

Scientific session of the Division of General Physics and Astronomy of the Russian Academy of Sciences (27 March 2002)

A scientific session of the Division of General Physics and Astronomy of the Russian Academy of Sciences (RAS) was held on March 27, 2002 at the P N Lebedev Physics Institute, RAS. The following reports were presented at the session.

(1) **Merkulov I A** (A F Ioffe Physicotechnical Institute, RAS, St. Petersburg, Russia) “Spin systems of quantum dots”;

(2) **Vedyaev A V** (Physics Department of M V Lomonosov Moscow State University, Moscow, Russia) “Use of spin-polarized current in spintronics”;

(3) **Ivchenko E L** (A F Ioffe Physicotechnical Institute, RAS, St. Petersburg, Russia) “Circular photogalvanic effect in nanostructures”;

(4) **Maksimov A A** (Institute of Solid-State Physics, RAS, Chernogolovka, Moscow Region, Russia) “Optical spectroscopy of semiconductor quantum dots”.

An abridged version of the first three reports is given below.

PACS numbers: 73.21.La, 78.67.Hc
DOI: 10.1070/PU2002v045n12ABEH001327

Spin systems of quantum dots

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(1) The magnetic properties of solids were used long before the physical models of solids were developed [1]. The understanding of the essence of the processes determining magnetic phenomena and their relation to electron spin became possible only with the development of quantum mechanics [2]. At present there can be no doubt about the leading role of spin processes in various phenomena of physics, chemistry, and other natural sciences (e.g., see Refs [3, 4]).

Spin phenomena in semiconductors have been studied for several decades. Optical orientation of electron spins, their depolarization in a magnetic field, and the alignment of the hot photocarrier momenta as a result of absorption of linearly polarized light have made it possible to study rapid relaxation processes in steady-state conditions and to build up the sources of polarized electrons [5].

In the last decade, the focus of investigations has shifted to spin phenomena in quantum-dimensional heterostructures, including quantum dots. Despite distinctions in the materials and the technologies used in fabricating these ‘artificial atoms’ [6–9], the main mechanisms governing the behavior of their spin systems are in many respects close and can be

examined within the approaches developed for the description of paramagnetic centers [3]. At present these systems generate a great deal of interest as possible elements of spintronic devices (for storing and processing data, and for quantum computations [10]).

(2) The main components of a spin system of a quantum dot are the spins of one or several carriers localized in it and a macroscopic number of nuclei forming the quantum dot of ions. For semimagnetic semiconductors, the spins of electrons filling the d and f shells of the ions of the transition or rare-earth metals are also added to the system.

Usually the description of the interaction between the spins in the quantum dot is restricted to pair Hamiltonians. First, this is the exchange interaction between the spins of charge carriers [11] and of carriers and electrons localized on the d and f shells of magnetic ions [12, 13], and the contact hyperfine interaction between the spins of the electrons and nuclei of the crystal lattice [14] (the Hamiltonians are directly proportional to scalar products of the spins). Second, the long-range (electrodipole) exchange electron–hole interaction [15–17] and the magnetodipole interaction between nuclear spins [14] also show their worth. The magnetodipole interaction between the spins of carriers and nuclei can usually be ignored.

(3) Spin–orbit coupling in the conduction band of diamondlike semiconductors is usually weak and can be interpreted as a perturbation. For the same reason, the interaction between electron spins and lattice vibrations is considered weak [18]. For electrons, the spin–orbit coupling begins to play an important role only if their kinetic energy is comparable to the band gap. In wide-gap semiconductors this is achieved only in the structures whose size approaches several lattice constants [19]. (In narrow-gap semiconductors, the requirements of the smallness of the localization region are not so stringent.) In these conditions, the magnitude and direction of the average electron spin differs from point to point [20, 21]. Such a dependence may lead to the formation of nonmagnetic (antiferromagnetic) magnetic polarons whose total magnetic moment is zero, while at each point the spins of the electrons and the surrounding magnetic ions are nonzero and correlated [21].

In diamondlike semiconductors, the hole spin $J = 3/2$, the effective mass depends on the helicity, and the spin–orbit coupling is described by the Luttinger Hamiltonian [11]. Generally speaking, irrespective of the size of the localization region, not one of the J components is a ‘good’ quantum number [22] and the quantum-mechanical average $\langle \mathbf{J} \rangle$ changes from point to point [23].

(4) The role of short-range and long-range exchange interactions between an electron and a hole, both localized at a quantum dot, has been thoroughly studied in Refs [16, 17, 24–27]. The strongest short-range exchange interaction splits the multiply degenerate ground state of the electron–hole

pair into two multiplets, namely, the ‘bright’ exciton (which interacts with photons) and the ‘dark’ exciton (which does not interact with photons). Because of the long-range exchange interaction, for quantum dots of anisotropic shape in a zero magnetic field, these multiplets are split into singlet non-magnetic states. As a result, the spin polarization of the electron–hole pair, which is introduced in a particular manner, disappears completely over long enough time intervals.

The luminescence from such states is linearly polarized. The mixing of the states in the presence of an external magnetic field (van Vleck paramagnetism) leads to circular polarization of equilibrium luminescence. In Faraday geometry, where the exciting light and the detected light propagate along the external magnetic field, the conversion effect emerges. Excitation by circularly polarized light gives rise to linear polarization of the luminescence, and conversely linear polarization of the exciting light gives rise to circular polarization of the recombination radiation. The data from optical experiments make it possible to estimate the shape anisotropy of quantum dots.

In conditions of pulsed excitation by circularly polarized light, when the recombination luminescence is caused by the coherent superposition of the transitions from the initial states with different energies Δ , polarization beats at the frequency Δ/\hbar should be observed in the luminescence. Such beats were indeed detected in the quantum dot luminescence spectra recorded in the Voigt geometry by the temporal resolution technique [28]. In the experiments of Kalevich et al. [28], the exchange interaction did not manifest itself, and the beat frequency equaled the Larmor precession frequency of the electron spin in the magnetic field. Apparently, this fact is related to the rapid spin relaxation of the hole, which leads to suppression of the exchange interaction because of the dynamic averaging effect.

(5) When describing spin relaxation of an electron or hole localized at a quantum dot, it is convenient to apply the approaches developed in the theory of paramagnetic resonance [3]. For an electron, Khaetskii and Nazarov [18] and Erlingsson et al. [29] examined the relaxation related to the interaction with acoustic phonons. Because of the weakness of the spin–orbit coupling in the conduction band, this process is characterized by long times ($\tau_S \geq 10^{-5}$ s), while for a hole τ_I may amount to several fractions of a nanosecond [17]. The hyperfine interaction has practically no effect on hole polarization, while for electrons and nuclei of a quantum dot this interaction may play the leading role [30–32]. At low values of the nuclear spin temperature, which may be reached via the optical cooling method, the hyperfine interaction leads to the formation of a nuclear spin polaron and the relaxation’s hyperfine channel is suppressed [33].

(6) In strong magnetic fields $B \gg B_L$ (B_L is the local field with which the neighbors act on the specified nuclear spin¹), nonequilibrium polarization of the electrons localized at the quantum dot is transferred to the nuclei in the process of flip–flop transitions in exactly the same way as happens in bulk samples [34–36]:

$$\left. \frac{\partial \langle \mathbf{I} \rangle \mathbf{B}}{\partial t} \right|_{f-f} = \frac{1}{T_e} \left[Q \langle \mathbf{S} \rangle \mathbf{B} - \langle \mathbf{I} \rangle \mathbf{B} \right]. \quad (1)$$

¹ For GaAs, the magnitude of B_L induced by the magnetodipole interaction is approximately 1.5 G.

Here, the reciprocal time T_e^{-1} is on the order of the product of the square of the precession frequency (ω) of nuclear spin in the hyperfine field of an electron by the time (τ_c) of coherent motion of the electron and nuclear spins in the hyperfine fields they generate on each other, and

$$Q = \frac{\langle I^2 \rangle B^2 - \langle (\mathbf{I} \mathbf{B})^2 \rangle}{\langle S^2 \rangle B^2 - \langle (\mathbf{S} \mathbf{B})^2 \rangle} \approx 5.$$

In steady-state conditions, one finds $\langle I \rangle \approx Q \langle S \rangle$.

When the magnetic fields are weak ($B \leq B_L$), the interaction between the spins of the neighboring nuclei gives rise to rapid relaxation of all the nuclear polarization components [35], and then

$$\langle \mathbf{I} \rangle \approx Q \frac{(\mathbf{S} \mathbf{B}) \mathbf{B}}{B^2 + \xi B_L^2}, \quad (2)$$

where ξ is a number of order unity. The characteristic drop in nuclear polarization in weak magnetic fields has been observed by many researchers (e.g., see Ref. [37]).

The developed theory of dynamic polarization can be applied directly to describe the situation where only one electron resides in the quantum dot. However, the case of quantum dots containing electron–hole pairs requires additional analysis. The nonmagnetic character of the steady states and the strong van Vleck paramagnetism that appears both in an external magnetic field and in the average hyperfine field of polarized nuclei lead to quantitative and even qualitative features.

In anisotropic quantum dots whose thickness is much smaller than their length and width, the relaxation time of nuclear polarization on electrons increases substantially [38]:

$$T_e \rightarrow T_e' \approx T_e \left(\frac{\delta_0}{\hbar} \right)^2 \tau_b \tau_d,$$

where δ_0 is the spacing between the doublets, and τ_b and τ_d are the lifetimes of bright and dark excitons. In the process of flip–flop transitions, the total energy of the electron–nuclear spin system changes by a magnitude practically equal to δ_0 (Fig. 1). Hence, the nuclear polarization by the oriented electrons may only occur against the background of other processes related to the absorption or emission of large energy packets. For instance, a flip–flop transition converts a dark exciton into a bright one, which immediately recombines and emits a photon with the appropriate energy.

Because of the slowing-down of dynamic polarization, the region of ‘small’ values of the external magnetic field, in which the average spin of dynamically polarized nuclei is negligible, increases substantially. As shown by Gammon et al. [38], the nuclear polarization is described in this case, too, by equation (2) in which, however, $\xi \rightarrow \tilde{\xi} \approx 10^5$. It occurs that dynamic polarization is mainly related to the nonequilibrium spin of electrons in long-lived states of the dark exciton.

In their theoretical model, Gammon et al. [38] did not allow for the effect of the level splitting of the bright and dark excitons in the hyperfine field of polarized nuclei on the dynamic polarization rate. As can easily be understood from Fig. 1, if we allow for this effect, the equation for the rate of nuclear spin relaxation becomes nonlinear:

$$\frac{d \langle I \rangle}{dt} = -\frac{1}{T_e'} \left[\frac{n_1 (\langle I \rangle - Q/2)}{(1 - aI)^2 + \gamma^2} + \frac{n_1 (\langle I \rangle + Q/2)}{(1 + aI)^2 + \gamma^2} + \langle I \rangle \frac{\tilde{\xi} B_L^2}{B^2} \right]. \quad (3)$$

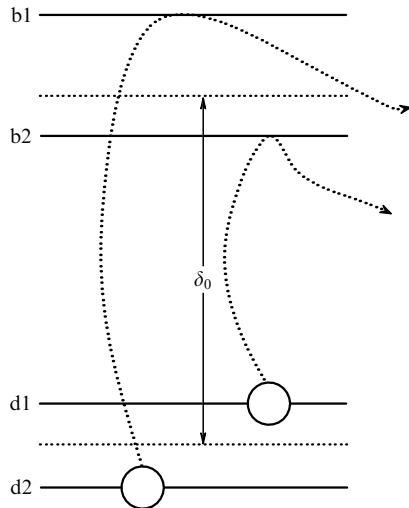


Figure 1. Spin sublevels of a bright exciton ($b1 = |1/2e, -3/2h\rangle$ and $b2 = |-1/2e, 3/2h\rangle$) and a dark exciton ($d1 = |1/2e, 3/2h\rangle$ and $d2 = |-1/2e, -3/2h\rangle$) in a thin cylindrical quantum dot, which are split by the exchange interaction (δ_0) and by the hyperfine nuclear field. The dotted curves represent the radiative recombination (allowed by flip–flop transitions) of dark excitons. The exciton level splitting in the hyperfine field of the polarized nuclei changes the spacing between the pairs of spin sublevels responsible for the opposite directions of dynamic polarization.

Here, n_{\uparrow} and n_{\downarrow} are the populations of the levels of the dark exciton with the electron spin parallel and antiparallel to the Z -axis, a is the hyperfine nuclear field parameter ($\mathbf{B}_N = a\langle\mathbf{I}\rangle$), and γ is the half-sum of the broadenings of the energy levels of the bright and dark excitons, measured in units of δ_0 . [Without the relaxation term $-\langle I \rangle \xi B_L^2 / T_e B^2$ [38], equation (3) for the nuclear polarization at a quantum dot was derived by Korenev [39].] Analysis of the steady-state solutions (Fig. 2) to the above equation predicts the occurrence of bistability and dynamic self-polarization [39, 40].

This work was supported financially by grants from the Russian Foundation for Basic Research (00-12-16991) and CRDF (RP1-2252).

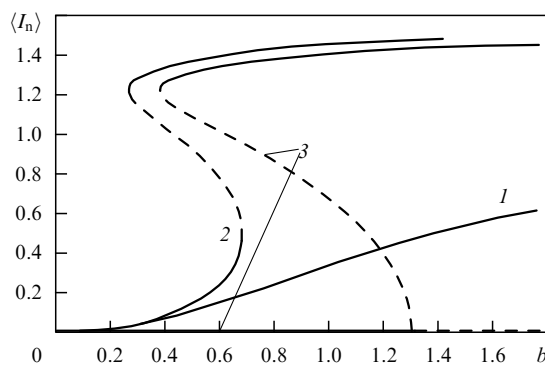


Figure 2. Polarization of the nuclear spin system of a GaAs quantum dot as a function of the external magnetic field $b = (B^2/\xi B_L^2)^{1/2}$ for the cases of weak, $a = 0.1$ (1), and strong, $a = 0.8$ (2, and 3), feedback via the hyperfine nuclear field. The curves 1 and 2 have been constructed for the maximum value of the average electron spin $\langle S_z \rangle = 1/2$, and 3 for $\langle S_z \rangle = 0$. The case of a weak feedback $\langle S_z \rangle = 0$ corresponds to a single trivial solution $\langle I_z \rangle = 0$. For the case of strong feedback there appears an additional solution corresponding to self-polarization of the nuclei. The unstable steady states are shown by dashed lines.

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PACS numbers: 73.40.Gk, 73.40.Rw, **75.70.-i**, 85.30.Mn

DOI: 10.1070/PU2002v045n12ABEH001270

Use of spin-polarized current in spintronics

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Spintronics is a relatively new branch of conventional electronics, where the electron spin (together with the electron charge) is an active element in storing and conveying information [1]. Devices that utilize electron spin may displace or supplement to a large measure traditional electronic devices. More than that, spintronics has real potential for use in new areas of technology, for example, in quantum calculations and quantum data transmission [2].

Nature has supplied us with a natural source of spin-polarized electrons in the form of ferromagnetic metals of the 3d group: Fe, Ni, and Co and their alloys. By now the origin of the ferromagnetic state of these metals has been established with rather great accuracy. For instance, there is no doubt that spontaneous magnetization arises from the part of the electron–electron Coulomb interaction that changes as a result of the permutation of two identical Fermi particles (electrons) and is called the exchange energy E^{exch} . This energy is lowest if the electron spins in the metal are parallel. True, in some cases the energy minimum is realized in a more complicated configuration of spins, for example, in a helical spin-density wave. On the other hand, the kinetic energy of an electron in the metal together with the Pauli exclusion principle tend to disrupt the spin order. Detailed first-principles calculations of the band structure of almost all the metals from the Periodic Table have shown that for 3d-metals (Fe, Ni, and Co) the electron density of states $\rho(\varepsilon_F)$ at the Fermi level is high enough for the so-called Stoner criterion $E^{\text{exch}}\rho(\varepsilon_F) > 1$ to be met. It is the emergence of spontaneous magnetization that is ensured by meeting this criterion, i.e., the population of subbands with different directions of spin is nonequivalent and the system acquires spontaneous magnetization. Notice that the main contribution to magnetization is provided by d-symmetry electrons whose density of states at the Fermi level is much higher than that of sp-symmetry electrons.

If, however, we turn to electron transfer phenomena, we are forced to conclude that the main contribution to the current in ferromagnetic metals is provided by the mobile sp-electrons rather than the heavier but strongly magnetized d-electrons. But is the current in a ferromagnetic d-metal spin-polarized? A direct answer to this question was obtained in 1988 as a result of a discovery [3] of what is known as the giant magnetoresistance (GMR) effect. Generally, the magnetoresistance phenomenon, or the change in the electrical resistance of a nonmagnetic or ferromagnetic metal on imposition of an external magnetic field, has been known for a long time. For ordinary samples this change is relatively small, however, amounting up to fractions of a percent for nonmagnetic metals and about 1% for ferromagnetic metals. On the other hand, GMR may be

as high as 100% at low temperatures. Let us describe this phenomenon in greater detail.

GMR was first observed in a multilayer thin-film $[\text{Fe}/\text{Cr}]_n$ structure, where Fe is a thin (several angstroms) layer of iron, Cr is a layer of chromium, and n is the number of repetitions of such a bilayer. What is important here is that the chromium layer is approximately 12 Å thick. Experiments provided an explanation for this fact — as the chromium layer thickness varies, the mutual orientation of the magnetizations peculiar to the adjacent iron layers changes from parallel to antiparallel, and it becomes almost strictly antiparallel when the Cr layer is 12 Å thick. According to theoretical investigations into this phenomenon, the magnetizations of the adjacent iron layers are coupled by the indirect exchange interaction via the conduction electrons in chromium. This interaction oscillates in space, changing its sign with a period determined by the Fermi-electron momentum in chromium and equal to 12 Å in the case of chromium. Thus, the magnetizations of the adjacent layers proved to be coupled antiferromagnetically (antiparallel) or ferromagnetically (parallel), depending on the thickness of the Cr interlayer. Now, if in the absence of an external magnetic field the magnetizations of the adjacent iron layers are antiparallel, a strong enough magnetic field (about 20 kOe in the case at hand) aligns these magnetizations parallel to each other and to the field direction. Measuring the electrical resistance R of the system with the current flowing in the plane of the layers (CIP) for the parallel (R^P) and antiparallel (R^{AP}) magnetization orientations, Schad et al. [4] found that the resistance R changes in such a way that $(R^{AP} - R^P)/R^P \approx 1.2$ at 1.5 K for the $[[\text{Fe}(4.5 \text{ \AA})/\text{Cr}(12 \text{ \AA})]_{50}]$ system. An even greater effect has been observed for analogous systems with the current flowing perpendicularly to the plane of the layers. Note that a similar effect has been observed in much weaker external fields (~ 20 Oe) for sandwich structures of the $F_1|P|F_2$ type, where F_1 and F_2 are the thin layers of ferromagnetic metals with different coercitivities (e.g., Permalloy and cobalt), and P is a nonmagnetic metal (e.g., copper). In this case, initially the magnetizations of the ferromagnetic layers are parallel, and an external magnetic field stronger than the smaller coercitive field of one layer (Permalloy) and weaker than the greater coercitive field of the other layer, when applied to the system, reverses the magnetism of the layer with the smaller coercitivity, so that the magnetization of that layer aligns itself antiparallel to the magnetization of the other (ferromagnetic) layer with the greater coercitivity. In other words, we are again in the situation we have just described and, measuring the resistance of the given system, we will again find that the resistance changes significantly when an external magnetic field is applied to the system, but the maximum effect is achieved in the fields of about 20 Oe, which are much weaker than in (Fe/Cr) multilayer structures.

Several researchers have provided theoretical interpretations of the GMR effect. These were based on the quasi-classical approaches using the Boltzmann equation [5–9] or on Kubo's quantum-mechanical formalism [10–12]. In this short report there is no sense discussing the ways in which the basic formulae describing the spin-polarized transport in the above structures were derived. Instead we will examine the GMR effect qualitatively.

As noted earlier, a ferromagnetic metal contains at least two groups of electrons: heavy d-electrons which practically do not participate in electron transport but whose band is