystals with x = 0.02, 0.04, 0.06 [5] are also shown for comparison.



Fig. 2. Mobility of conducting holes versus their concentration in our $Sn_{1-x}Mn_xTe$ and SnTe layers as well as bulk crystals. Literature data (Ref. [5]) for bulk $Sn_{1-x}Mn_xTe$ crystals with x = 0.02, 0.04, 0.06 are presented for comparison.

By adjusting the additional flux of Te, we have grown SnMnTe layers with carrier (hole) concentration in the range of $p = 7 \times 10^{19} \div 2 \times 10^{21}$ cm⁻³. Particularly interesting is the possibility to grow SnMnTe crystals with a carrier concentration below 10^{20} cm⁻³ which is not possible in Bridgman-grown bulk crystals. The mobilities of conducting holes in our SnMnTe layers are rather low $(\mu_{77} = 15 \div 200 \text{ cm}^2/(\text{V s})$ depending on carrier concentration), and correspond to bulk SnMnTe with a high Mn content (see Fig. 2).

Magnetic properties of $\text{Sn}_{1-x}\text{Mn}_x$ Te layers were studied by ac magnetic susceptibility and by electron paramagnetic resonance (EPR) measurements. The temperature dependence of ac magnetic susceptibility presented in Fig. 3a gives clear evidence for the ferromagnetic phase transition at $T_C \leq 7$ K in SnMnTe layers grown with an extra flux of Te. These layers have a high carrier concentration $p = 10^{21}$ cm⁻³. Layers grown with no (or very low) additional Te flux exhibit low carrier concentrations (about 10^{20} cm⁻³) and remain paramagnetic in



Fig. 3. Temperature dependence of the ac magnetic susceptibility of a $Sn_{1-x}Mn_x$ Te layer (a 1 μ m thick layer on a 0.2 μ m buffer). In figure (a) a ferromagnetic phase transition is visible. Figure (b) shows the expanded paramagnetic region described by the Curie-Weiss law (solid line). From the Curie constant determined as a fitting parameter in (b) follows x = 0.04, whereas the electron microprobe analysis of the sample gives x = 0.09.

the temperature range studied $T = 4.5 \div 70$ K. We have, therefore, demonstrated that carrier-concentration induced paramagnet-ferromagnet transition observed in bulk crystals can be also realized in thin epitaxial layers. A necessary variation of the carrier concentration is achieved by a proper control of the growth conditions. Such control of magnetic properties by changing the carrier concentration is in the case of MBE grown SnMnTe layers much easier than in bulk crystals [6].

The analysis of the magnetic susceptibility of $\operatorname{Sn}_{1-x}\operatorname{Mn}_x$ Te layers shows that for $T > T_{\rm C}$ the magnetic susceptibility follows the Curie-Weiss law (see Fig. 3b) with the Curie constant indicating the Mn^{2+} ions content $x \leq 0.04$. This result agrees well with the observed ferromagnetic-transition temperatures as compared with the data for bulk crystals.

EPR measurements show a single line with the g-factor equal to 2.0. Temperature dependence of the intensity of the EPR line follows the Curie-Weiss law and supports the conclusions drawn from the measurements of magnetic susceptibility. A careful analysis of the intensity of the EPR line provides also an independent estimate for the Mn^{2+} content in our layers. There is satisfactory agreement between the Mn^{2+} concentration determined from magnetic susceptibility and from EPR data. This concentration however turned out to be generally lower than the Mn content x determined from electron microprobe measurements and from X-ray fluorescence analysis. The higher x value, the greater the difference. In $\text{Sn}_{1-x}\text{Mn}_x$ Te layers, obtained under the growth conditions applied in our experiments, the highest determined concentration of Mn^{2+} ions — that substitute for Sn^{2+} ions — is about 4% of all cations.

In conclusion, we have shown that thin layers of SnMnTe grown by molecular beam epitaxy exhibit transport and magnetic properties depending on the growth conditions. By controlling the Te flux from additional Te effusion cell we have grown SnMnTe layers with carrier concentrations in the range of $p = 7 \times 10^{19} \div 2 \times 10^{21}$ cm⁻³. Magnetic properties reveal the effect of carrier concentration dependent ferromagnetic transition at $T_{\rm C} \leq 7$ K.

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