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Electrotransport and magnetic properties of metastable phases in the system GaSb-Mn synthesized under high pressure.

M.V.Kondrin¹, S.V.Popova¹, V.R.Gizatullin¹, O.A.Sazanova¹, N.V.Kalyaeva¹, A.G.Lyapin¹, V.V.Brazhkin¹, A.A.Pronin², S.A.Gudoshnikov³, Yu.V.Prokhorova³

¹Institute for High Pressure Physics, RAS, Troitsk, Moscow region, 142190 Russia
²General Physics Institute, RAS, 117942 Moscow, Russia
³Institute of Terrestrial Magnetism, Ionosphere, and Radio Wave Propagation, RAS, Troitsk, Moscow region, 142190 Russia

Abstract. The paper reports on electrophysical and magnetic properties of recently discovered high-pressure phases in the (GaSb)₁₋ₓMnx system, which is a possible candidate for spintronic material. It was shown that the simple cubic and tetragonal high-pressure phases display magnetic ordering although the critical parameters and electrotransport properties of these two phases significantly differ. Also we observed a magnetic ordering in the solid solution of Mn in the semiconductor sphalerite phase of GaSb prepared under high pressure, although in this case magnetic moment per Mn ion is an order of magnitude smaller than in the first two cases.

Close attention has been recently focused on the search for new materials for spintronics, i.e., electronics that involve both magnetic field and spin-polarized electron transport for signal transmission and modulation. In this respect, III-V semiconductors doped with magnetic ions are most attractive. However, the application of such compounds is yet complicated, because the Curie temperature is much lower than room temperature. To date, the properties of Ga₁₋ₓMnxAs with Tc=110 K are studied most completely and the attention of researchers is primarily focused on the search for new similar compounds with the different matrices and substitution of the Mn ion for the Group-III element [1]. A ferromagnetic phase has recently been found in wide gap semiconductor(Zn,Cr)/Te [2].

All the samples mentioned above were prepared as a thin films by the molecular beam epitaxy. On the other hand the preparation of bulk spintronics materials can provide a mean to rule out possible dimensional effects and to let us correlate electro transport and magnetic properties with crystal structure of synthesized samples. The possible candidate was the solid solution of Mn in GaSb matrix, whose thin-film analogies were studied in [3, 4]. Two previously unknown high-pressure phases of (GaSb)₁₋ₓMnx that are significantly different in structure were obtained in [5], where it was shown that the structure of the samples synthesized at high pressures near 6 GPa was primarily determined by the synthesis temperature Tsyn. For temperatures of Tsyn <470 K, solid solutions with the sphalerite structure are likely to be formed. This phase is structurally the same as conventional low-pressure phase GaSbI. A further increase in temperature to Tsyn=690-870 K leads to the synthesis of a phase with the tetragonal structure. In a narrow intermediate synthesis-temperature range of Tsyn=620-670 K, the simple cubic phase is formed.
The X-ray spectra of the tetragonal and simple-cubic phases are shown in Fig. 1. Although tetragonal phase is clearly identified as $I4/mcm$ CuAl$_2$-like structure the identification of simple-cubic one is still questionable. The number of data (X-ray diffraction and density measurements) provided an evidence that the unit cell of this structure is quite small with lattice parameter of about 2.94 Å (whose value slightly depends on initial Mn content and varies in the range ± 0.1 Å) and contains only 1.2 atoms per unit cell [5] (for 50% Mn initial concentration). The same structure is synthesized in a wide concentration range (10-60 at.% of initial Mn). It is also worth mentioning that on X-ray spectra we observe only small traces of pure Mn in negligible amounts (in comparison to its initial content) and no other elemental components in pure forms or binary compounds. So the X-ray data gives an evidence of quite high degree of phase homogeneity, the conclusion which is also supported by a preliminary scanning microscopy with elemental analysis.

We have examined the magnetic and electrophysical properties of all types of the (Ga, Mn)Sb compounds synthesized through temperature treatment under high pressure. In all three cases the samples synthesized under pressure has magnetic ordering although critical parameters of these phases differ. The existence of these high-pressure phases with magnetic ordering may account for the diversity of magnetic properties observed on the films of GaSbMn [3].
authors there suggested that the different properties observed for the samples grown at higher temperatures (above 560°C) and the one grown at temperatures below 300°C are due to the formation of the second ferromagnetic phase MnSb and/or MnSb$_2$ ($T_c = 587$ and 550 K). So the above-room Curie-temperature found in this type of material [4] was attributed to the ferromagnetic properties of this phase. But in the bulk samples prepared in [5] and investigated by us, X-ray study and chemical analysis revealed no traces of binary compounds of Mn and Sb.

The high temperature tetragonal phase demonstrates a metallic-like transport properties and is ferromagnetic in all temperature range where it does exist ($T<450\text{K}$). The temperature dependence of resistivity and magnetization curve at 293 K are shown in Fig. 2-e) and Fig. 3-b) respectively. The phase prepared at intermediate temperatures with simple cubic structure demonstrates strong dependence of kinetic coefficients on Mn content. With the rise of Mn concentration from 0.1 to 0.6 the $T_c$ is increased from 220 K up to 280 K. But at the same time this phase undergoes a metal-insulator transition. This fact is demonstrated by the resistivity data (Fig. 2 a-d) and Drude formula for “minimal” metallic conductivity:

$$\sigma_0 = (3\pi^2)^{-2/3}(e^2/\hbar)n^{1/3}$$

Taking into account the data shown on Fig. 2 and an order of magnitude estimate for carrier concentration $n \sim x/a^3$ (where $x$ is the Mn concentration and $a = 2.94 \text{Å}$ - lattice parameter [5]) we can see that the samples with $x < 0.5$ are on the insulator side of the metal-insulator transition while the samples with $x > 0.5$ are well onto the metal side. The magnetization curves for these type of samples taken at liquid nitrogen temperatures are shown in Fig. 3-a. The dependencies in the near-critical temperature region of magnetic polarization measured by SQUID magnetometer and magnetic susceptibility measured by AC inductive method for the sample with largest $T_c$ attained in our experiments are displayed in Fig. 4.

The samples of sphalerite phase have quite limited solubility of Mn atoms. Our upper-bound estimate is well below 5% of Mn. This number of metallic atoms has a little impact on electrotransport properties of sphalerite phase which is obviously a semiconductor, although by variation of synthesis procedure the resistivity of the obtained samples at helium temperature can take values in the range 10-10000 Ohm cm. The typical temperature dependence of resistivity is shown in Fig. 5. Despite the small Mn content the samples exhibit magnetic ordering at room temperature. The evidence for that is provided by magnetization hysteresis observed on these samples (Fig. 3-c), although in this case the magnetic moment per Mn ion is an order of magnitude less than the one observed in the tetragonal phase. It seems unlikely that this magnetic ordering is due to the second magnetic phase (i.e. tetragonal one), because under high pressure the regions of stability of tetragonal and sphalerite phases have no common border and are separated by the region where simple-cubic phase are formed with the $T_C$ a little lower than the room temperature. The possible mechanism may be like the one suggested for the description of diluted wide-gap semiconductors (Zn,Cr)Te [2], where it was provided an evidence for “patterned” distribution of magnetic-ion in semiconductor matrix, so the ferromagnetic ordering does exist in the region of rich Cr content.

Thus, new ferromagnetic phases with high Curie temperatures close to or above room temperature are formed in the GaSbMn system under high pressure. Solid solutions with the primitive cubic structure, which are formed in a wide range of the manganese content, are of most interest. Variation of the initial manganese content allows one to control not only the magnetic but also the transport properties of these compounds. An increase in the manganese content, on the one hand, enhances the ferromagnetic properties, which is manifested in an increase in the ferromagnetic transition temperature from $T_c = 230 \text{K}$ for $x = 0.1$ to $T_c = 220 \text{K}$ for $x = 0.6$, and, on the other hand, strongly reduces the resistivity in this content range. The phase with the tetragonal structure is ferromagnetic over the entire range of its stability ($T < 450 \text{K}$) and exhibits metallic conductivity. The magnetization at $T=77.3 \text{K}$ is equal to $M=0.58$
Figure 2. Resistivity of the high-pressure phases of the (GaSb)$_{1-x}$Mn$_x$ samples with various structures and various manganese contents: the simple cubic structure with $x = (a) 0.1$, (b) 0.2, (c) 0.5, and (d) 0.6 and (e) the tetragonal GaSbMn phase

Figure 3. Magnetization per Mn ion of three high-pressure phases of GaSb-Mn: a) simple-cubic at 77 K ($x=0.2$) b) tetragonal at 293K ($x=0.5$) and c) sphalerite phase at 293 K ($x=0.1$)
Figure 4. DC magnetic moment and AC magnetic susceptibility of the simple cubic sample at the vicinity of ferromagnetic transition.

Figure 5. Mott’s resistivity of the (GaSb)\textsubscript{0.8}Mn\textsubscript{0.2} sample with sphalerite structure.

\(\mu_B\) and 0.28 \(\mu_B\) per manganese ion for the simple cubic and tetragonal phases, respectively. The sphalerite phase with low Mn content (\(x<0.05\)) is semiconductor and exhibits magnetic ordering at room temperature with magnetic moment per Mn about 0.05\(\mu_B\).

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