Physics-Chemical Analysis of Semiconductor-Ferromagnet Systems as a Basis of Synthesis of Magnetic-Granulated Spintronic Structures

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Abstract. An analytical review devoted to the physicochemical principles of the synthesis of granular structures in semiconductor-ferromagnet systems is represented. In these systems, as semiconductors, compounds are $\text{A}^{\text{II}}\text{B}^{\text{IV}}\text{C}^{\text{V}}_2$, $\text{A}^{\text{III}}\text{C}^{\text{V}}$, $\text{A}^{\text{II}}_2\text{C}^{\text{V}}_3$ and $\text{A}^{\text{II}}\text{C}^{\text{V}}_2$ and manganese compounds ($\text{MnP}$, $\text{MnAs}$ and $\text{MnSb}$) as ferromagnets. It is shown that in magneto-transmitter devices magneto-granular structures are an alternative to superlattices, and the effects of GMR and TMR are also possible. Advantages of magneto-granular structures are considered, such as: less labor-intensive methods of production, milder requirements for the dimension of a ferromagnet and a non-magnet, the possibility of forming a stable interface, soft requirements to the thickness of layers than in the case of superlattices, etc. It is shown that, due to the high mobility of charge carriers, the use of semiconductors as a matrix is more preferable than metals or dielectrics. The basic principles for the creation of granular structures with high values of magnetoresistance based on eutectic-type systems are formulated. During the crystallization of the eutectic, the simultaneous crystallization of all the phases that make up the eutectic takes place, leading to the formation of a specific fine-dispersed structure. The use of ultrahigh supersaturations leads to significant supercooling, which contributes to metastable crystallization. This causes a synergistic effect that stimulates nanostructuring, and promotes the creation of granular structures. The results of investigations of semiconductor-ferromagnet systems are presented and the possibility of obtaining magnetogranular structures with high magnetoresistance in them is shown.

Keywords: spintronics, magneto-granular structures, semiconductors, ferromagnets

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1. Introduction
Granular structures are an alternative to superlattices in spintronic devices. Currently, spintronics is one of the most dynamically developed areas of electronics. This is due to the fact that it is more energy-efficient to control the electron spin than the electron charge. In this regard, research on the creation of spin diodes, transistors and other traditional electronics devices is intensively carried out in Russia and abroad. However, while research on the creation of such devices are at the model levels [1, 2]. Actually existing spintronic devices are magnetic memory devices. Magnetic memory devices are produced in large quantities, because are part of all modern computers [3, 4]. Magnetic memory
devices are based on two effects, the effects of giant (GMR) and tunnel (TMR) magnetoresistance. [5, 6]. Effects were found on superlattices, which consisted of nanolayers of a magnetic and non-magnetic ones. Heterogeneous magnetically ordered systems are considered as analogues of superlattices, better known in the literature as magnetically granulated structures [7-11]. The effects of GMR and TMR are also observed on such structures [12–16]. Magnetogranular structure consists of a nonmagnetic matrix and ferromagnetic nanoparticles. Metals, semiconductors and dielectrics can be used as matrix materials [17–23].

The nature of the GMR effect in magnetically granular structures and superlattices is similar. Figure 1 shows the scheme of the appearance of the HMS effect. Despite the fact that the magnitudes of the effects of GMR and TMR in magnetically granulated structures are smaller than in superlattices, however, these structures have several advantages [24]. Of the advantages, there are: less labor-intensive methods of obtaining, softer requirements for the size of a ferromagnet and non-magnetics, better possibilities for creating a stable phase boundary between a ferromagnet and a nonmagnet, softer requirements for the thickness of layers, etc. Therefore, the synthesis and study of the properties of magnetically granular structures are rather intensive. Nonmagnetic metals or dielectrics, usually oxides, are used as the matrix material. Iron, cobalt, nickel or alloys based on them are chosen as ferromagnets. In our works, semiconductors are used as the matrix material. Semiconductors have high mobility of charge carriers; this allows increasing the spin relaxation time and increasing the polarization efficiency of the spin transport.

2. PHASE EQUILIBRIA IN SEMICONDUCTOR-FERROMAGNET SYSTEMS

For the synthesis of a magnetically granular structure in semiconductor-ferromagnet systems, components are needed that would form a eutectic between themselves and have minimal inter-solubility. During eutectic crystallization, simultaneous crystallization of all phases that make up the eutectic occurs, which forms a specific fine structure. The use of ultra-high supersaturation, leads to a significant overcooling of the medium, and cause metastable crystallization. Both of these factors cause a synergistic effect that contributes to nanostructuring when creating a granular structure.

Manganese compounds (MnP, MnAs, MnSb) were chosen as ferromagnetic components of eutectic type systems. These compounds are semi-metallic ferromagnets with high Curie temperatures [25, 26]. Compounds A\textsuperscript{II}B\textsuperscript{IV}C\textsuperscript{V}, A\textsuperscript{II}C\textsuperscript{V}, A\textsuperscript{II}2C\textsuperscript{V} and A\textsuperscript{II}C\textsuperscript{V}\textsuperscript{2}, which include an element of the fifth group, were chosen as semiconductors. The crystal structure of these semiconductors is significantly different from the crystal structure of MnP, MnAs and MnSb, which suggests the possibility of the formation of systems of the eutectic type or systems with significant areas of delamination.

The study of semiconductor-ferromagnet systems was performed using a complex of physicochemical analysis methods: XRD, DTA and DSC. Microstructural studies were performed using SEM, AFM and optical microscope "Epicuant". The results of these studies are presented in Table 1. The table shows the composition, melting points and types of eutectic microstructures. As can be
seen from the data in the table, the systems were of the eutectic type, the microstructures of the eutectics depending on the composition were plate-like, columnar, needle-shaped [27, 28].

### 3. SYNTHESIS AND PROPERTIES OF MAGNETIC GRANULATED STRUCTURES

Based on the works of Ya.B. Zeldovich on nucleation, under conditions of significant supersaturation [29, 30], composites were synthesized with nanoclusters of MnAs and MnSb ferromagnets with $T_C = 290–600$ K. Subsequently, the effect of the dispersion factor on the electrical and magnetic properties in a wide range of temperatures and magnetic fields was studied on these composite alloys [31]. In the ZnGeAs$_2$-MnAs system, two types of samples were prepared. Upon receipt of the first type, the melt crystallized under normal conditions; in the off-furnace mode, the cooling rate was $\sim 1.5 \cdot 10^{-2}$ deg/s. The melts of the second type crystallized in quenching mode. The cooling rate was $\sim 1 \cdot 10^2$ deg/s. Salt solutions with high thermal conductivity were used as a quenching medium. Microstructural studies have shown that the synthesized alloys were of the eutectic type, the size of the manganese arsenide crystallites in them depended on the cooling rate. With an increase in speed of 4 orders of magnitude, the average crystallite size of manganese arsenide changed by 3 orders of magnitude from $5 \cdot 10^4$ to $\leq 6 \cdot 10^1$ nm. In samples of the second type, unlike samples of the first type, the DSC curves showed no thermal effect of the structural transformation $\alpha$–$\beta$ MnAs, while the Curie temperature shifted towards higher values, from 318 to 351 K, respectively. Significant differences in the properties show the increasing role of surface phenomena with increasing dispersion.

The magnetoresistance was measured on granular structures of alloys: ZnGeAs$_2$ and CdGeAs$_2$ with MnAs. In Fig. 2 (a) shows the change in resistance from the magnetic field for the ZnGeAs$_2$ alloy with MnAs. At the beginning, the resistance dropped sharply with increasing magnetic field, and then saturation was observed. The magnitude of the magnetoresistance reached 60%. The type of magnetoresistance practically corresponded to the results observed by A. Fert and P. Grünberg when opening the GMR effect on superlattices [5, 6]. For CdGeAs$_2$ alloys with MnAs, the change in resistance had a different appearance. The resistance increased sharply with increasing magnetic field and there was no saturation (Fig. 2b). The known models of magnetoresistance cannot explain this nature of the change in resistance. As an attempt to find an explanation, alloys of ZnGeAs$_2$ solid solutions with CdGeAs$_2$ were prepared with the subsequent introduction of manganese arsenide. To determine the boundaries of solid

![Fig. 2. Magnetic field/resistance dependence for alloys: a) ZnGeAs$_2$ with MnAs, b) CdGeAs$_2$ with MnAs.](image_url)
solutions, the ZnGeAs$_2$–CdGeAs$_2$ system was studied. The results were unexpected [32, 33]. Despite the proximity of the crystal structures of compounds, the formation of solid solutions between them occurs in narrow boundaries (Fig. 3) with a large decay region. Therefore, magnetic field measurements were performed on alloys of solid solutions with a composition close to the compositions of ZnGeAs$_2$ and CdGeAs$_2$ compounds. The nature of the change in resistance (Fig. 4) from the magnetic field, as in the case of solid solutions from the side of ZnGeAs$_2$, and from the side of CdGeAs$_2$ did not change, but the magnitude of the effects decreased, as if their convergence occurred. The different type of change in resistance from the magnetic field for composite alloys ZnGeAs$_2$ and CdGeAs$_2$ with MnAs corresponds to the difference in the microstructures of their eutectic eutectics. In the first case, it is lamellar; in the second, needle-like, a form effect is observed. The needle-like view of the granular structure of CdGeAs$_2$ with MnAs is a classical solenoid, which leads either to the appearance of percolation processes or to the formation of a complex spin-valve structure. In both cases, a positive effect of magnetoresistance is possible [34, 35].

The interesting results were obtained on granular structures of alloys of the Ga(In) Sb–MnSb systems. Of greatest interest were the data of the magnetoresistance of granular structures. The average size of ferromagnetic MnSb clusters in granular structures was 24 nm [36-40]. Fig. 5 presents the temperature dependence of the magnetization of the eutectic alloy of the GaSb-MnSb system. The Curie temperature ~ 600 K was determined by the MnSb clusters. The dependence of resistance in a magnetic field had a complex form. At small values of the magnetic field (up to a saturation of 0.8 T), the resistance dropped sharply, and at large values it grew slowly. At low magnetic fields, negative magnetoresistance was observed, and at high magnetic fields, which was observed both at low temperatures of 5 K and at room 300 K (Fig. 6). The same type of change in resistance from the magnetic field was observed by A. Fert and P. Grünberg when the GMR effect was discovered [5, 6]. The change in the mechanism of charge carrier scattering occurs due to the fact that at low magnetic fields
up to saturation, scattering associated with the spin-dependent effect predominates, and at high magnetic fields, the resistance changes due to the Lorentz force. The change in resistance is insignificant in magnetic fields above saturation, but it is positive. Using pulsed laser deposition, using composite alloys of eutectics of the GaSb-MnSb and InSb-MnSb systems as a target, films were obtained. GaSb films with MnSb were synthesized on leucosapphire substrates, the film thickness was 80-100 nm. According to electron and atomic force microscopy data, the films were homogeneous [38, 41]. The electrical conditions of deposition largely influenced their electrical properties. The best samples had a hole type with a specific resistance of $7 \cdot 10^{19}$ cm$^{-3}$ at 300 K. The dependences of the specific magnetization and resistance on the magnetic field are presented in Fig. 7a,b respectively. The type of change in film resistance coincided qualitatively with bulk samples (Fig. 6). There was only a difference in the magnitude of the saturation magnetic field. In films, it is lower than $\sim 0.2 \, T$, and in bulk samples it was $\sim 0.8 \, T$.

Films of the InSb + MnSb composite with hole type conductivity were formed on single-crystal n-type InSb substrates by pulsed laser deposition and diode structures were formed [42, 43]. Fig. 8a shows the formation of these diode structures, Fig. 8b shows the current-voltage characteristic (VAC) of the $p$-(96% InSb + 4% MnSb)/n-InSb diode structure at 300 K: 1 - $B = 0 \, T$; 3 - $B = 0.15 \, T$ in the perpendicular field of the heterojunction plane.

Diodes based on this granular structure showed high sensitivity to a magnetic field. The magnitude of the current in a magnetic field of 0.15 T, applied perpendicular to the plane of the diode structure, decreased more than 9 times from 0.35 A to 0.04 A at a voltage of 1 V.

4. CONCLUSION
It is shown that the use of semiconductors as a matrix of magnetically granulated structures due to the high mobility of charge carriers is preferable to metals or dielectrics. The basic principles of creating granular structures with high values of magnetoresistance based on eutectic type systems are formulated. During eutectic crystallization, simultaneous crystallization of all phases takes place, and a specific fine-dispersed structure of eutectic alloys is formed. The use of ultra-high supersaturation leads to metastable crystallization, which causes a synergistic effect leading to nanostructuring, which is necessary when creating granular structures.
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