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MAGNONS IN CUBIC MBE-GROWN $A_{1-x}Mn_xTe$ LAYERS (A = Cd, Zn, Mg)*

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One-magnon excitations in MBE-grown $A_{1-x}Mn_xTe$ layers (where A = Cd, Zn, Mg and $x > 0.7$) were investigated by means of the Raman scattering measurements at low temperatures ($\approx$ 20 K). The composition dependence of the anisotropy energy — as extracted from these measurements — is discussed. Further, the elastic neutron scattering measurements were performed in layers of cubic MnTe, which constitute the end point material of the ternary alloys series. Abundance of variously oriented antiferromagnetic domains in MnTe layers as a function of temperature was studied. We confirm occurrence of a pronounced magnetostriction effect.

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Molecular beam epitaxy (MBE) has been successfully used to grow new materials that cannot be produced by traditional growth techniques. In particular, cubic (zinc blende) diluted magnetic semiconductors (DMS), such as $Cd_{1-x}Mn_xTe$ and $Zn_{1-x}Mn_xTe$, were obtained by this technique for the composition range corresponding to very high magnetic ion concentrations ($x > 0.7$). Similarly, new magnetic semiconducting alloys $Mg_{1-x}Mn_xTe$, with both constituent binaries that do not crystallise normally in the cubic form, were also grown by MBE. The interesting aspect of these new possibilities introduced by MBE is that, as it is known, zinc blende Mn chalcogenides constitute unique cases of fcc Heisenberg spin systems with strongly dominating antiferromagnetic nearest-neighbour interactions (resulting in AFIII antiferromagnetic phase — see, e.g., [1, 2]). Therefore, they can be used as model systems for studies of magnetic properties and interactions to be confronted with existing theoretical predictions.

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Our knowledge of the details of magnetic properties of the materials under consideration is still not complete. There are many papers in the literature devoted to the problem of magnetic phase transitions in $Cd_{1-x}Mn_xTe$ and $Zn_{1-x}Mn_xTe$ thin layers and superlattices, including discussions on possible influence of internal strains on these properties (see, e.g., [1, 2] and references therein). Very recently, magnetic properties of $Mg_{1-x}Mn_xTe$ have been also analyzed [3] in this context. In order to determine important parameters characterizing magnetic structure of three ternary compounds mentioned above several methods were used, such as magnetization, magnetooptical and elastic neutron scattering measurements. It was demonstrated that the magnetic phase diagrams corresponding to three discussed diluted antiferromagnetic systems are quite similar [3].

The goal of the present paper is to study some properties of the collective magnetic excitations (magnons) in $A_{1-x}Mn_xTe$ crystals (A = Cd, Zn or Mg) for the composition range $x > 0.7$. Our investigations were performed by means of the Raman scattering measurements at low temperatures. The results of these optical studies were complemented by elastic neutron scattering data obtained on cubic MnTe, which constitutes the end point case of all three ternary alloys being investigated by us. Layers of cubic $Mg_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ (thickness ranging from 1 to 6 micrometers) were grown by MBE on (001) GaAs substrates (2 degrees off) with 1–3 μm CdTe buffer layers deposited prior to the magnetic layer growth. In the case of growth of $Zn_{1-x}Mn_xTe$, a thin layer of ZnTe was used as the most convenient buffer between substrate and the magnetic layer. The crystal quality of all DMS layers, as well as their chemical composition, were determined by X-ray diffraction using Co K_α radiation. The X-ray measurements did not reveal any crystallographic phases other than the cubic phase. They did show a presence of some residual strain in the epilayers.

The Raman spectra were recorded in a quasi-backscattering geometry at about 20 K using a U1000 Jobin-Yvon double monochromator equipped with holographic grating and a S20 photomultiplier. Samples were mounted on a cold finger of continuous flow helium cryostat, the Ar^+ laser lines were used for excitation. The elastic neutron scattering measurements were performed on two MnTe samples (having 4 and 6 micrometer thickness) in the usual "transmission" and "reflectivity" modes, using triple-axis neutron spectrometers installed on both thermal and cold neutron sources of ORPHEE reactor in Laboratoire Léon Brillouin (Saclay).

From the elastic neutron scattering measurements performed on our samples we were able to estimate the population of three types of antiferromagnetic (AF) domains. It was found that the dominant orientation of the domains tends to be the plane perpendicular to the MnTe film plane upon a decrease in temperature. At the lowest investigated temperature ($\approx$ 12 K) only about 10% of the sample volume corresponds to the domain with AF sheets plane parallel to MnTe epilayer. Our data demonstrate that the fraction of the sample volume corresponding to each of the two remaining orientations of AF domains (with AF sheets perpendicular to the MnTe plane) is temperature dependent and it increases to the value equal to about 0.45 at $T = 12$ K. Figure 1a shows the ratio of the sample volume related to two different (one of the perpendicular and the parallel) orientations of AF domains as a function of temperature. These findings complete precise experimental data

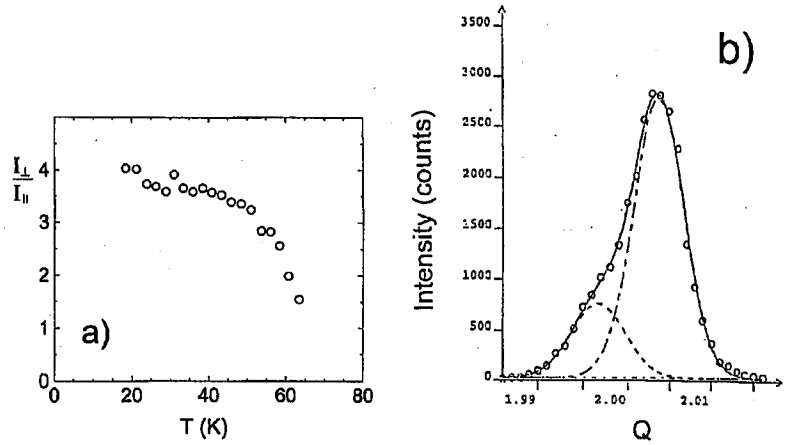

Fig. 1. (a) Temperature dependence of the ratio of the population of AF domains: a parallel domain and one of the perpendicular domains to the MnTe layer plane. (b) Elastic neutron scattering data taken for MnTe (002) Bragg peak at $T = 26$ K. Broken curves show the contributions from two lattice parameters, solid line corresponds to their sum. MnTe lattice spacing values in the growth direction, resulting from the fitting of measured spectrum, correspond to the values reported in the literature.

previously taken for 1 μm thick MnTe [1, 2] and $Zn_{0.06}Mn_{0.94}Te$ [4] layers deposited on ZnTe buffers. One can conclude that, as it was expected, the observed effect depends on the lattice mismatch between the investigated AF layer and the buffer layer. It is not clear whether it depends on the layer thickness or not. It is likely then that this effect is an intrinsic property of AF cubic MnTe at low temperatures and is related to magnetostriction (see below). Similar investigations performed in the case of MBE grown MnSe or MnS layers would be helpful in order to confirm this suggestion.

Direct lattice parameter measurements, performed by means of elastic neutron scattering demonstrate that in MnTe epilayers at low temperatures a magnetostriction effect takes place along the growth direction. In order to analyse this phenomenon we studied (002) Bragg peak. Above the Néel temperature ($T_N = 68$ K) the cubic lattice parameter value decreases slightly with a decreasing temperature. Below the Néel point — in the antiferromagnetic phase — the crystal unit cell is no longer cubic, because of the magnetostriction effect, and there are two lattice parameter values. Figure 1b shows the typical experimental curve taken by triple-axis neutron spectrometer. This effect was found earlier by X-ray diffraction measurements in cubic MnTe [2, 3] and $Zn_{0.06}Mn_{0.94}Te$ [4] layers. However, the temperature at which the lattice parameter begins to split, reported in [2–4], was equal to about 50–55 K (well below the Néel temperature determined for cubic MnTe). In our case the temperature at which the splitting appears is, within the accuracy of the experiment, equal to the magnetic phase transition temperature. The difference between these two results may be due to various strains present in two kinds of samples studied by us and in [2–4], respectively.

Features resulting from Raman scattering on magnons were observed for the first time in the optical spectra of two next DMS systems: cubic $Mg_{1-x}Mn_xTe$ and $Zn_{1-x}Mn_xTe$ (see Fig. 2). One-magnon structures found at the Raman spectra taken for these compounds at low temperatures can be compared with the earlier Raman scattering data obtained for MnTe and $Cd_{1-x}Mn_xTe$ layers [5, 6]. The observed composition dependence of the magnon frequency is quite similar in all three diluted AFIII systems. It is well known that the magnon frequency at the zone centre is given by a simple formula involving the exchange energy (depending on Mn–Mn exchange integrals) and the anisotropy energy (see, e.g., [7]). Our data show that the exchange energy is much greater than the anisotropy energy in the case of cubic MnTe (as it was previously suggested in the literature). One cannot exclude that Mn–Mn exchange integrals depend on the composition of the ternary mixed crystal. However, the exchange integrals change only very slowly with the mixed crystal composition within one ternary system as it was demonstrated in [8]. Thus, as the first approximation, one can assume that appropriate Mn–Mn exchange integrals should be the same in all investigated samples.

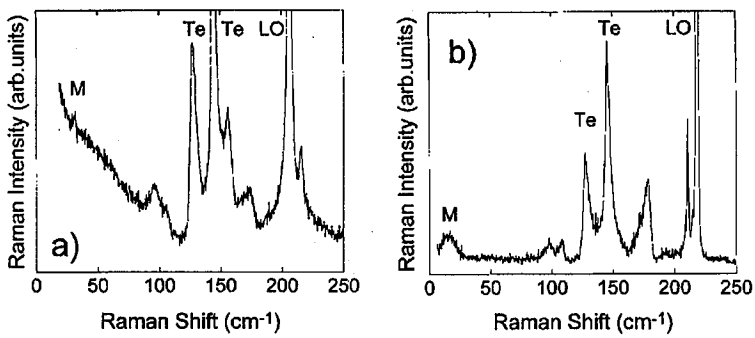

Fig. 2. Part of the Raman spectrum taken in $-z(x,y)z$ configuration at $T \approx 22$ K for $Mg_{0.15}Mn_{0.85}Te$ (a) and $Zn_{0.27}Mn_{0.73}Te$ (b) epilayers. Ar^+ 514.5 nm laser excitation line was used (spectral resolution better than 1 cm^{-1}). Structures denoted as M result from the scattering by magnons, the group of well pronounced structures in the centre are due to tellurium precipitates, LO peak corresponds to the longitudinal phonon branch for the "MnTe-like" optical mode.

By analysing the composition dependence of the magnon frequency we obtained that the anisotropy term decreases strongly with the dilution of AFIII magnetic system. In particular, assuming that two first exchange integrals J_1 and J_2 (describing interactions between the nearest neighbour and next nearest neighbours, respectively) for all mixed crystals under consideration are constant and equal to the values suggested in [8], we obtain a drop of the anisotropy energy by about one order of magnitude when decreasing Mn content from $x = 1$ to $x = 0.7$ in $A_xMn_{1-x}Te$ mixed crystals regardless the cation species. This unexpected, interesting phenomenon needs further experimental and theoretical investigations. It should be also mentioned that apart from magnon features and optical phonon

related structures a group of lines resulting from the Raman scattering on crystalline Te precipitates was found in the spectra taken for all investigated samples. This problem will be discussed elsewhere.

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