ODMR evidence of electronic cascade in multiple asymmetrical (CdMn)Te quantum wells

A. S. Gurin, D. O. Tolmachev*, N. G. Romanov*[1)], B. R. Namozov*, P. G. Baranov*, Yu. G. Kusrayev*, G. Karczewski*[×]

**Ioffe Institute, 194021 St. Petersburg, Russia*

[×]*Institute of Physics, Polish Academy of Sciences, PL-02608 Warsaw, Poland*

Submitted 29 May 2015
Resubmitted 7 July 2015

Exchange-coupled complexes consisting of Mn ions and holes were revealed by optically detected magnetic resonance in the narrowest quantum wells of asymmetrical multiple-quantum-well structures (CdMn)Te/(CdMg)Te. Calculations were performed to estimate the parameters of the complexes (exchange interactions and hole g-factors) and simulate the spectra. The formation of such complexes implies the directional electron tunneling from narrow to larger wells, i.e. an electronic cascade, which results in the creation of the excess hole concentration in the narrowest and intermediate-width quantum wells.

DOI: 10.7868/S0370274X15160080

In diluted magnetic semiconductors (DMS), strong interaction between the two spin subsystems – free electrons and localized spins of impurity transition ions is known to give rise to striking effects, e.g., the giant Zeeman splitting of both the conduction and valence bands [1]. Since the magneto-optical properties of DMS are determined by magnetic impurity ions (Mn in most cases) it is important to clarify the structure of Mn-related centers. Electron paramagnetic resonance (EPR) is a method of choice for the study of transition metal ions [2]. However, conventional radiospectroscopy techniques are hardly applicable for low-dimensional systems because of a small active volume, not high enough sensitivity and the absence of spatial selectivity. High sensitivity, extreme resolution and spatial selectivity of optically detected magnetic resonance (ODMR) allow to overcome these difficulties and make this technique very suitable for a study of defects, carriers and excitons in quantum wells (QWs), superlattices (SLs), quantum dots (QDs) and nanocrystals [3].

Using ODMR the giant exchange interaction of free carriers with the localized spins of magnetic ions was discovered [4]. Later, ODMR was used for investigation of bulk DMS (ZnMn)S [5], (ZnMn)Te [6], (CdMn)Te [7, 8] and DMS-based nanostructures (see [9, 10] and references therein) but no structural information on Mn-related centers was reported. Anisotropy of ODMR spectra found in (CdMn)Se QDs [11] and sub-monolayer QWs [12] allowed to reveal a strain-induced axial fine structure splitting of the Mn^{2+} energy levels that appears because of low dimensionality.

It was shown [13] that manipulating the density of the two-dimensional hole liquid affects the ferromagnetic properties of magnetic quantum wells and drive the system between the ferromagnetic and paramagnetic phases, in a direction which can be selected by an appropriate design of the structure. This offers new tools for patterning magnetic nanostructures as well as for information writing and processing. Excess hole concentration can be created in not specifically doped QWs owing to the surface states [14]. In such (CdMn)Te quantum wells with 2D hole gas, unusual behavior of Mn spin system was observed in spin-flip Raman-scattering experiments [15] where a reduction of the effective Mn g-factor was found. The effect was ascribed to magnetic soft mode [16] of collective excitations in strongly coupled spin system. New anisotropic ODMR spectra in (CdMn)Te quantum wells containing 2D hole gas [17] were ascribed to the complexes consisting of manganese ions exchange-coupled to localized holes.

In this work, we report on peculiarities of ODMR spectra in a different type of systems, i.e. asymmetrical QW structures containing (CdMn)Te QWs with different width separated by large (CdMg)Te barriers. The evidence of the excess hole concentration that appears because of the cascade charge transfer between QWs is presented.

A schematic of the (CdMn)Te/(CdMg)Te heterostructure under study is shown in the inset in Fig. 1. The structure was grown by molecular-beam epitaxy on a (001)-oriented GaAs substrate with a thick (100 nm)

1)e-mail: nikolai.romanov@mail.ioffe.ru

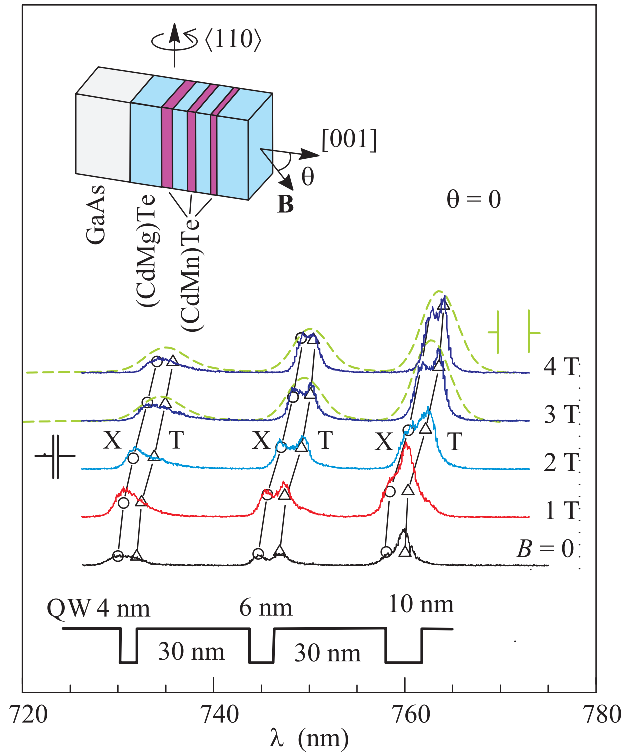

Fig. 1. The scheme of the triple (CdMn)Te/(CdMg)Te QW structure under study with QWs of 4, 6, and 10 nm (inset) and high resolution photoluminescence (PL) spectra recorded at 1.5 K at different magnetic field values. Dashed lines show the PL spectra measured at 2 K with lower spectral resolution, which was used in ODMR experiments

(CdMg)Te buffer layer and contained three (CdMn)Te quantum wells with the widths of 4, 6, and 10 nm. The Mn content was ca. 1 %. The QWs were not doped. They were separated by rather large (30 nm) (CdMg)Te barriers and covered with a 100 nm (CdMg)Te cap layer. Our ODMR study of QWs covered with such a thick cover layer have shown that no excess hole concentration is created in such QWs.

Photoluminescence (PL) was excited above the band gap using a 650 nm semiconductor laser (ca. 1 W/cm^2), and detected with a triple monochromator and a CCD camera. In ODMR measurements, a single-grating monochromator and a photomultiplier tube were used. The 35 and 94 GHz ODMR spectra were recorded at a temperature of 1.8 to 2 K via PL intensity at a fixed wavelength which was chosen in such a way that the PL intensity were proportional to the PL line shift in the absence of the microwave field. For ODMR measurements at 94 GHz a quasi-optical microwave circuit shown in the inset in Fig. 2 was used. Its specific features were a high power microwave generator (100 mW) and a field concentrator described in Ref. [18]. In the 35 GHz ODMR spectrometer the sample was placed

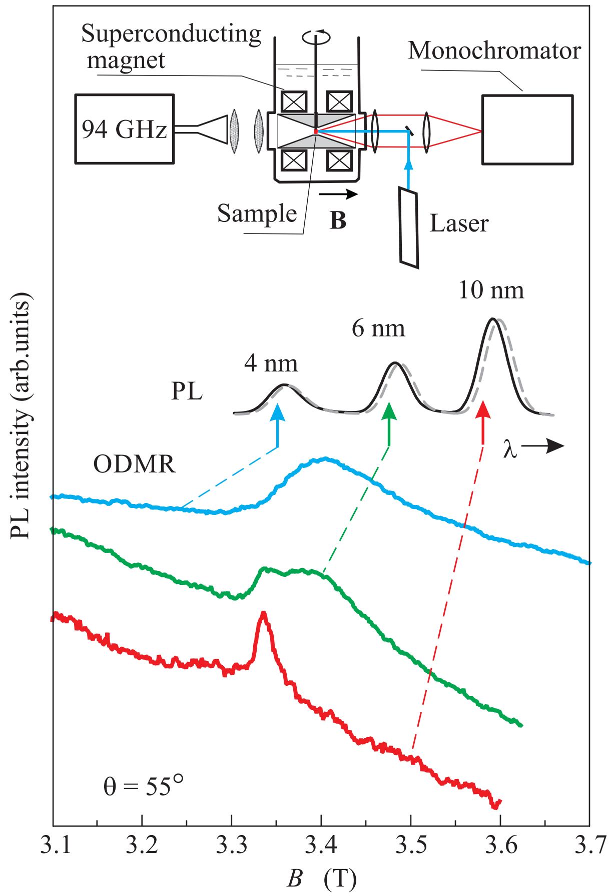

Fig. 2. Block diagram of the 94 GHz ODMR spectrometer (inset) and ODMR spectra recorded at 2 K in the (CdMn)Te QW structure by monitoring the PL intensity at the wavelengths marked by arrows. Solid and dashed lines show PL spectra in magnetic fields of 3 and 4 T, respectively

in the center of a cylindrical microwave cavity with holes for excitation and detection of PL. The sample was mounted on a rotating sample holder, which enables the ODMR measurements at different orientations of the sample relative to the magnetic field.

High-resolution PL spectra measured in different magnetic fields at a temperature of 1.5 K in the (CdMn)Te/(CdMg)Te QWs structure under study are shown in Fig. 1. The magnetic field was directed along the growth axis [001]. For each QW the spectrum consists of two emission lines – excitons (X) and charged excitons (trions, T). They manifest similar shifts to lower energy (larger wavelength) with increasing magnetic field. In ODMR experiments, we used a lower spectral resolution. The corresponding PL spectra measured at the magnetic field values of 3 and 4 T are shown in Fig. 1 by dashed lines.

Fig. 2 shows 94 GHz ODMR spectra of three QWs recorded via PL intensity at the wavelengths marked by arrows with microwaves applied in cw mode. The angle between the growth axis [001] and the magnetic field was $\theta = 55^0$. In the absence of microwaves the field dependencies of the PL intensity were close to linear and the shifts of the PL lines produced by increasing magnetic field and resonant microwaves were inside these linear dependencies. Thus, the PL intensity was proportional to the shift of the PL line. The ODMR spectra measured at both slopes of the PL line were found to be identical, which confirms reasonableness of such an approach.

The ODMR spectra are anisotropic. The baseline corrected 94 and 35 GHz ODMR spectra in three QWs are shown in Figs. 3a and b for the sample orientation

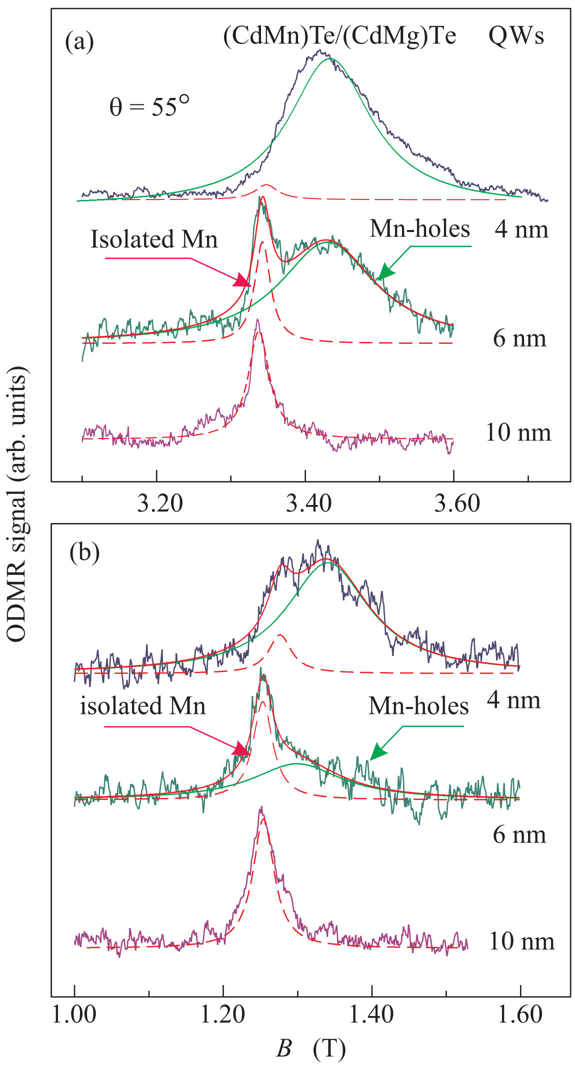

Fig. 3. 94 GHz (a) and 35.1 GHz (b) ODMR spectra for 4, 6, and 10 nm QWs measured at 2 K. The baseline is subtracted. The angle θ between the c-axis and magnetic field is 55^0

$\theta = 55^0$ in which two ODMR lines are better resolved. They belong to two different spin systems, i.e. the narrow lines can be attributed to isolated Mn ions [11, 12] and the wide lines can be ascribed to exchanged-coupled complexes [17] formed by a localized hole and Mn ions. The narrow ODMR line shifts slightly to lower magnetic fields while the wide ODMR line shifts from lower (effective g-factor > 2) to higher fields with increasing angle θ between the magnetic field and [001] growth axis of the structure.

The 94 and 35 GHz spectra are similar but they are very different for 4, 6, and 10 nm QWs. It is clearly seen that the narrow ODMR line of isolated Mn ions is observed in the widest QW and is present in the intermediate 6 nm QW while the wide ODMR line of the complexes (Mn-hole) dominates the spectrum of the narrowest 4 nm QWs, is still observed in 6 nm QW and is nearly absent in 10 nm QW. The formation of such complexes implies that an excess hole concentration is created in the narrowest (4 nm) and intermediate (6 nm) QWs. It is the largest in the narrowest well.

The excess hole concentration seems to appear as a result of the directional electron tunneling from narrower to wider QWs. Accumulation of excess holes happens in spite of very low tunneling probability caused by a large (30 nm) barrier thickness. Surface states have no effect on the hole concentration in our sample because of a thick cup layer [14]. Although ODMR spectra can not provide direct information on the hole concentration it can be roughly estimated from comparison of the ODMR spectra in the structure under study and the (CdMn)Te/(CdMg)Te QW with 2D hole gas that was studied in Refs. [15] and [17] as being of the order of 10^{11}. It is the largest in the narrowest QW.

The g-factor of individual Mn^{2+} ions is known to be isotropic and close to $g = 2$, and therefore cannot give rise to an anisotropy of the ODMR signal. An anisotropy can appear in the ODMR spectra owing to the zero field splitting of the energy levels, which can be due to strain-induced fine-structure splitting [11, 12] or exchange interactions of Mn ions with localized holes [17]. Because of a large Boltzmann factor only the lowest energy sublevels of the spin system are populated in high magnetic fields at a low temperature and the ODMR lines corresponding to EPR transitions between these levels can only be detected.

A spin Hamiltonian for isolated Mn^{2+} is given by:

$$\hat{H}_{\mathrm{Mn}} = g\mu_{\mathrm{B}}\mathbf{B}\cdot\mathbf{S} + D\left[S_z^2 - \frac{1}{3}S(S+1)\right], \qquad (1)$$

where μ_{B} is Bohr magneton, $g = 2.0032$ and is isotropic, $S = 5/2$, and D is a zero-field splitting parameter for the axial crystal field. Hyperfine interactions are not included. The symmetry of the fine-structure splitting, which is due to the crystal field, is consistent with the [001] growth direction.

At thermal equilibrium the $M_S = -5/2 \leftrightarrow M_S = -3/2$ and $M_S = -3/2 \leftrightarrow M_S = -1/2$ EPR transitions dominate the spectra. This results in a shift of the observed ODMR line from higher to lower fields with increasing the angle θ. The parameters of the Spin Hamiltonian (1) for isolated Mn ions in 10 and 6 nm QWs were found to be the same within the experimental error: $g = 2$, $D = 15 \cdot 10^{-4}\,\text{cm}^{-1}$.

The energy levels of complexes formed by a localized hole and Mn^{2+} ions with $S = 5/2$ can be described by the Hamiltonian

$$\hat{H} = \hat{H}_{\text{Mn}} + \hat{H}_h + \hat{S}_h^* \sum c_i \hat{S}_{\text{Mn}i}, \qquad (2)$$

where the first two terms are the spin Hamiltonians for the isolated Mn^{2+} with $S = 5/2$ and the localized hole. The third term is the exchange interaction between the localized hole and the Mn^{2+} ions. $\hat{S}^*$ is the effective spin of the hole. For the ferromagnetic interaction the coefficients c_i are negative. Here we neglect interactions between Mn^{2+} ions.

In zinc blend semiconductors, the top of the valence band consists of the heavy- and light-hole subbands, each twofold degenerate in angular-momentum projection. For the magnetic fields used in the ODMR experiments the Zeeman energy splittings are much smaller than the difference in confinement energy for the light and heavy holes. At liquid-helium temperature only the heavy-hole energy levels $J_z = \pm 3/2$ are populated and we can describe the magnetic properties of heavy holes using an effective spin $S^* = 1/2$:

$$\hat{H}_h = \mu_{\text{B}}[g_{h\parallel} B_z S_z^* + g_{h\perp}(B_x S_x^* + B_y S_y^*)], \qquad (3)$$

where $g_\parallel$ and $g_\perp$ are the components of the $\hat{g}$ tensor along the [001] growth axis (z-axis) and perpendicular to it. According to a theoretical consideration the heavy hole spin is frozen in the growth direction due to quantum confinement or, equivalently, its in-plane g-factor is vanishing. In an ideal QW having D_{2d} symmetry, only the longitudinal component of the heavy-hole g-factor tensor g_{zz} is appreciable.

In Fig. 4a, angular variations of the ODMR spectra in the narrowest (4 nm wide) QW are shown. They are typical for complexes formed by Mn ions and holes and suggest the presence of these complexes in the narrowest QWs. The ODMR lines have an asymmetrical shape that is changed depending on the microwave power as shown in Fig. 4b where the ODMR spectra measured with the maximum microwave power and at a power decreased by 10 and 20 dB are presented. With increasing microwave power the ODMR line becomes more symmetrical and its center of weight shifts to the field corresponding to $g = 2$.

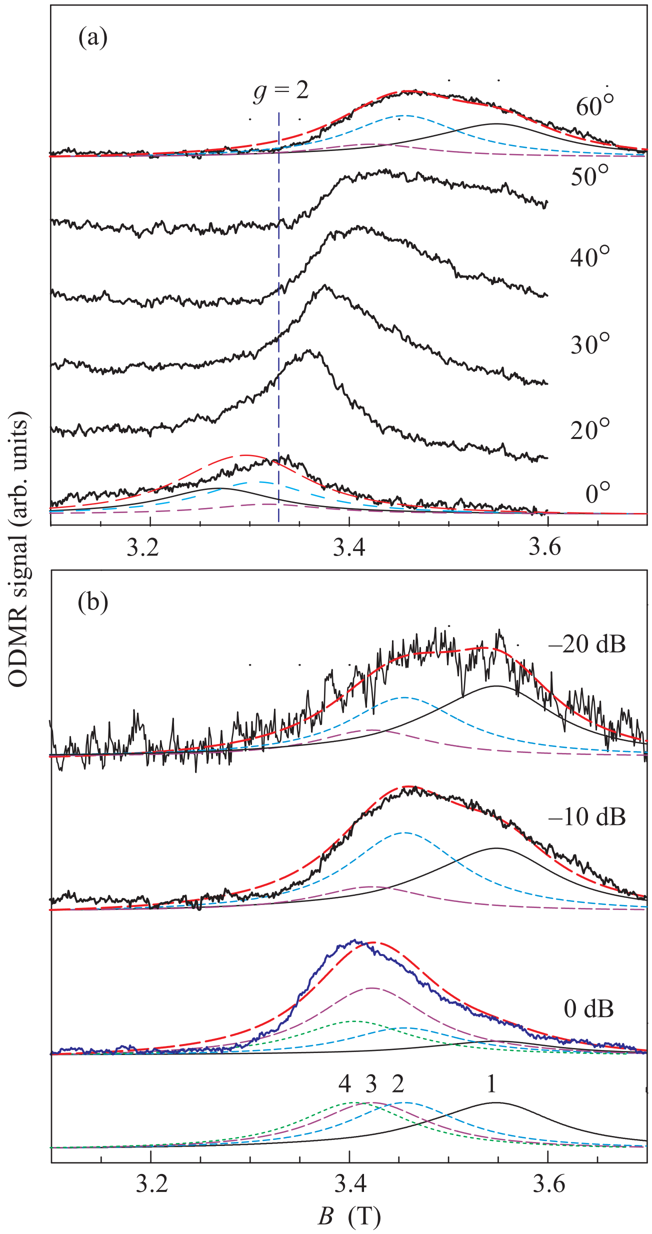

Fig. 4. (a) – Variations of 94 GHz ODMR spectra for 4 nm QW with the angle between the magnetic field and the [001] growth axis. The baseline is subtracted. (b) – Experimental and simulated ODMR spectra for 4 nm QW recorded with the maximum microwave power (0 dB) and with 10 and 20 dB attenuation. Calculated spectra for exchange-coupled complexes formed by a localized hole and 1, 2, 3, and 4 Mn ions are shown in bottom. In simulated spectra different contributions of complexes with several Mn ions for different levels of microwave power were taken into account. $T = 2\,\text{K}$; $\theta = 60^0$. The spectra in panel a were measured at $-10\,\text{dB}$

The shape of the ODMR spectra can be explained if we suppose that there exists a distribution of exchanged-coupled complexes in which a hole is coupled to several

Mn ions. It is known that the spin-lattice relaxation time shortens with an increased number of particles in exchange-coupled systems [19]. To observe ODMR signal it is necessary that the microwaves could change the population difference between the spin levels, i.e. the rate of the microwave transitions should be larger than the spin-lattice relaxation rate. At low microwave power only centers with long enough spin-lattice relaxation time can be detected. By increasing the microwave power the ODMR signals from complexes with shorter relaxation, i.e. complexes including several Mn ions appear and increase.

Model calculations for the exchange-coupled complexes in which a hole interacts with 1, 2, 3, and 4 Mn ions were performed using the EasySpin program [20] that took into account the Boltzmann population distribution between spin sublevels. The following parameters were used in calculations: $g = 2$ for Mn, $g_{||} = 2.4$ and $g_{\perp} = 1.1$ for a hole, and the exchange interaction constants $c_i = -1\,\mathrm{cm}^{-1}$. A 160 mT wide Lorentzian line was used to account for spreading of the exchange interactions. The results of calculations for exchanged-coupled complexes consisting of a localized hole and 1, 2, 3, and 4 Mn ions are shown in the bottom of Fig. 4b. The ODMR line shifts to $g = 2$ with increasing number of Mn ions in the complex. For the simulated ODMR spectra recorded at a higher microwave power an increased contribution of complexes including larger amount of Mn ions was taken into account. The simulated spectra shown in Figs. 4a and b are in good agreement with the experimental spectra.

In summary, analysis of ODMR spectra in the system of multiple anisotropic (CdMg)Te QWs separated by large (CdMg)Te barriers has shown that different Mn-related centers are responsible for magneto-optical effects in QWs with different width. Exchange-coupled complexes consisting of manganese ions and holes were found in the narrowest and intermediate-width QWs and an evidence of the existence of a distribution of such complexes that differ in a number of Mn ions interacting with a hole is presented. The spin Hamiltonian parameters of the complexes were estimated and simulation of their ODMR spectra was performed. The formation of such complexes implies creation of the excess hole concentration in the narrowest QWs of the structure and suggests directional tunneling of photo-created electrons from narrower to wider wells.

This work has been supported by the Megagrant of the Russian Government 14.Z50.31.0021, Russian Foundation of Basic Research under grant # 13-02-00821, and Russian Ministry of Education and Science under grant # 14.604.21.0083.

1. J. K. Fudyna, J. Appl. Phys. **64**, R29 (1988).
2. A. Abragam and B. Bleaney, *Electron Paramagnetic Resonance of Transition Ions*, Oxford University Press, Oxford (1970).
3. P. G. Baranov and N. G. Romanov, Appl. Magn. Resonance **21**, 165 (2001).
4. A. V. Komarov, S. M. Ryabchenko, O. V. Terletskii, I. I. Zheru, and R. D. Ivanchuk, ZhETF **73**, 608 (1977) [Sov. Phys. JETP **46**, 318 (1977)].
5. P. G. Baranov, M. F. Bulanyi, V. A. Vetrov, and N. G. Romanov, JETP Lett. **38**, 623 (1983).
6. A. V. Malyavkin, Phys. Stat. Sol. B **115**, 353 (1983).
7. S. J. C. H. M. van Gisbergen, M. Godlewski, R. R. Galazka, T. Gregorkiewicz, C. A. J. Ammerlaan, and N. T. Khoi, Phys. Rev. B **48**, 11767 (1993).
8. S. Zeng, L. C. Smith, J. J. Davies, D. Wolverson, S. J. Bingham, and G. N. Aliev, Phys. Stat. Sol. B **243**, 887 (2006).
9. M. L. Sadowski, M. Byszewski, M. Potemski, A. Sachrajda, and G. Karczewski, Appl. Phys. Lett. **82**, 3719 (2003).
10. V. Yu. Ivanov, M. Godlewski, D. R. Yakovlev, M. K. Kneip, M. Bayer, S. M. Ryabchenko, and A. Waag, Phys. Rev. B **78**, 085322 (2008).
11. P. G. Baranov, N. G. Romanov, D. O. Tolmachev, R. A. Babunts, B. R. Namozov, Yu. G. Kusrayev, I. V. Sedova, S. V. Sorokin, and S. V. Ivanov, JETP Lett. **88**, 631 (2008).
12. D. O. Tolmachev, R. A. Babunts, N. G. Romanov, P. G. Baranov, B. R. Namozov, Y. G. Kusrayev, S. Lee, M. Dobrowolska, and J. K. Furdyna, Phys. Stat. Sol. B **247**, 1511 (2010).
13. H. Boukari, P. Kossacki, M. Bertolini, D. Ferrand, J. Cibert, S. Tatarenko, A. Wasiela, J. A. Gaj, and T. Dietl, Phys. Rev. Lett. **88**, 207204-1 (2002).
14. S. Tarasenko, M. Bertolini, W. Maslana, H. Boukari, B. Gilles, J. Cibert, D. Ferrand, P. Kossacki, and J. A. Gai, Opto-Electron. Rev. **11**, 133 (2003).
15. C. Kehl, G. A. Astakhov, K. V. Kavokin, Yu. G. Kusrayev, W. Ossau, G. Karczewski, T. Wojtovicz, and J. Geuts, Phys. Rev. B **80**, 241203(R) (2009).
16. K. V. Kavokin, Phys. Rev. B **59**, 9822 (1999).
17. D. O. Tolmachev, A. S. Gurin, N. G. Romanov, A. G. Badalyan, R. A. Babunts, P. G. Baranov, B. R. Namozaov, and Yu. G. Kusrayev, Pis'ma v ZhETF **96**, 247 (2012).
18. R. A. Babunts, A. G. Badalyan, N. G. Romanov, A. S. Gurin, D. O. Tolmachev, and P. G. Baranov, Technical Phys. Lett. **38**, 887 (2012).
19. A. Bencini and D. Gatteschi, *Electron Paramagnetic Resonance of Exchange Coupled Systems*, Springer-Verlag, Berlin-Heidelberg (1990).
20. S. Stoll and A. Schweiger, J. Magn. Reson. **178**, 42 (2006).