Optical probing of the spin state of a single magnetic impurity in a self-assembled quantum dot

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Optical spectra of a self-organized quantum dot with a single magnetic impurity are studied. The quantum dot potential and exchange interactions result in an additional fine structure of an exciton localized in a quantum dot with a magnetic impurity. In contrast to the undoped quantum dots, the ground state excitons in the quantum dot with a magnetic impurity are bright. It is also shown that an initial spin state of the Mn atom leaves a unique pattern in the optical response. This suggests the possibility to read out optically the spin polarization of a single Mn atom.

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Electrons in solids move in a very complex environment interacting with a phonon bath and experiencing spin-orbit and exchange interactions. These interactions affect the spin state of electrons and cause a variety of interesting spin-dependent phenomena. New, rapidly developing fields of research, spintronics and quantum computation, utilize spin-dependent effects and provide the principles to control and manipulate the spin states of carriers in semiconductor and magnetic materials.[1,2] One technologically important class of spintronics materials is diluted magnetic semiconductors.[3] A diluted magnetic semiconductor combines some of the properties of high-quality semiconductor crystals with magnetic properties of impurities. This combination may certainly provide important advantages for spintronics since the semiconductor technology is very well developed and semiconductor crystals can be grown in the form of multi-layer structures. Experimentally, carriers and spins in semiconductors can efficiently be manipulated via polarized optical pulses[4,5] because many semiconductors have optically active interband transitions. Presently, one of the main materials under study is GaMnAs. For this material and other magnetic semiconductors, the main emphasis in current research concerns a behavior of large collections of Mn-spins and mobile carriers.[6] In particular, optical and magnetic effects were studied in semimagnetic II-VI quantum dots (QDs) where photogenerated excitons interact with a large number of magnetic atoms.[7,8] In contrast, this paper will discuss a single Mn-impurity system in a quantum dot. In such a system, the quantum state of a single magnetic atom can be controlled optically via the generation of excitons. So far, QDs with single Mn impurities were considered only for the case of spherical nanocrystals with a strong confinement.[9] In particular, it was shown in Ref. 9 that the optical response of quantum dots depends on the spin state of Mn impurity. Here we will focus on a different case, an oblate self-organized QD with a relatively weak confinement. In other words, we will assume that the radius of the Mn acceptor state is smaller than that of a QD. This approach is applicable for the InGaAs/GaAs QD systems since a Mn-impurity in GaAs forms a deep acceptor level with the radius essentially smaller than the typical size of the self-assembled QDs.

Here, we study theoretically the physical properties of a single Mn impurity embedded in a self-assembled InGaAs/GaAs quantum dot. We think that the QD system would be advantageous compared to a Mn atom in a uniform crystal because the optically excited electron and hole in a QD are well confined and can be well controlled.[10] We note that, in the case of Mn impurities in GaAs, the previous optical studies reported the conduction band-acceptor transitions without details on the electron binding.[11] Technologically, a realization of self-assembled QDs with embedded single impurities looks feasible since all the steps of the growth process, when taken separately, are well known. At the same time, single-dot optical spectroscopy of QDs is also well developed.[12–14] In this paper we will focus on optical responses of a negatively charged Mn acceptor A^-. The presence of the charged state A^- implies that the structure contains some amount of donor impurities or a back contact with electrons. Historically, the state A^- was studied first[15] whereas the details of the neutral state A^0 became known later.[16–18]

We start with the initial state of A^- in a QD. This state has the completely filled valence band and the Mn spin $I=\frac{5}{2}$ [Fig. 1(a)]. Therefore the initial state of A^- can be in either of six states: $I_z=\pm\frac{5}{2},\pm\frac{3}{2},\pm\frac{1}{2}$. In the optical absorption process, an incident photon creates an exciton composed of an electron and hole and we now consider the lowest energy states associated with the exciton in a QD [Fig. 1(a)]. The lowest exci-

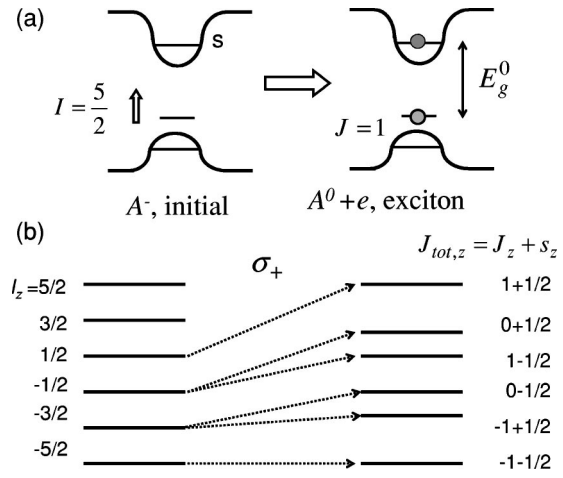

FIG. 1. (a) Schematics of the optical absorption process. (b) Optical transitions in the system induced by the σ_+ photon.

ton states can be regarded as a system of A^0 and an electron in the lowest s state. It is know that the Mn impurity in bulk GaAs forms a deep acceptor level ($\approx$112 meV above the top of the valence band) and the size of hole wave function is quite small, about 1 nm.[19] At the same time, a size of QD is typically larger, about a few nm. The hole of a Mn atom is exponentially localized and, therefore, we can start from the case of Mn impurity in bulk GaAs and treat the QD potential as a perturbation. In the envelope-function approximation, the Hamiltonian of the Mn impurity has the form:

$$\hat{H}_{imp} = \hat{T} + U_{imp}(\mathbf{r}_h) + \hat{H}^{exc}_{\text{hole-Mn}} + U_h^{\text{QD}}, \tag{1}$$

where $\hat{T}$ is the "kinetic energy" of the Luttinger model, $U_{imp}(\mathbf{r})$ is the impurity potential energy, and U_h^{QD} is the valence-band potential in an undoped QD. The exchange interaction between the valence band hole and Mn spin is taken in the conventional form,

$$\hat{H}^{exc}_{\text{hole-Mn}} = A_h \hat{\mathbf{I}}\hat{\mathbf{j}}\delta(\mathbf{r}_h - \mathbf{R}_{imp}), \tag{2}$$

where $\hat{\mathbf{I}}$ and $\hat{\mathbf{j}}$ are the angular momenta operators of the Mn-spin and hole $\left(j=\frac{3}{2}\right)$, respectively; A_h is the exchange interaction constant; and $\mathbf{r}_h$ and $\mathbf{R}_{imp}$ are the coordinates of the hole and impurity, respectively. The total angular moment of the impurity becomes then: $\hat{\mathbf{J}}=\hat{\mathbf{I}}+\hat{\mathbf{j}}$. The wave functions of the hole in bulk GaAs, $|J,J_z>$, are eigenfunctions of the operator $\hat{T}+U_{imp}(\mathbf{r}_h)+\hat{H}^{exc}_{\text{hole-Mn}}$ and were studied in detail in Refs. 16 and 19. These wave functions correspond to the state $1S_{3/2}$ with $J=1$:

$$|1,+1\rangle = \frac{1}{\sqrt{2}}\psi_{5/2,-3/2} - \frac{\sqrt{30}}{10}\psi_{3/2,-1/2} + \frac{\sqrt{15}}{10}\psi_{1/2,1/2} - \frac{\sqrt{5}}{10}\psi_{-1/2,3/2}$$

$$|1,0\rangle = \frac{1}{\sqrt{5}}\psi_{3/2,-3/2} - \frac{\sqrt{30}}{10}\psi_{1/2,-1/2} + \frac{\sqrt{30}}{10}\psi_{-1/2,1/2} - \frac{\sqrt{5}}{5}\psi_{-3/2,3/2}$$

$$|1,-1\rangle = -\frac{1}{\sqrt{2}}\psi_{-5/2,3/2} + \frac{\sqrt{30}}{10}\psi_{-3/2,1/2} - \frac{\sqrt{15}}{10}\psi_{-1/2,-1/2} + \frac{\sqrt{5}}{10}\psi_{1/2,-3/2}. \tag{3}$$

Here, $\psi_{I_z,j_z}=|I_z\rangle\phi_{j_z}$, where ϕ_{j_z} are the eigenstates of a bound hole in the absence of the exchange interaction and $|I_z\rangle$ are the states of Mn spin. The wave functions ϕ_{j_z} are given by

$$\phi_{j_z} = R_0(r)Y_{00}u_{j_z} + \sum_{\nu=\pm 3/2,\pm 1/2} C^{3/2,j_z}_{j_z-\nu,\nu} R_2(r) Y_{2,j_z-\nu} u_\nu, \tag{4}$$

where u_{j_z} are the Bloch functions of the hole $j=\frac{3}{2}$ and $C^{3/2,j_z}_{j_z-\nu,\nu}$ are the Clebsch-Gordan coefficients. The functions $R_{0(2)}$ are the solutions of the coupled equations of the Baldereschi-Lipari theory.[20] The energy of the $1S_{3/2}$ states as a function of J is given by $E_{imp}=E^0_{imp}+(\epsilon/2)J(J+1)$, where $\epsilon = A_h R_0^2(0)/4\pi$, and $J=1,2,3$, and 4. The energy spacing between the two lower states $J=1$ and 2 is equal to 2ϵ. The experimental value for the above parameter is $2\epsilon \sim 10$ meV.[18]

Since the radius of the impurity state is smaller than the size of QD, the QD potential [the last term in Eq. (1)] can be regarded as perturbation. For the QD model, we choose a parabolic valence-band potential: $U_h^{\text{QD}}(\mathbf{r})=m_h\omega_z^2 z^2/2 + m_h(\omega_x^2 x^2+\omega_y^2 y^2)/2$, where m_h is the heavy-hole effective mass; the frequencies $\omega_{z,x,y}$ describe an anisotropic confinement of a QD; and x and y are the in-plane coordinates (Fig. 2). If the center of the QD and the impurity position do not coincide, it is convenient to shift the QD potential: $U^h_{\text{QD}}(\mathbf{r}'+\mathbf{R}_{imp})$. Calculation of matrix elements of $U^h_{\text{QD}}(\mathbf{r}'+\mathbf{R}_{imp})$ in the basis of the functions (3) is straightforward:

$$\begin{pmatrix} am_h\omega_z^2 + cm_h(\omega_x^2+\omega_y^2) & 0 & fm_h(-\omega_x^2+\omega_y^2) \\ 0 & bm_h\omega_z^2 + dm_h(\omega_x^2+\omega_y^2) & 0 \\ fm_h(-\omega_x^2+\omega_y^2) & 0 & am_h\omega_z^2 + cm_h(\omega_x^2+\omega_y^2) \end{pmatrix}, \tag{5}$$

where $a=0.233$ nm^2, $b=0.262$ nm^2, $c=0.261$ nm^2, $d=0.247$ nm^2, and $f=0.014$ nm^2. To obtain the above matrix, the wave functions $R_{0,2}$ (Ref. 19) were approximated as follows: $R_0(r)=3.95e^{-1.7r}$ and $R_2(r)=1.64re^{-1.5r}$, where r should be taken in nm. We note that the matrix (5) does not contain the impurity coordinate.[21] Typically, a self-assembled QD has the shape of a disk and therefore the strongest contribution in U_h^{QD} is due to the confinement in the z direction. The terms in U_h^{QD} with $\omega_{x(y)}$ are expected to be smaller. In self-assembled QDs, the typical in-plane size of the hole potential, $l_{h,x(y)}=\sqrt{\hbar/m_h\omega_{x(y)}}$, is about 4 nm.[22] For the vertical size of the QD, we take $l_{h,z}=2$ nm. From these numbers and the heavy-hole mass $m_h=0.5\,m_0$, we obtain the frequencies: $\hbar\omega_z=47$ meV and $\hbar\omega_{x(y)}\sim 10$ meV. Using the above parameters, we see that the main effect comes from the z confinement which splits the triplet (3) into a singlet ($J_z=0$) and doublet ($J_z=\pm 1$). This splitting is $\Delta_0=380$ μeV. In a symmetric QD, the states $J_z=\pm 1$ remain degenerate. In the case of asymmetric QD, they are split. Taking $\hbar\omega_x=8$ meV and $\hbar\omega_y=12$ meV ($l_{h,x}\approx 4.9$ nm and $l_{h,y}\approx 4$ nm), we obtain a weak splitting of the $J_z=\pm 1$ states: $\delta_1=fm_h(-\omega_x^2+\omega_y^2) \sim 12$ μeV. It is also worthwhile to note that our perturbation

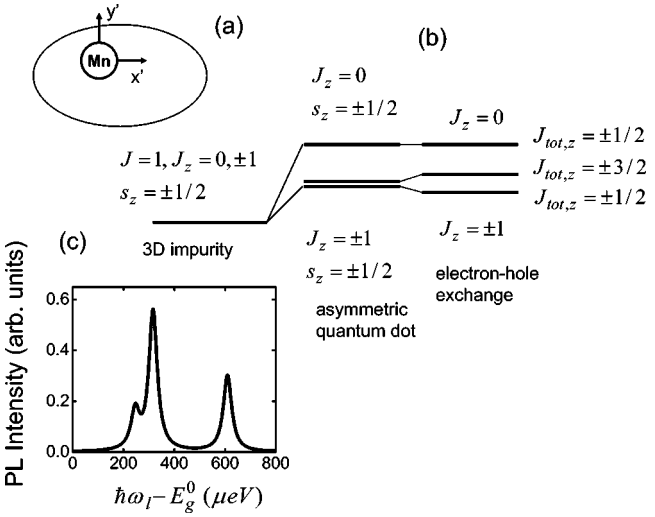

FIG. 2. (a) Geometry of an asymmetric quantum dot with a Mn impurity. (b) The lowest states of an exciton in a dot with Mn impurity. (c) Calculated PL spectrum; the width of Lorentzian peaks is 30 μeV; for typical self-assembled InGaAs/GaAs QDs, E_g^0 is about 1 eV.

theory is valid if the impurity is situated not far from the center of a QD [$R_{imp,z} \sim l_z$ and $R_{imp,x(y)} \sim l_{x(y)}$] and all the energy shifts are less than 2ϵ. The latter condition is well satisfied since $\Delta_0, \delta_1 \ll 2\epsilon$.

Along with the hole–Mn exchange interaction, the optically generated exciton in a QD can experience the electron–Mn and electron–hole exchange interactions. The first interaction is expected to be very weak because the electron wave function is much more delocalized. In addition, the constant of electron–Mn exchange interaction is typically several times smaller than that of the hole–Mn interaction. Indeed, our estimations led us to the electron–Mn interaction energy of about 3 μeV. To obtain the above number, we assumed that $|A_h/A_e| \sim 5$, where A_e is the constant of electron–Mn exchange interaction.[9] For the sizes of electron wave function, we assumed $l_{e,z} = 2$ nm and $l_{e,x(y)} \sim 5$ nm.[22] The electron–hole exchange interaction in the exciton can be stronger. This interaction is described by the Hamiltonian:[23]

$$\hat{H}_{e-h}^{exc} = \sum_{i=x,y,z} \Delta_i^0 \hat{j}_i \hat{s}_i + \Delta_i^1 \hat{j}_i^3 \hat{s}_i, \tag{6}$$

where $\Delta_i^{0,1}$ are constants and $\hat{\mathbf{s}}$ is the electron spin. Using the basis (3), we find the non-zero matrix elements of the operator (6): $\langle 1,+1;s_z|\hat{H}_{e-h}^{exc}|1,+1;s_z\rangle = -\langle 1,-1;s_z|\hat{H}_{e-h}^{exc}|1,-1;s_z\rangle \approx -(1.50\Delta_z^0 + 3.07\Delta_z^1)/2$, where $s_z = \pm 1/2$ is the electron spin. In the above matrix elements, we used the $|J,J_z;s_z\rangle$ basis. The exchange splitting for excitons in QDs was examined experimentally in several publications and was found in the range 200–300 μeV.[13] Theoretically, this splitting is proportional to the integral: $A = \int d^3\mathbf{r} |\Psi(\mathbf{r},\mathbf{r})|^2$, where $\Psi_{exc}(\mathbf{r}_e,\mathbf{r}_h)$ is the wave function of an exciton. In the case of the usual exciton in an undoped QD, the function $\Psi_{exc}(\mathbf{r}_e,\mathbf{r}_h) = \Psi_e(\mathbf{r}_e)\Psi_h(\mathbf{r}_h)$ and contains two strongly overlapping functions Ψ_e and Ψ_h which describe the electron and hole motions, respectively. Also, in the first approximation, we can assume that the electron and hole occupy equal volumes, $V_e \sim V_h$. Then, the integral can be estimated as $A \sim 1/V_e$. However, in our case, the overlap between the electron and hole functions is strongly reduced because the hole is strongly localized in the vicinity of a Mn impurity. We can roughly estimate the above integral by taking $\Psi_e \sim V_e^{-1/2}$ and $\Psi_h \sim V_h^{-1/2}$. Then, assuming $V_e \gg V_h$ we obtain $A \sim 1/\max(V_e,V_h) = 1/V_e$. This demonstrates that the integral A should not be strongly reduced in the case of a QD with a Mn impurity. Nevertheless, we calculated numerically the integral A for the exciton in both undoped and doped QDs. We found that the integral A in the case of a QD with impurity is reduced about three times. For our calculations, we used harmonic-oscillator wave functions with the typical lateral sizes of the QD wave functions: $l_{e,x(y)} \sim 5$ nm and $l_{h,x(y)} \sim 4$ nm. For the vertical size of the QD, we took 2 nm. In the following, we will assume that the exchange energy $E_{e-h}^{exc} = -(1.50\Delta_z^0 + 3.07\Delta_z^1) = 70$ μeV.

Since the electron–hole exchange interaction is expected to be stronger than the anisotropic splitting δ_1 and the electron–Mn impurity exchange energy, we can neglect the latter two contributions. We note that the neglected interactions induce small energy shifts in the spectrum and do not lead to any additional splittings. Thus, using the basis $\Phi_{J_z,s_z} = |J,J_z;s_z\rangle$, we obtain the following set of excitonic eigenstates:

$$\Phi_{0,\pm 1/2} = |+1,0;\pm 1/2\rangle,$$

$$\Phi_{\pm 1,\pm 1/2} = |+1,\pm 1;\pm 1/2\rangle. \tag{7}$$

The states $\Phi_{0,\pm 1/2}$ have the energy $E_{0,\pm 1/2} = bm_h\omega_z^2 + dm_h(\omega_x^2 + \omega_y^2)$. The energies of the other states are $E_{+1,-1/2} = E_{-1,1/2} = am_h\omega_z^2 + cm_h(\omega_x^2 + \omega_y^2) + E_{e-h}^{exch}/2$ and $E_{-1,-1/2} = E_{1,1/2} = am_h\omega_z^2 + cm_h(\omega_x^2 + \omega_y^2) - E_{e-h}^{exch}/2$. Since the coefficients $\Delta_z^{0,1}$ are typically negative, the ground states are $\Phi_{\pm 1,\mp 1/2}$ and correspond to $|J_z| = 1$ and $J_{tot,z} = J_z + s_z = \pm 1/2$ [see Fig. 2(b)].

We now calculate the interband optical matrix elements responsible for the photoluminescence (PL) process, assuming that the exciton can be found in either of the lowest states $|1,J_z;s_z\rangle$ with the same probability. The calculated PL spectrum is not polarized and shown in Fig. 2(b) as a function of $\omega_l - E_g^0$, where ω_l is the photon energy and E_g^0 is the characteristic interband energy [see Fig. 1(a)]. For this energy we have $E_g^0 = E_{cv}^{dot} - E_{\text{binding}}^{\text{Mn}} + E_c^e + am_h\omega_z^2 - E_{cv}^{\text{Coul}}$, where E_{cv}^{dot} is the conduction-to-valance band spacing at the position of the Mn impurity in the QD, $E_{\text{binding}}^{\text{Mn}} = 112$ meV is the binding energy of the Mn acceptor in bulk GaAs, and E_c^e is the quantization energy of electron in the conduction band of a QD. E_{cv}^{Coul} is the direct Coulomb interaction in the exciton, which is typically about 10–20 meV in the InGaAs QD systems. In our model, the QD confinement and the hole confinement for the deep Mn-impurity level are essentially stronger than the interband Coulomb interaction in the exciton and, therefore, the Coulomb interaction leads only to a shift of the interband energy E_g^0. For InGaAs/GaAs QDs, the energy E_g^0 is expected to be about 1 eV.

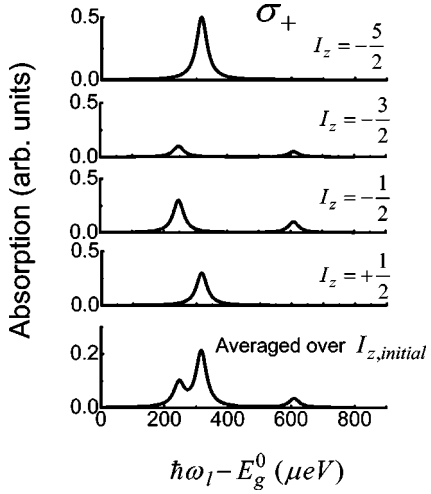

FIG. 3. Absorption spectra for different initial Mn states I_z of the A^- impurity; the photon circular polarization is σ_+. The absorption for the initial Mn states with $I_z=+3/2$ and $+5/2$ is zero. The lowest panel presents the absorption spectrum of the Mn atom with a randomized spin in the initial state. For the broadening of the peaks we take 30 μeV.

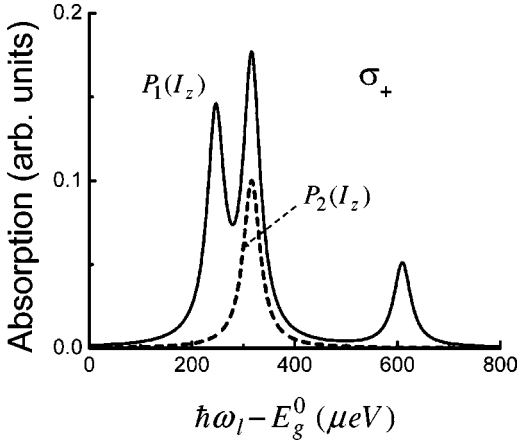

FIG. 4. Absorption spectra of the A^- impurity in a QD for two different initial spin distribution functions, $P_1(I_z)$ and $P_2(I_z)$; the polarization if incident photons is σ_+.

It is interesting to note that optical transitions are allowed from all the lowest states of the exciton [Fig. 2(b)], including the ground state, because several final states with different spins I_z are available. This is in contrast to the usual self-assembled QDs where the ground state of exciton is dark.[13]

Another interesting feature of the exciton spectrum is the possibility to observe *circularly polarized PL at zero magnetic field*. It is known that, in the absence of Mn impurity, the PL spectrum of QDs at zero magnetic field is linearly polarized because of the anisotropic exchange interaction [Eq. (6)].[13] In the presence of the Mn impurity, the strong hole–Mn exchange interaction dictates circular symmetry and the exciton wave functions $\Phi_{J_z,s_z}=|J,J_z;s_z\rangle$ [Eq. (7)] have well defined angular momenta. Therefore, if the exciton population is generated by a circularly polarized light and has nonzero angular momentum, the resulting PL emission can also be circularly polarized. The anisotropic splitting δ_1 can slightly mix the above states and bring a small degree of linear polarization in the emission of excitons. The degree of mixing and linear polarization of individual excitons is given by the small parameters: δ_1/Δ_0 and $\delta_1/\Delta_{e-h}^{exch}$, where Δ_0 and Δ_{e-h}^{exch} are the splittings coming from the QD confinement in the z direction and e–h exchange interaction, respectively. Note that circularly polarized PL emission at zero magnetic field was recently observed in II-VI quantum dots doped by Mn impurities.[8] In the experiment,[8] it is likely that a single QD contains several Mn impurities. We think that our single-impurity model suggests a qualitative explanation for the experiment.[8]

Allowed optical transitions for absorption of a σ_+ photon are shown in Fig. 1(b). The corresponding matrix elements can be found using the wave functions (3) and the electron-photon interaction $\hat{V}_{opt}=V_0(\mathbf{e}_x+i\mathbf{e}_y)\hat{\mathbf{p}}$, where $\mathbf{e}_{x,y}$ are the unit vectors and $\hat{\mathbf{p}}$ is the electron momentum. We observe that the absorption spectrum strongly depends on the initial spin state of a Mn impurity (Fig. 3) and therefore by using the absorption spectrum one can distinguish the initial state of the Mn spin. The lower panel in Fig. 3 shows the spectrum when the Mn spin in the initial state is randomized. In other words, we assumed that the initial state is in thermal equilibrium and therefore $P_{I_z}=1/6$, where P_{I_z} is the probability to find the Mn spin in the state I_z.

If the Mn atom in the initial state is not spin-polarized, the circularly polarized laser pulse can create the polarization of the Mn spin. If the pulse is shorter than the spin relaxation time in the system, we can calculate the change of the averaged Mn spin in the following way: $\Delta\overline{I_z}=(\Delta t/6)\Sigma_\alpha W_\alpha\langle\alpha|\hat{I}_z|\alpha\rangle$, where the index α denotes the exciton states (Φ_{J_z,s_z}), Δt is the pulse duration, W_α is the rate of optical-generation for the exciton in the state α, and $\langle\alpha|\hat{I}_z|\alpha\rangle$ is the average Mn spin in the state $|\alpha\rangle$. Using the interband optical matrix elements, we find that the σ_+ light induces negative spin polarization of the Mn atom: $\Delta\overline{I_z}=-\Delta tW_0\frac{7}{16}<0$, where the parameter W_0 contains the interband optical matrix element P_{cv} between the Bloch states. At the same time, the average spin of the photoexcited hole is positive: $\Delta\overline{j_z}=\Delta tW_0\frac{3}{16}>0$. The negative sign of $\Delta\overline{I_z}$ comes from the inequality $I>j$. It follows from the above calculations that circularly polarized resonant light can induce the nonzero spin polarization in the Mn impurity. In bulk materials, the possibility to polarize the Mn spin has been demonstrated in several publications[24] where the Mn spins were partially polarized by using nonresonant and resonant optical excitations.

We now consider a pump-probe scheme for write and read optical experiments. First, assume that the Mn-spin in a QD becomes polarized by an intense polarized laser pulse. It is known that such a spin polarization can be long lived.[24] The pump pulse can be either resonant or nonresonant.[24] Then, the probe pulse can test the nonequilibrium state of Mn. As an example, we consider two strongly nonequilibrium distribution functions, $P_{1,2}(I_z)$, for the initial state of Mn-spin: $P_1(I_z)=1/3$ if $I_z<1/2$ and 0 otherwise, and $P_2(I_z)=1/3$ if $I_z>-1/2$ and 0 otherwise. In the first case, the Mn spin in

the initial state is predominantly negative, while, for the second distribution function, the average spin in the initial state is positive. The calculated optical absorption of σ_+ photons for the distributions $P_{1,2}$ strongly differ in intensity and shape (see Fig. 4). The absorption spectrum for the initial state P_1 has more lines due to the momentum-conservation selection rules. The above results demonstrate that the initial spin polarization of the Mn impurity can be recognized by recording the absorption spectrum.

In this paper we considered single-dot absorption spectroscopy as a method to read out the spin polarization of an embedded magnetic impurity. It is worthwhile to note that absorption spectra of single self-assembled QDs were recorded in recent experiments.[14] Therefore, proposed absorption spectroscopy of a single impurity is accessible in the state-of-the-art experimental studies. In real doped systems, several Mn impurities can be present in a QD. In this case, the optical manipulation of single spins can become a much more difficult task as we have to control more degrees of freedom.

In conclusion, we propose a self-assembled QD with a single magnetic impurity as a system with efficient optical control of the spin state. We investigated the specific fine structure and optical spectrum of a magnetic impurity embedded into a self-assembled QD and showed that the spin state of a magnetic impurity can be read optically.

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[1] G. A. Prinz, Science **282**, 1660 (1998); S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, *ibid.* **294**, 1488 (2001).

[2] D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).

[3] J. K. Furdina, J. Appl. Phys. **65**, 29 (1988).

[4] *Optical Orientation*, edited by F. Meier and B. P. Zakharchenya (North-Holland, Amsterdam, 1984).

[5] J. M. Kikkawa and D. D. Awschalom, Science **287**, 473 (2000).

[6] T. Dietl, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, Science **287**, 1019 (2000).

[7] R. N. Bhargava, D. Gallagher, X. Hong, and A. Nurmikko, Phys. Rev. Lett. **72**, 416 (1994); P. S. Dorozhkin, A. V. Chernenko, V. D. Kulakovskii, A. S. Brichkin, A. A. Maksimov, H. Schoemig, G. Bacher, A. Forchel, S. Lee, M. Dobrowolska, and J. K. Furdyna, Phys. Rev. B **68**, 195313 (2003); J. Seufert, G. Bacher, M. Scheibner, A. Forchel, S. Lee, M. Dobrowolska, and J. K. Furdyna, Phys. Rev. Lett. **88**, 027402 (2002).

[8] S. Mackowski, T. Gurung, T. A. Nguyen, H. E. Jackson, L. M. Smith, G. Karczewski, and J. Kossut, Appl. Phys. Lett. **84**, 3337 (2004).

[9] A. K. Bhattacharjee and J. Perez-Conde, Phys. Rev. B **68**, 045303 (2003); D. M. Hoffman, B. K. Meyer, A. I. Ekimov, I. A. Merkulov, Al. L. Efros, M. Rosen, G. Couino, T. Gacoin, and J. P. Boilot, Solid State Commun. **114**, 547 (2000).

[10] T. Lundstrom, W. Schoenfeld, H. Lee, and P. M. Petroff, Science **286**, 2312 (1999).

[11] I. Y. Karlik, I. A. Merkulov, D. N. Mirlin, L. P. Nikitin, V. I. Perel, and V. F. Sapega, Fiz. Tverd. Tela (Leningrad) **24**, 3550 (1982) [Sov. Phys. Solid State **24**, 2022 (1982)]; J. M. Kikkawa, J. J. Baumberg, D. D. Awschalom, D. Leonard, and P. M. Petroff, Phys. Rev. B **50**, 2003 (1994).

[12] J.-Y. Marzin, J.-M. Gerard, A. Izrael, D. Barrier, and G. Bastard, Phys. Rev. Lett. **73**, 716 (1994).

[13] See, for example, M. Bayer, A. Kuther, A. Forchel, A. Gorbunov, V. B. Timofeev, F. Schafer, J. P. Reithmaier, T. L. Reinecke, and S. N. Walck, Phys. Rev. Lett. **82**, 1748 (1999).

[14] J. R. Guest, T. H. Stievater, X. Li, J. Cheng, D. G. Steel, D. Gammon, D. S. Katzer, D. Park, C. Ell, A. Thrnhardt, G. Khitrova, and H. M. Gibbs, Phys. Rev. B **65**, 241310 (2002); B. Alen, F. Bickel, K. Karrai, R. J. Warburton, and P. M. Petroff, Appl. Phys. Lett. **83**, 2235 (2003).

[15] N. Almeleh and B. Goldstein, Phys. Rev. **128**, 1568 (1962); R. Bleekrode, J. Dieleman and H. J. Vegter, Phys. Lett. **2**, 355 (1962).

[16] J. Schneider, U. Kaufmann, W. Wilkening, M. Baeumler, and F. Kohl, Phys. Rev. Lett. **59**, 240 (1987).

[17] V. F. Masterov, S. B. Mikhrin, B. E. Samorukov, and K. F. Shtelmakh, Fiz. Tekh. Poluprovodn. (S.-Peterburg) **17**, 1259 (1983) [Sov. Phys. Semicond. **17**, 796 (1983)].

[18] M. Linnarsson, E. Janzen, B. Monemar, M. Kleverman, and A. Thilderkvist, Phys. Rev. B **55**, 6938 (1997).

[19] A. K. Bhattacharjee and C. Benoit a la Guillaume, Solid State Commun. **113**, 17 (2000).

[20] A. Baldereschi and N. O. Lipari, Phys. Rev. B **8**, 2697 (1973).

[21] This comes from the fact that all the matrix elements of the operator $\mathbf{r}'\mathbf{R}_{imp}$ are zero. However, the matrix elements of $\mathbf{r}'\mathbf{R}_{imp}$ become nonzero if we involve the impurity wave functions with $J>1$. The corresponding corrections can be obtained with second-order perturbation theory and therefore are essentially smaller than the matrix elements given by Eq. (5).

[22] R. J. Warburton, B. T. Miller, C. S. Durr, C. Bödefeld, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro, P. M. Petroff, and S. Huant, Phys. Rev. B **58**, 16221 (1998).

[23] E. L. Ivchenko and G. E. Pikus, *Superlattices and Other Heterostructures. Symmetry and Optical Phenomena* (Springer, Berlin, 1997).

[24] H. Krenn, W. Zawadzki, and G. Bauer, Phys. Rev. Lett. **55**, 1510 (1985); H. Krenn, K. Kaltenegger, T. Dietl, J. Spalek, and G. Bauer, Phys. Rev. B **39**, 10918 (1989); D. D. Awschalom, J. Warnock, and S. von Molnar, Phys. Rev. Lett. **58**, 812 (1987).