Nano- and heterostructures of magnetic semiconductors for spintronics

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The magnetic, electron transport, and optical properties of ordered nano- and heterostructures of Group III—V and Group IV diluted magnetic semiconductors are studied. A comparative analysis of the magnetic and electron transport properties of nanowires, thin films, and bulk semiconductors doped with transition metals is carried out. The size effect on the spin dynamics, magnetic properties, and magnetoresistance is discussed. Prospects for application of nano- and heterostructures with combined magnetic, electrical, and optical properties are considered.

Key words: diluted magnetic semiconductors, thin films, nanowires, heterostructures, spin dynamics.

The development of physics of magnetism began with studies of natural magnetic materials, such as magnetite, pyrrhotite, *etc*. Among the variety of known minerals, only a few are characterized by natural magnetic ordering. Therefore, rapid progress in physics of magnetic phenomena has been since elaboration of methods for the preparation of pure ferromagnetic metals and various alloys based on them (including alloys based on rare-earth metals). The variety of practical applications of such systems is due to the possibility of targeted preparation of magnetic alloys with specified properties. This technique was replaced by a more elegant approach, *viz*., chemical design of molecular magnets. By finely tuning the magnetic ordering conditions (types of magnetic atoms, atom-atom distances, types of chemical bonds, crystal lattice symmetry, *etc*.) one can in some cases predict the sign and magnitude of the exchange coupling constant J from "chemical" considerations (generally, this is impossible in spite of centuries-old history of the physics of magnetism). Chemical design is used to prepare polyfunctional materials whose magnetic properties are combined with some other properties (optical, electrical, *etc*.). However, practical application of molecular magnets is complicated because they usually have low Curie temperatures, low charge carrier mobility, and are poorly integrable with modern Si-based electronics. These difficulties can be avoided using yet another, "technological", approach to manufacturing of polyfunctional materials with specified magnetic properties, *viz*., the design of diluted magnetic semiconductors. They are obtained by partial replacement of atoms in the crystal lattice of a semiconductor by transition metal ions (usually, the proportion of impurity atoms is 1—10 at.%); indirect exchange coupling between these ions through charge carriers leads to a long-range magnetic order. This approach offers considerable prospects for fine "tuning" of properties, namely, an increase in the Curie temperature and magnetoresistance, *etc*. The foundations of physics of macroscopic magnetic semiconductors were developed earlier.[1—4] At present, researchers work on the design of nano- and heterostructures of ferromagnetic semiconductors for spintronic applications.

The aim of the present study is to carry out a comparative analysis of different types of Group III—V and Group IV diluted magnetic semiconductors, nano- and heterostructures based on them, as well as their magnetic and electron transport properties.

Experimental

The temperature dependences of the magnetization (M) of samples were measured on a Quantum Design MPMS 5XL SQUID magnetometer. Measurements were carried out in the

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temperature range 2—300 K in a constant magnetic field of strength 1 kOe.

Electron spin resonance spectra were obtained on a Bruker E 500 X-band spectrometer operating at about 9.650 GHz with an H102 rectangular cavity (modulation frequency was 100 kHz and the constant magnetic field sweep range was 0—14 kOe). The temperature was varied in the range 4—300 K using an Oxford Instruments ESR 900 cryostat.

Images of the samples were taken on a Philips Tecnai F 20 TEM transmission electron microscope at an accelerating voltage of 200 keV; the resolution of the microscope was 0.10 to 0.24 nm.

Circularly polarized photoluminescence was measured in magnetic fields H varied from 0 to 50 kOe in a cryostat with a superconducting magnet in liquid helium or in an Oxford Instruments variable-temperature cryostat ($T \geq 8$ K). Photoluminescence was excited by a 632.8 nm He—Ne laser and recorded in the Faraday geometry.

Results and Discussion

Spin-wave resonance and microwave magnetoresistance in $Ge_{1-x}Mn_x$ thin films. $Ge_{1-x}Mn_x$ thin films (Fig. 1) were obtained by ion implantation of manganese ions into single-crystalline germanium wafers. This resulted in the appearance of near-surface germanium layers enriched with manganese ions with an average concentration x of 2, 4, and 8 at.%. The penetration depth of manganese ions into single-crystalline germanium was at most 120 nm and the distribution profile of Mn ions in the manganese-enriched germanium layer was nearly Gaussian. Earlier, it was established that the films contain Ge_3Mn_5 clusters (average diameter of about 10 nm) and isolated Mn^{2+} ions at the sites in the germanium crystal lattice.

The magnetic properties of the $Ge_{1-x}Mn_x$ films depend on two subsystems, *viz.*, Ge_3Mn_5 clusters and individual Mn^{2+} ions dissolved in the Ge matrix.**[5—8]** At $T < 20$ K, the 9.6 GHz electron spin resonance spectra of the $Ge_{1-x}Mn_x$ films exhibited a number of peaks**[9,10]** (Fig. 2). For each line, we calculated the corresponding magnetic susceptibility χ proportional to the integrated intensity of the line. A comparison of the χ values thus obtained with the values for a copper sulfate test sample showed that for all three lines the χ values are two orders of magnitude higher than the magnetic susceptibility values of all Mn^{2+} ions present in the films, calculated assuming noninteracting (paramagnetic) Mn^{2+} ions. This means that the observed magnetic resonance lines cannot be ascribed to paramagnetic Mn^{2+} ions. These lines can be due to ferromagnetic resonance in the Ge_3Mn_5 clusters or ferromagnetic resonance of the entire film in the case of long-range magnetic order. The study of the orientational dependence of these lines at $T = 4$ K by rotating the sample about the axis parallel to the film plane revealed the same anisotropy of the lines. This suggests that the lines characterize a collective spin density excitation in the sample rather than the contributions of particular different-type clusters because in the latter case one would expect differences in the angular dependences of the resonance field for these clusters. Thus, the most plausible reason for the appearance of several resonance peaks with the same angular symmetry of the resonance field is spin-wave resonance.**[9,10]**

Spin-wave resonance indicates a long-range magnetic order in the $Ge_{1-x}Mn_x$ films. At low temperatures, the temperature dependence of the film magnetization $M(T)$ has a smooth (non-Bloch) shape (Fig. 3) characteristic of percolation-type ferromagnetic ordering in disordered fer-

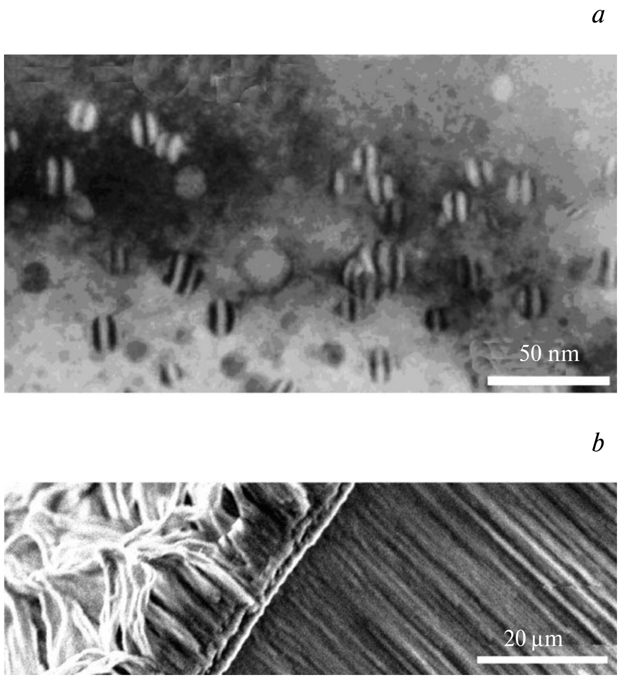

Fig. 1. Micrograph of cross-section of $Ge_{1-x}Mn_x$ thin films with a manganese content $x = 8$ at.% (*a*) and anodized alumina membranes with nanowires (*b*).

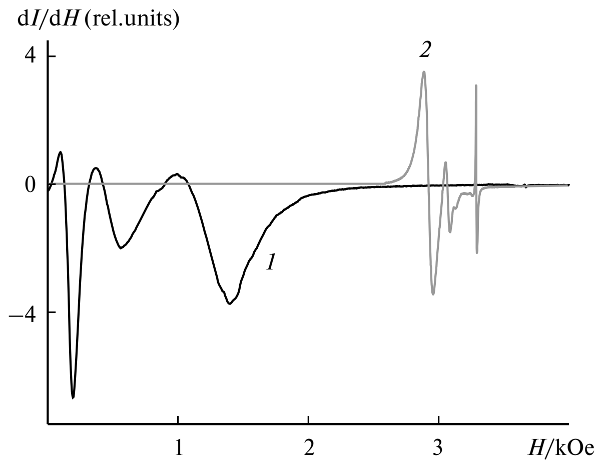

Fig. 2. Electron spin resonance spectra of $Ge_{1-x}Mn_x$ thin films (*1*) and nanowires (*2*) at $T = 4$ K. The constant magnetic field is directed normal to the film plane.

romagnets.[11,12] The onset of ferromagnetism in disordered diluted magnetic semiconductors is usually associated to indirect exchange coupling of transition metal ion impurities through charge carriers. A large spin polarization "cloud" (polaron) is formed near the impurity ion. At relatively high temperatures, the spin orientation of the impurity ions belonging to different polarons are oriented randomly. As the temperature decreases, the radii of magnetic polarons increase and at a particular temperature, the wave functions of the neighboring polarons begin to overlap through impurity ions belonging to different polarons. The overlap of the wave functions leads to ordering of the spins of the impurity ions belonging to different polarons. Near the percolation threshold, when the cluster size of correlated polarons reaches the sample size, a ferromagnetic ordering occurs. The temperature of the percolation-type ferromagnetic transition in the subsystem of dispersed manganese ions can be increased by cooling single-crystalline germanium wafers during ion implantation. Cooling leads to suppression of diffusion-controlled aggregation of manganese ions in the clusters and, as a consequence, to an increase in the concentration of dispersed Mn^{2+} ions and in the Curie temperature of this subsystem.

The magnetoresistance is a key property of thin films of diluted magnetic semiconductors, which determines their practical value. Earlier, only the d.c. magnetoresistance of $Ge_{1-x}Mn_x$ thin films was studied.[13,14] Note that the microwave magnetoresistance can be due to other groups of charge carriers (not those responsible for the d.c. magnetoresistance). But it is these charge carriers manifesting themselves at high frequencies that play the decisive role in high-frequency devices for information storage and recording.

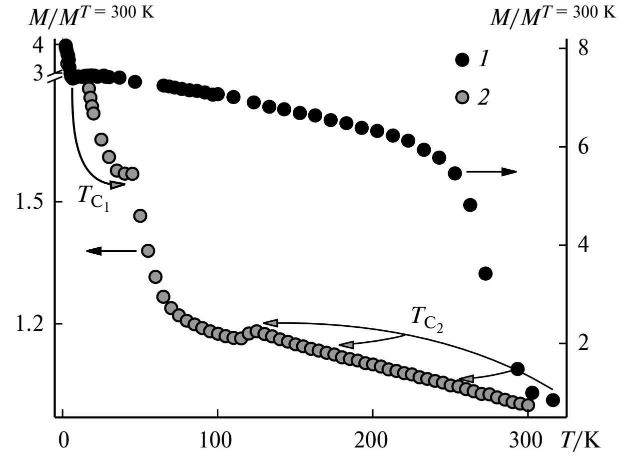

Fig. 3. Temperature dependences of the magnetization of $Ge_{1-x}Mn_x$ thin film (*1*) and nanowires (*2*). The magnetic field of strength $H = 1$ kOe is directed normal to the film plane (parallel to the nanowire axis). The magnetic moment was normalized to its value at $T = 300$ K. Arrows denote the shifts of the Curie temperatures T_{C1} and T_{C2} on going from the thin film to nanowires.

Usually, resonance absorption of microwave power is studied by EPR spectroscopy. However, microwave absorption depends not only on the resonance transfer of the spin energy, but also on the electrical loss in the sample and on the resistance of the sample in the magnetic field. For low-ohmic samples characterized by the microwave resistance $\rho \approx 10^{-3}$ Ohm (*cf.* about 250 Ohm for the resistance of the microwave circuit of the spectrometer), the absorbed power is proportional to the resistance of the sample.

At $T > 60$ K, the signal of microwave absorption $dP/dH(H)$ is significantly different from that observed at low temperatures (see Fig. 2) and represents a strongly nonlinear, very broad background whose amplitude much exceeds the signal of the spectrometer in blank experiments.[9,10] It was found that the observed signal has a nonresonance nature. The reason for the appearance of the broad signal is the magnetoresistance of the sample. Note that the undoped (manganese-free) germanium sample shows no broad signals at the same sensitivity of the spectrometer, which is determined from the signal amplitude of the reference sample. An analysis of the nonmonotonic magnetic field dependence of the microwave magnetoresistance revealed three components, *viz.*, a positive classical Lorenz magnetoresistance, a negative magnetoresistance due to the Zeeman splitting of localized states near the Fermi level, and a component originating from spin-dependent scattering of charge carriers by impurities.[9,10]

Spin dynamics in $Ge_{1-x}Mn_x$ nanowires. To compare the magnetic properties of the quasi-two-dimensional thin films and quasi-one-dimensional nanowires of magnetic semiconductors of the same composition and to reveal the effect of reduced dimension on the magnetic properties, samples of $Ge_{1-x}Mn_x$ nanowires were investigated (see Fig. 1). Nanowires 60 nm in diameter and 100 μm long were synthesized using the supercritical fluid inclusion-phase technique in pores of anodized alumina membranes as a result of decomposition of diphenyl germanite and dicarbonylmanganese.[15]

A comparison of the magnetoresistance of the $Ge_{1-x}Mn_x$ films and nanowires demonstrates the effect of reduced dimension on the spin-dependent scattering of charge carriers in the $Ge_{1-x}Mn_x$ system and suppression of the microwave magnetoresistance on going to the quasi-one-dimensional system.[16—18]

Spin-wave resonance in thin films also differs from the resonance of standing spin waves in nanowires.[16—18] This is due to limitation imposed on the transversal dimension of the nanowire. When the nanowire diameter becomes smaller then the magnetic correlation length, "one-dimensional" spin-wave resonance can be excited in the nanowires. For the $Ge_{1-x}Mn_x$ nanowires, the magnetic correlation length much exceeds the nanowire diameter (about 250 *vs.* 60 nm, respectively). Therefore, the resonance lines from the $Ge_{1-x}Mn_x$ nanowires can be interpreted as manifestation of "one-dimensional" spin-wave resonance in which the distribution regularities of the resonance spectral lines differ from the case of thin films.

The results of preliminary experiments showed that limitations imposed on the size and dimension of the samples can affect the temperature of ferromagnetic ordering in $Ge_{1-x}Mn_x$ (see Ref. 19). For example, "bulk" $Ge_{1-x}Mn_x$ samples ($x = 1$—17 at.%) are characterized by two critical temperatures of about 150 K and 285 K corresponding to the antiferromagnetic and ferromagnetic ordering in different Ge_nMn_m clusters (Ge_3Mn_5, Ge_8Mn_{11}, *etc.*), respectively.[20] The $Ge_{1-x}Mn_x$ thin films (x = 8 at.%) are characterized by two reliably reproducible critical temperatures $T_{C_1} \approx 16$ K and $T_{C_2} \approx 290$ K which correspond to ferromagnetic ordering in dispersed Mn^{2+} ions and in Ge_3Mn_5 clusters, respectively. It was also reported[21] that ferromagnetic ordering in $Ge_{1-x}Mn_x$ nanocolumns (x = 1—6 at.%) can be observed up to 400 K, which is higher than the Curie temperatures of all known types of Ge_nMn_m clusters. The quantum dots $Ge_{1-x}Mn_x$ (x = 1—5 at.%) are characterized by two critical temperatures of about 230 and 300 K.[22] Of particular interest is the increase in the critical temperature T_{C_1} corresponding to percolation-type long-range ordering in the subsystem of dispersed Mn^{2+} ions. In this study, we established the role of the dimension of diluted magnetic semiconductors in the formation of magnetic properties and the temperature of percolation transition to the ferromagnetic state in the $Ge_{1-x}Mn_x$ single-crystalline thin films and nanowires. Nanostructuring of $Ge_{1-x}Mn_x$ single-crystalline alloy leads to an increase in the Curie temperature of the percolation transition (see Fig. 3). The increase in T_{C_1} on going from thin films to nanowires can be associated with an increase in the solubility and/or nonuniform distribution of manganese ions within the nanowire due to the presence of regions with locally high concentration of dispersed manganese ions in the nanowires. Transition from thin films to nanowires is characterized by a long flat portion on the $M(T)$ graph instead of a step-like dependence observed for thin films (see Fig. 3). This can be due to broadening of the scatter of cluster sizes and, correspondingly, Curie temperatures. It is known that the Curie temperatures of nanoparticles depend on the particle size because there is a deficiency of exchange bonds in the near-surface atoms of the nanoparticles. This reduces the Curie temperatures in them compared to those of the "bulk" samples.

Effect of ferromagnetic ordering in δ-<Mn>-layer on the quantum well photoluminescence in InGaAs/GaAs heterostructures. In the text above, we considered magnetic semiconductors based on germanium whose great advantage is high mobility of charge carriers. Germanium is an indirect band gap semiconductor which is difficult-to-use in optoelectronics. Therefore, it was interesting to study a diluted magnetic semiconductor based on GaAs. Magnetic quasi-two-dimensional (2D) heterostructures based on $Ga_{1-x}Mn_xAs$ deserve particular attention from the standpoint of practical applications. For instance, by δ-doping (forming a thin dopant layer a few nanometers thick in the bulk of a lightly doped semiconductor) a heterojunction with Mn atoms the Curie temperatures T_C can be increased up to 250 K due to the overlap of the wave functions of high concentration of 2D holes with those of the atoms of the δ-Mn layer.[23] However, implantation of Mn directly into the 2D channel leads to low hole mobility (≤ 10 cm^2 V^{-1} s^{-1}) and to suppression of radiative recombination at $x > 0.1$ at.% (see Ref. 24). Therefore, enhancement of magnetic properties is accompanied by weakening of the electrical and optical responses of the system to magnetization. As a result, the main goal to combine interrelated magnetic, electrical and optical properties is not attained. This difficulty can be avoided by spatially separating charge carriers and the Mn δ-layer. This increases the mobility of charge carriers in the 2D channel by more than two orders of magnitude[25] and retains the sensitivity of the electrical and optical properties to magnetization because the characteristic path length of charge carriers with conservation of spin polarization appears to be longer than the distance between the δ-<Mn>-layer and the quantum well.

In molecular epitaxy, the substrate orientation significantly affects the quality and ordering of growing nanostructures.[26,27] For instance, when the angle θ between a crystallographic plane and the corresponding substrate face differs from zero, various types of fragmentary, irregularly filled nanolayers can be grown; this significantly affects the optical and electron-transport properties of samples.[26,27] The reasons for the substrate orientation effect remain unclear. Therefore, we tried to disclose the driving force of the changes in the optical properties of the InGaAs/GaAs quantum well at different orientations of GaAs and to establish how does it affect the magnetic properties of a thin layer of a δ-doped $Ga_{1-x}Mn_xAs$ solid solution with a high concentration of manganese $x \approx 4$—10 at.% (δ-<Mn>-layer) and their interrelations with the optical properties of the InGaAs/GaAs quantum well.

InGaAs/GaAs/δ-<Mn> heterostructures were grown by combined stepwise epitaxial growth. First, a GaAs buffer layer, a δ-<C> layer, an undoped GaAs layer, an undoped InGaAs quantum well and, finally, a GaAs separating layer (Fig. 4) were successively grown by organometallic hydride epitaxy on a GaAs (001) substrate. Next, a δ-<Mn> layer and a GaAs cover layer were successively grown in the same reactor by laser sputtering of Mn and GaAs targets. The δ-<Mn> layer represented a thin layer (about 2—3 nm thick) of a $Ga_{1-x}Mn_xAs$ solid solution with high concentration of manganese ($x \approx 4$—10 at.%).[25] The procedure for preparation and certification of InGaAs/GaAs/δ-<Mn> heterostructures was described in more detail elsewhere.[28]

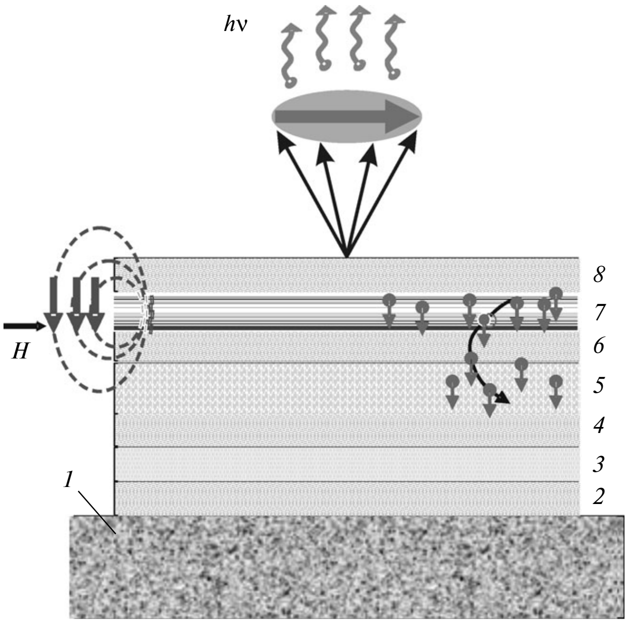

Fig. 4. A heterostructure: a schematic view. Shown is the magnetic field *H* of the δ-<Mn>-layer with spin-polarized holes and the polarized photoluminescence *h*ν from the InGaAs quantum well. The order of layers: GaAs substrate 0.5 mm thick (*1*), GaAs buffer layer 0.5 μm thick (*2*), δ-<C>-layer 1 mm thick (*3*), GaAs separating layer 15 nm thick (*4*), InGaAs quantum well 10 nm thick (*5*), GaAs separating layer 3 nm thick (*6*), δ-<Mn>-layer 2 nm thick (*7*), and GaAs cover layer 30 nm thick (*8*).

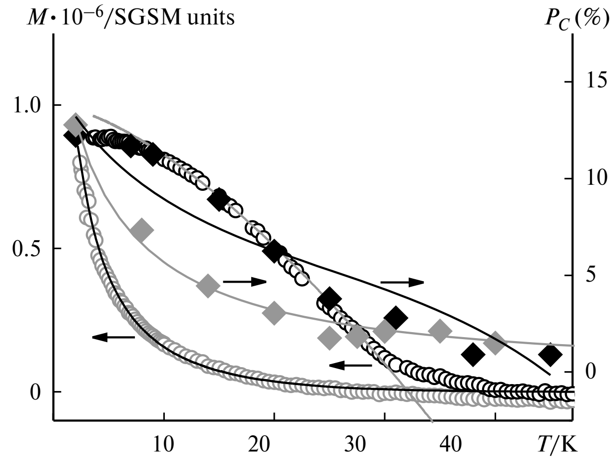

Fig. 5. Temperature dependences of the magnetization M (open circles) and degree of circular polarization P_C (rhombi) for the sample Nos 4838 (singular) (black rhombi) and 4831 (vicinal) (grey rhombi). Solid lines denote the theoretical dependences.

In this work, two series of samples were investigated. The first series included singular heterostructures with a InGaAs/GaAs quantum well and δ-<Mn>-layer on exactly oriented GaAs (001) substrate, *viz.*, the angle θ between the (001) crystallographic plane and the corresponding substrate face was equal to zero. The second series of samples included vicinal heterostructures containing a InGaAs/GaAs quantum well and a δ-<Mn> layer on the GaAs (001) substrate; the angle θ = 3° between the growth direction and the [001] crystallographic direction. The substrate orientation was controlled by X-ray diffraction using a DRON-4 diffractometer.

In this work, we studied three singular samples for which qualitatively similar results were obtained. We will discuss them taking the sample 4838 as an example. The temperature dependence of its magnetization, $M(T)$, measured with the SQUID magnetometer is shown in Fig. 5. As for conventional bulk ferromagnets, the decrease in the magnetization of the δ-<Mn>-layer with an increase in temperature is correctly described by the Bloch expression (see Fig. 5) in a wide temperature range below the Curie temperature $T_C = 30 \pm 5$ K.

The shape of the $M(T)$ dependence for the vicinal structures significantly differs from the smooth Bloch pattern, being characteristic of percolation-type ferromagnetic ordering in disordered ferromagnets.[11,29] The $M(T)$ dependences obtained for highly disordered ferromagnets $Ga_{1-x}Mn_xAs$ with no quantum well also exhibit a characteristic extended flat portion near T_C similarly to the vicinal sample 4831 (see Ref. 30). The onset of ferromagnetism in disordered diluted magnetic semiconductors including $Ga_{1-x}Mn_xAs$ is due to indirect exchange coupling of transition metal impurity ions through charge carriers (holes) analogously to the case for $Ge_{1-x}Mn_x$ (see above).

The photoluminescence spectra of the InGaAs/GaAs/δ-<Mn> samples below 1.4 eV correspond to radiative recombination of photoexcited electrons relaxed to the quantum well and holes in the quantum well (the holes are in the well owing to doping with manganese).[31,32] In the magnetic field, the emission line of the quantum well is split into two circularly polarized components due to the Zeeman splitting of the electron and hole states. High degrees of circular polarization $P_C = (I_+ - I_-)/(I_+ + I_-)$ of photoluminescence of the InGaAs/GaAs quantum well (here I_+ (I_-) are the intensities of the components with right (left) polarization) and spin polarization of holes in the quantum well are due to the magnetization of adjacent δ-<Mn>-layer rather than external magnetic field.[32] The photoluminescence intensity of the quantum well in the structures containing the δ-<Mn>-layer is more than 100 times weaker than in the "nonmagnetic" structure (sample 4847) having no δ-<Mn>-layer. Therefore, the "infinitely thin" δ-<Mn>-layer creates prerequisites for domination of nonradiative recombination.

The temperature dependences of P_C in the singular and vicinal structures are significantly different. They are similar to the corresponding $M(T)$ dependences for the same samples (see Fig. 5). We believe this similarity is not accidental and can generally be understood within the framework of the polaron percolation theory. The degree of polarization should be proportional to the magnetic field produced by the δ-<Mn>-layer and, therefore, to the magnetization of this layer. On the other hand, it is the ferromagnetic regions that contribute to high P_C values because $P_C \approx 0$ in the nonmagnetic structures. Thus, one can expect that the function $P_C(T)$ will be proportional to $M(T)$ for a given sample. These simple considerations are indeed confirmed by the experimental data. In Fig. 5, the solid lines denote the temperature dependences of P_C calculated based on the considerations mentioned above. The recalculated $P_C(T)$ dependence is in good agreement with the initial experimental data for the sample 4831.

Summing up, in the present work we studied the static and microwave dynamic magnetic properties of the nanocomposites containing arrays of ordered quasi-one-dimensional $Ge_{1-x}Mn_x$ ferromagnetic semiconductor nanowires in anodized alumina membranes and quasi-two-dimensional thin films of the same composition. A comparison of the microwave magnetoresistance, electron spin resonance spectra, and static magnetization data for the $Ge_{1-x}Mn_x$ thin films and nanowires indicates that the sample size affects the spin-dependent scattering of charge carriers, the Curie temperature, and the spin dynamics in the $Ge_{1-x}Mn_x$ nanostructures. Oriented $Ge_{1-x}Mn_x$ nanowires with readily controllable and reliably reproducible geometries can be prototypes of memory microchips. The effect of the magnetic field of the δ-<Mn>-layer on the polarization of quantum well photoluminescence in InGaAs/GaAs/δ-<Mn> heterostructures was revealed. The heterostructures studied can be considered as prototypes of spin light-emitting diodes.

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