Investigation of the magnetic properties of manganese silicide grown on i-GaAs substrate by pulsed laser deposition

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Abstract. In this paper we have investigated the properties of the MnSi ferromagnetic layer grown on a semi-insulating GaAs (100) substrate by pulsed laser deposition method. Basing on the analysis of the temperature and magnetic-field dependencies of the Hall voltage and magnetization, as well as on analysis of the electron paramagnetic resonance spectra, it was shown that the manganese silicide film with Mn content of about 45\% is ferromagnetic with a Curie temperature of \(\sim 370\) K.

1. Introduction
Spintronics is one of the main trends for the development of microelectronics. The important task of the spintronics technology is the fabrication of ferromagnetic materials with Curie point close to room temperature or above. Silicon - the main element of all semiconductor electronics, is typically a non-magnetic semiconductor. A number of papers have shown [1]-[3] that the introduction of manganese atoms into the silicon with high enough concentration films allows one to confer the magnetic properties to the material. In the present work we report on the investigation of thin manganese silicide films with near 45x55 \% Mn and Si composition.

2. Method of experiment
The magnetic properties of the structure were investigated by analyzing the magnetic field dependence of the Hall voltage, the Nernst-Ettingshausen effect [4], the EPR spectrum [2] and the magnetic field dependence of the magnetization recorded by the variable field gradient method [5].

Figure 1 shows a schematic diagram of the mounting of a sample for recording the magnetic field dependences of the Hall voltage and Nernst-Ettingshausen voltage. The Hall voltage was measured in the Wan-der-Pauw geometry with the sample temperature kept uniform along the sample.

Figure 1. Schematic picture of mounting the sample when recording the magnetic field dependences of the Hall voltage and Nernst-Ettingshausen; 1 - holder, 2 - sample, 3 - thermal grease, 4 - heating element (resistor), 5 - contacts, 6 - cooling radiator.

For the thermoelectric and thermomagnetic measurements one of the sample edges was kept under an increased temperature. For that reason an electric current was supplied on the resistor (FE) thus the Joule
heat raised the temperature of the corresponding edge. Another edge of the structure was kept in thermal contact with the radiator. The temperature gradient created this way causes a number of effects: a thermal current, a Seebeck voltage generation and the Nernst-Ettingshausen (NE) effect under the applied magnetic field. The latter phenomenon consists in the generation of a transverse potential difference under the applied both the temperature gradient at the edges of the sample and the external magnetic field normal to sample surface (Fig.1). The analysis of Nernst-Ettingshausen voltage, in conjunction with the Hall voltage, gives information on the scattering behavior of free carriers. According to [4], the Hall and Nernst-Ettingshausen voltages are equal to:

\[ U_{NE} = Q B \Delta T, \]
\[ U_H = R_H B I, \]

where \( Q \) – Nernst-Ettingshausen constant, \( R_H \) – is the Hall constant, \( B \) – external magnetic field, \( I \) – electric current passing through the sample (in the Hall measurements), and \( \Delta T \) – temperature difference between the edges of the structure under study (for NE measurements).

The Hall and Nernst-Ettingshausen constants depend on the true mobility of free charge carriers:

\[ Q = \left( \frac{1}{2} - r \right) A_r \frac{k}{e} \mu, \]
\[ R_H = - \frac{A_r \mu}{\sigma}, \]

where \( k \) – Boltzmann constant, \( e \) – elementary charge, \( \mu \) – true mobility of the free charge carriers, and \( \sigma \) – electric conductivity, \( r \) – is a Hall factor and \( A_r \) – is the scattering factor.

The Hall factor and the scattering factor are related by:

\[ A_r = \frac{3\sqrt{\pi} \Gamma(\frac{3}{2} + 2r)}{4 \Gamma^2(2 + r)}, \]

where \( \Gamma(x) \) – is gamma function.

Solving the obtained system with respect to mobility and the scattering factor, we obtain the expression:

\[ r = \frac{1}{2} - \frac{QeR_{sl}}{R_H k} \rightarrow \mu = \mu(r). \]

From expression (6), one can determine the true mobility. Knowing the scattering factor one can also determine the contribution of each scattering center (impurity or phonons).

In ferromagnetic semiconductors, the scattering of the charge carrier depends on its spin orientation, therefore, an additional term appears in expressions (1) and (2), leading to distortion of the magnetic field dependences of the Hall and Nernst-Ettingshausen voltages:

\[ U_H(B) = R_H B I + R_b M(B) I, \]
\[ U_{NE}(B) = Q_b B \Delta T + Q_M M(B) \Delta T, \]

where \( R_b \) and \( Q_M \) – are the constants of the anomalous Hall effect and anomalous Nersent-Ettinghausen effect, respectively.

By analyzing the characteristic form of these magnetic field dependences, it is possible to determine the magnetic state of the structure under study.
3. Experimental
Figure 2 illustrates the magnetic-field dependences of the Hall voltage recorded for the investigated sample at \( T = 300K \), \( T = 77K \) and \( T = 370K \).

![Figure 2](image)

**Figure 2.** The magnetic field dependence of the Hall voltage recorded at 300K (dashed line) and 77K (solid line) and 370K (dot).

The 77 K and 300 K dependences are non-linear evidencing on the presence of an anomalous Hall component in the Hall constant. A clear hysteresis loop can be seen at the \( U_{\text{Hall}}(B) \) dependence measured at 77 K. With the temperature increase, the coercive field decreases, and thus, no hysteresis can be detected at 300 K. At a temperature of \( T = 370K \) the \( U_{\text{Hall}}(B) \) dependence can be approximated by a linear function. This temperature can be estimated as the Curie point of investigated MnSi film.

To further confirm the presence of ferromagnetic properties, the magnetic field dependence of the magnetization of the structure was recorded by the alternating force gradient magnetometer [5]. Figure 3 illustrates the dependences of the magnetization of the investigated structure on the magnitude of the external magnetic field with the \( B \) applied perpendicular to the film surface (in-plane geometry) and parallel to the film surface (out-of-plane).

![Figure 3](image)

**Figure 3.** The magnetic field dependence of the magnetization of the structure under study with the external field is oriented in-plane (solid line) and out-of-plane (dashed line).

The presence of a hysteresis loop within the in-plane measurements indicates the presence of ferromagnetic ordering in the structure under study. In the out-of-plane measurement, no hysteresis loop was detected which is in agreement with the anomalous Hall effect measurements (Figure 2). This fact is believed to be due to the easy magnetization axis of the MnSi lies mostly in the plane of the film.

Figures 4a and 4b illustrate the magnetic field dependences of the Hall and Nernst-Ettingshausen voltages recorded at an average sample temperature \( T_{av} = 196.75 \text{K} \) and \( T_{av} = 390 \text{K} \), respectively. The average temperature was calculated by the formula: \( T_{av} = \left( T_{\text{hot}} + T_{\text{cold}} \right) / 2 \).
Figure 4. Magnetic field dependences of the Hall voltage and Nernst-Ettingshausen, registered with: (a) \( T_{av} = 196.75 \) K, (b) \( T_{av} = 390 \) K.

From the graph illustrated at Figure 4a it can be seen that the magnetic-field dependences of the Hall and Nernst-Ettingshausen voltages have a clearly nonlinear form, whereas at 390 K a linear dependence is clearly visible (see Figure 4b). The change of the magnetic field dependence of the Nernst-Ettingshausen voltage, along with the change of the magnetic-field dependence of the Hall voltage, mentioned above, additionally attests to the presence of the "ferromagnet-paramagnet" phase transition point in the structure under study. From the analysis of the recorded magnetic field dependences of the Hall voltage and the Nernst-Ettingshausen voltage over a wide temperature range (20 ± 430) K, the temperature dependence of the true mobility of free charge carriers (Figure 5a), the scattering factor and Hall factor (Figure 5b) were calculated. In this case the \( R_H \) and \( Q_0 \) coefficients below the Curie point were derived by analyzing the linear part of \( U_H(B) \) and \( U_{NE}(B) \) dependences respectively. The temperature dependence of the mobility is given in the Arrhenius plot.

Figure 5. Calculated temperature dependences: (a) - true mobility of free charge carriers, (b) - scattering factor and Hall-factor.

From the graphs illustrated in Figures 5a and 5b it is seen that up to 70 K the carrier scattering was due to impurity ions only - the mobility increases with increasing temperature, the scattering factor is about 1.5. After passing through this temperature point, the contribution of scattering by acoustic phonons increases, which leads to a decrease in the scattering factor and a decrease in the mobility of the charge. The last consequence leads to the fact that the values of the Hall voltage signal and the Nernst-Ettingshausen voltage decrease, which can be seen on the graphs above.

The "ferromagnet-paramagnet" phase transition was also studied by analyzing the spectra of electron paramagnetic resonance of EPR recorded in the temperature range of 300-500 K. Figure 6a illustrates the EPR spectrum of the sample under investigation at 300 K, and Figure 6b shows the temperature
dependence of the magnitude of the peak amplitude on the spectrum, which is normalized to the maximum value in the investigated range (by the peak amplitude at room temperature).

**Figure 6.** (a) - EPR Spectrum of the structure under study, (b) is the experimentally obtained temperature dependence of the peak value on the EPR spectrum of the sample under study.

One can see the rapid drop of the magnetization at ~ 350 K, however above 350 K the value of $M$ does not drop down to zero and is preserved even at the highest temperature used. The latter is believed to be due to the presence of at least two magnetic phases in the film. The Curie temperature of the first phase, as shown by other studies, is about 370K, the Curie point of the other phase is at a temperature above 525K. We note that the presence of the second magnetic phase was not detected neither via magnetotransport measurements, nor via anomalous Nernst-Ettingshausen effect measurements. This can lead us to conclusion that the ratio of the second magnetic phase with respect to the first one is relatively low thus it does not have a significant influence on the electronic transport.

**In conclusion**

In this work, the magnetic properties of manganese silicide film were studied. It was shown that the presence of a large amount of manganese in silicon can lead to a ferromagnetic ordering of a structure with a Curie temperature of $T = 370 \text{ K}$, which opens prospects for the instrumental use of this material.

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