

## Reentrant formation of magnetic polarons in quantum dots

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We propose a model of magnetic polaron formation in semiconductor quantum dots doped with magnetic ions. A wetting layer serves as a reservoir of photogenerated holes, which can be trapped by the adjacent quantum dots. For certain hole densities, the temperature dependence of the magnetization induced by the trapped holes is reentrant: it disappears for some temperature range and reappears at higher temperatures. We demonstrate that this peculiar effect is not an artifact of the mean-field approximation and persists after statistical spin fluctuations are accounted for. We predict fingerprints of reentrant magnetic polarons in photoluminescence spectra.

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Long spin memory time,<sup>1</sup> giant magnetoresistance,<sup>2</sup> robust magnetic ordering, and its versatile control in epitaxial and colloidal quantum dots (QDs)<sup>3–10</sup> are attributed to magnetic polarons (MPs), known for fifty years in bulk semiconductors.<sup>11</sup> The MP formation can be viewed as a “cloud” of localized spins, aligned through exchange interaction with a confined carrier spin. While the seminal studies of MPs in the bulk<sup>12–14</sup> assumed that an impurity binds only one carrier, many experiments in QDs demonstrate multiple occupancies.<sup>15–19</sup> We show that varying QD occupancy has important consequences for MP formation. It is conventionally understood that MPs form at low temperature ( $T$ ) and vanish at high  $T$ , owing to thermal fluctuations of Mn spins. Here, we propose an unexpected scenario where the temperature can *enhance*, rather than quench, MP formation and lead to reentrant magnetism.<sup>20,21</sup>

We formulate a model of MPs formed in II-VI semiconductor QDs doped with Mn ions and apply it to simulate photoluminescence (PL) spectra.<sup>3,4,22–24</sup> In a PL experiment,<sup>25</sup> the number of carriers captured by QDs depends on the total density of the photoexcited carriers, determined by the laser intensity. Focusing on holes in type-II QDs,<sup>4</sup> we find nontrivial dependencies of MP binding energies on both the laser intensity and  $T$ .

Figure 1 shows our model. Epitaxial QDs typically reside on a two-dimensional (2D) wetting layer (WL) in which electron-hole pairs are created by interband absorption of light.<sup>26</sup> Under illumination, a quasi-Fermi distribution is established in the WL 2D hole gas. Since the total area of the QDs is low relative to that of the WL, the quasi-Fermi level,  $\mu(T, p) = +k_B T \ln\{\exp[p\pi\hbar^2/(m_h^*k_B T)] - 1\}$ , is pinned by the nonequilibrium WL holes with 2D density  $p$ , where  $m_h^*$  is the heavy-hole effective mass. The population of the QDs by the captured holes is controlled by  $\mu$ . This assumption is justified by the fact that in many QD systems, the times of capture and intradot relaxation are much shorter than carrier radiative recombination time. Therefore quasiequilibrium can be established in the valence band.<sup>27</sup>

The exchange interaction of heavy holes with Mn spins in a flat QD is highly anisotropic and described by an Ising Hamiltonian,<sup>8</sup>

$$H_{\text{ex}} = -\beta \sum_{i,j} \delta(\mathbf{r}_i - \mathbf{R}_j) s_{zi} S_{zj}, \quad (1)$$

where  $\beta$  is the exchange coupling constant,  $\mathbf{r}_i$  ( $\mathbf{R}_j$ ) and  $s_{zi}$  ( $S_{zj}$ ) are the position and the spin projection of the carrier (Mn).<sup>28</sup> The exchange interaction results in the Mn-magnetization  $M_z$  and exchange splitting  $\Delta$  of hole levels. We assume a uniform hole wave function throughout the QD volume  $\Omega$ ,<sup>29</sup> thus we can relate  $M_z$  and  $\Delta = \beta M_z/g\mu_B$ . The maximum  $M_z$  is  $M_{\text{max}} = x_{\text{Mn}} N_0 \Omega S g \mu_B$ , where  $x_{\text{Mn}}$  is the Mn fraction per cation,  $N_0$  is the density of cation sites,  $S = 5/2$ ,  $g = 2.0$  is the  $g$  factor,  $\mu_B$  is the Bohr magneton, and  $N_0 \beta \sim -1.0$  eV.<sup>28</sup>

The Gibbs free energy of the system is expressed as

$$G_{\text{sys}}(\xi) = G_{\text{Mn}}(\xi) + F_h(\xi), \quad (2)$$

in terms of the order parameter  $\xi = M_z/M_{\text{max}}$ , where  $G_{\text{Mn}}(\xi)$  is the Mn-spins contribution and  $F_h(\xi)$  is the hole grand canonical (GC) free energy.  $G_{\text{Mn}}(\xi)$  can be obtained by expressing the free energy of the Mn spins as a function of an external magnetic field and then applying a Legendre transformation:<sup>20,30</sup>

$$G_{\text{Mn}}(\xi) = k_B T N_{\text{Mn}} \left\{ \xi B_S^{-1}(\xi) - \ln \frac{\sinh[(1 + 1/2S)B_S^{-1}(\xi)]}{\sinh[B_S^{-1}(\xi)/2S]} \right\}, \quad (3)$$

where  $B_S^{-1}(\xi)$  is the inverse Brillouin function<sup>30</sup> and  $N_{\text{Mn}}$  is the number of Mn in the QD.  $F_h(\xi)$  is obtained from the QD states with 0, 1, or 2 holes [see Fig. 1(c)]. For transparency, we consider only one nonmagnetic single-hole level and neglect the possibility of magnetization in the presence of two holes.<sup>6,31</sup> This yields

$$F_h(\xi) = -k_B T \ln \left[ 1 + 2e^{-(\varepsilon-\mu)/k_B T} \cosh \left( \frac{\Delta_{\text{max}} \xi}{2k_B T} \right) + e^{-(2\varepsilon+U-2\mu)/k_B T} \right], \quad (4)$$

where  $\varepsilon$  is the single-hole confining energy,  $U$  is the repulsive Coulomb (charging) energy, and the maximum splitting is  $\Delta_{\text{max}} = x_{\text{Mn}} |N_0 \beta| S$ . To elucidate some interesting phenomena, we first use a standard mean field (MF) approximation,<sup>12–14</sup> commonly used by many authors.<sup>3,6,7,32</sup> By numerically minimizing Eq. (2), we obtain the value  $\xi_{\text{MF}}$ . The MP energy,  $E_{\text{MP}}$  is defined as the expectation value of Eq. (1). We calculate it from  $E_{\text{MP}} = -k_B T \Delta_{\text{max}} d(\ln Z_{\text{sys}})/d\Delta_{\text{max}}$ .<sup>34</sup> The MF partition function

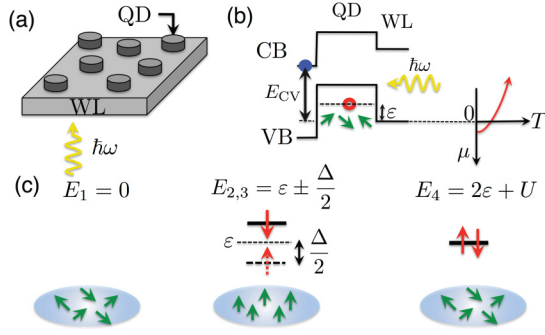


FIG. 1. (Color online) (a) A scheme of QDs grown on a wetting layer (WL), which is excited by light (cladding layer not shown). Type-II conduction/valence band (CB/VB) profile of a II-VI QD doped with Mn spins (green).  $\varepsilon$  and  $E_{CV}$  are the confinement and band gap energies. Hole quasi-Fermi level  $\mu(T)$  lies in the continuum of WL states for  $T = 0$ . (c) QD states with corresponding energies, occupancies, and Mn-spin alignment:  $E_1$  state has zero energy and QD occupancy;  $E_{2,3}$  state has one hole with spin  $+3/2$  ( $-3/2$ ),  $\Delta$  is the exchange splitting; and  $E_4$  has two holes with a repulsive Coulomb energy  $U$ .

$Z_{\text{sys}}(\xi_{\text{MF}}) = Z_{\text{Mn}}(\xi_{\text{MF}})Z_h(\xi_{\text{MF}})$  is expressed in terms of the Mn and hole contributions:  $Z_{\text{Mn}}(\xi_{\text{MF}}) = e^{-G_{\text{Mn}}(\xi_{\text{MF}})/k_B T}$ ,  $Z_h(\xi_{\text{MF}}) = e^{-F_h(\xi_{\text{MF}})/k_B T}$ . We obtain

$$E_{\text{MP}} = -\frac{\Delta_{\text{max}}\xi_{\text{MF}}}{Z_h(\xi_{\text{MF}})} e^{-(\varepsilon-\mu)/k_B T} \sinh\left(\frac{\Delta_{\text{max}}\xi_{\text{MF}}}{2k_B T}\right). \quad (5)$$

In Fig. 2, MF predicts multiple phase transitions and reentrant magnetism. For low-hole densities (solid line), the system exhibits a second-order phase transition at  $T_{C3} = 27$  K. In Fig. 2(b), the location of minimum  $G_{\text{sys}}(\xi)$  continuously goes to zero, a signature of second order transition. This is similar to the usual MP case (single particle), since for any  $T < 30$  K, the probability of finding a single hole in the QD is dominant. For high-hole densities (dotted line), MF

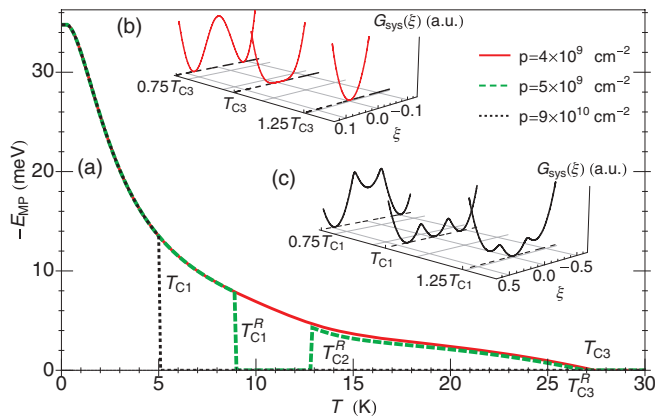


FIG. 2. (Color online) (a) Reentrant behavior in the MP energy,  $E_{\text{MP}}$  [see Eq. (5)], and various critical temperatures. Free energy evolution reveals the second- (b) and first-order (c) phase transitions for  $p = 4 \times 10^9$  and  $p = 9 \times 10^{10}$  cm $^{-2}$ , respectively. Parameters, partially guided by (Zn,Mn)Te/ZnSe QDs:<sup>4</sup>  $\varepsilon = -36$  meV,  $U = 30$  meV,<sup>16,33</sup>  $\Delta_{\text{max}} = 69$  meV (see Fig. 1),  $x_{\text{Mn}} = 2.6\%$ ,  $N_0\beta = -1.05$  eV, and  $m_h^* = 0.21$ . QD volume,  $\Omega$ , is cylindrical with a 50-Å radius and a 25-Å height.

predicts a first-order transition at  $T_{C1} = 5$  K, consistent with the discontinuous shift of the  $G_{\text{sys}}(\xi)$  minimum to  $\xi = 0$  at  $T_{C1}$  in Fig. 2(c). At  $T < T_{C1}$ , magnetism is present since the QD is occupied by 1, rather than 2 holes (despite  $\mu \sim 1$  meV in the continuum), according to  $E_2 - \mu < E_4 - 2\mu$  [see Fig. 1(c)]. This inequality is satisfied as long as the ordering of Mn-spins sufficiently lowers the energy of the single hole state  $E_2$ . When  $T > T_{C1}$ ,  $E_2 - \mu > E_4 - 2\mu$  and the QD becomes doubly occupied suppressing MP formation. Above  $T_{C1}$ , magnetism does not reappear since thermal Mn excitations completely quench magnetic order, before  $\mu$  approaches  $\varepsilon + U$  to promote single occupancy.

The QD exhibits reentrant magnetism at  $p = 5 \times 10^9$  cm $^{-2}$  (dashed line). For  $T < T_{C2}^R$ , the scenario is the same as the dotted line ( $T_{C1}^R$  plays the role of  $T_{C1}$ ). At  $T > T_{C2}^R$ , with  $E_2 - \mu < E_4 - 2\mu$ , the QD is singly occupied as a result of  $\mu$  moving quickly towards  $\varepsilon$ , thus MP reappears by a first-order transition. The transition at  $T = T_{C3}^R$  has the same origin as the solid curve. For  $T \gg T_{C3}^R$ , the dot becomes emptied.

However, MF theory neglects the possibility for the system to deviate from the equilibrium value  $\xi_{\text{MF}}$ . This leads to unphysical thermodynamic phase transitions in small systems. Could this also imply that the described reentrant magnetism is only an artifact of the MF theory? To address this question and better understand the validity of the behavior predicted at the MF level, we formulate a fluctuation approach (FA). Statistical fluctuations are included in the partition function by integrating over all possible values of the order parameter.<sup>12</sup> Correspondingly, we employ  $Z_{\text{sys}} = \int_{-1}^1 e^{-G_{\text{sys}}(\xi)/k_B T} d\xi$  to implement the framework used for MF [recall Eq. (5)], and obtain the average exchange energy

$$E_{\text{MP}} = -\frac{\Delta_{\text{max}}}{Z_{\text{sys}}} e^{-(\varepsilon-\mu)/k_B T} \int_{-1}^1 d\xi \xi e^{-G_{\text{sys}}(\xi)/k_B T} \times \sinh\left(\frac{\Delta_{\text{max}}\xi}{2k_B T}\right). \quad (6)$$

This  $E_{\text{MP}}$  for the GC ensemble is similar to that of the canonical ensemble.<sup>35</sup> However,  $Z_{\text{sys}}$  now contains multiple occupancies and the numerator is weighted by  $e^{\mu/k_B T}$ , which decreases with increasing  $T$  [see Fig. 1(b)].

We are now able to directly compare MF and FA results. The sharp MF phase transitions [ $E_{\text{MP}}$  in Fig. 2], become smeared out, as seen in Fig. 3. FA yields finite  $E_{\text{MP}}$  at any finite  $T$ . This is expected from averaging of  $G_{\text{sys}}$  over  $\xi$ , implicit in Eq. (6), including strong contributions from the competing local minima [Fig. 2(c) at  $T_{C1}$ ]. For example, MF reentrant magnetism from Figs. 2(a) and 3(b) for  $p = 5 \times 10^9$  cm $^{-2}$  is absent in FA (see Fig. 3), since the local minima of  $G_{\text{sys}}$  at  $|\xi| > 0$  contribute strongly in the temperature range of 9–13 K. Surprisingly, the dotted and solid curves in Fig. 3 show FA reentrant  $E_{\text{MP}}$  even at room temperature, while for the same  $p$  no MF reentrant behavior was seen. In FA, the increase in  $|E_{\text{MP}}|$  at higher  $T$  is due to the the QD occupancy decreasing from 2 to 1 holes. The maximum reentrant  $|E_{\text{MP}}(T)|$  coincides with the average occupancy of 1. Even though inclusion of statistical fluctuations removes reentrance for some hole densities, it also yields a smoothed version of the same effect for higher  $T$ , where MF predicts

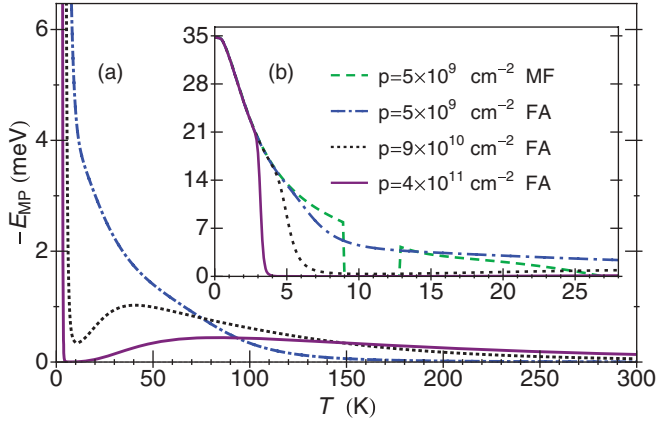


FIG. 3. (Color online) (a) Reentrant magnetism in the fluctuation approach (FA) [see Eq. (6)] for  $p = 5 \times 10^9$ ,  $9 \times 10^{10}$ ,  $4 \times 10^{11}$   $\text{cm}^{-2}$ . (b) Comparison of the FA and mean field (MF) FA (dot-dashed line) shows no reentrant magnetism predicted for MF and  $p = 5 \times 10^9$   $\text{cm}^{-2}$  (dashed line). Other QD parameters are from Fig. 2.

$E_{\text{MP}} = 0$ . A peculiar nonmonotonic  $E_{\text{MP}}(T)$  is therefore not limited to the MF description.

Furthermore, the reentrant MP is not restricted to the above parameters, but occurs for a range of  $x_{\text{Mn}}$ ,  $\varepsilon$ ,  $U$ , and  $p$ . For example, we find reentrant  $E_{\text{MP}}$  for  $1.5 \times 10^{10} \text{ cm}^{-2} \leq p \leq 2 \times 10^{12} \text{ cm}^{-2}$ , with other parameters fixed. Conversely, the reentrant MP is present for  $1.5\% \leq x_{\text{Mn}} \leq 2.6\%$ , if the remaining parameters are fixed.

We next discuss how reentrant magnetism could be observed in PL experiments. To obtain the PL spectrum, we assume low QD density and sufficiently high intensity of exciting light for two CB electrons to always be in the vicinity of each QD, occupying the lowest possible energy  $\sim E_{\text{CV}} > 0$ <sup>36</sup> (see Fig. 1) and with opposite spins. The total spectrum  $I_{\text{tot}}$  is the superposition of the lines generated by the  $2 \rightarrow 1$  and  $1 \rightarrow 0$  transitions:

$$I_{\text{tot}}(\omega) = I_{1 \rightarrow 0}(\omega) + I_{2 \rightarrow 1}(\omega). \quad (7)$$

We assume that the Mn-configuration does not change during a recombination event. The intensity of each PL line is  $I = \sum_{i,f} p_i w_{if} \delta[\hbar\omega - (E_i - E_f)]$ , where  $w_{if}$  is the transition rate,  $p_i$  is the thermodynamic probability of the initial state,  $\hbar\omega$  is the energy of the emitted photon, and  $E_f$  ( $E_i$ ) is the energy of the final (initial) state of the system. We replace the above  $\sum$  with  $\int d\xi$ . For  $1 \rightarrow 0$  transitions, the system is in an initial state with a hole of spin up (down), which later recombines with a spin-down (up) electron.<sup>32</sup> The intensity of this line is

$$I_{1 \rightarrow 0}(X) = c(T) e^{-(\varepsilon - \mu)/k_B T} e^{-X/k_B T} \times e^{-G_{\text{Mn}}(2X/\Delta_{\text{max}})/k_B T} \theta(\Delta_{\text{max}} - |2X|). \quad (8)$$

Here,  $X(\omega) = \hbar\omega - E_{\text{CV}} - \varepsilon$  is the shifted frequency,  $c(T) \propto (\Omega/\beta) |d_{\text{cv}}|^2 / Z_{\text{sys}}$ ,  $d_{\text{cv}}$  the dipole matrix element, and  $\theta$  is the step function. For the  $2 \rightarrow 1$  transition, there are two holes and two electrons of opposite spin in the initial state. The

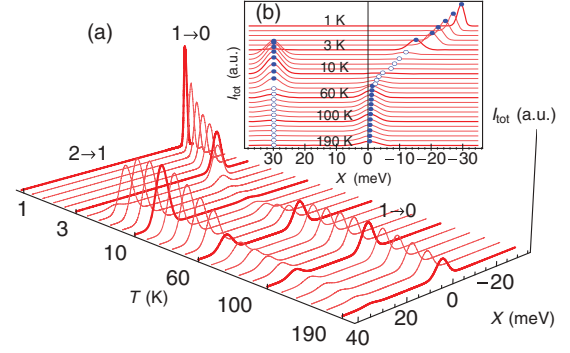


FIG. 4. (Color online) (a) A PL spectrum for reentrant magnetism showing  $2 \rightarrow 1$  and  $1 \rightarrow 0$  hole occupancy transitions ( $p = 9 \times 10^{10} \text{ cm}^{-2}$ ). The thick lines emphasize important features in the PL spectrum, while the thin lines between them show their evolution at intermediate equidistant  $T$ . (b) Overall PL peak position marked with filled circles. As  $T$  increases,  $1 \rightarrow 0$  peak shifts to zero energy. QD parameters are from Fig. 2.

intensity is

$$I_{2 \rightarrow 1}(X) = c(T) e^{-(2\varepsilon + U - 2\mu)/k_B T} e^{-G_{\text{Mn}}(2(X-U)/\Delta_{\text{max}})/k_B T} \times \theta(\Delta_{\text{max}} - 2|X - U|). \quad (9)$$

In Fig. 4, the PL spectrum shows the evolution of the peaks for transitions  $1 \rightarrow 0$ , centered at  $X < 0$ , and  $2 \rightarrow 1$ , centered at the charging energy,  $X = U$ , since  $\hbar\omega = E_i - E_f = (2\varepsilon + U + E_{\text{CV}}) - \varepsilon$ .<sup>37</sup> From Eqs. (3), (8), and (9) it follows that the Mn-contribution to the PL is  $T$  independent: the amplitude of the  $1 \rightarrow 0$  ( $2 \rightarrow 1$ ) peak at different  $T$  is proportional to the probability of finding a single (double) occupied QD. For  $T < 5$  K, the  $1 \rightarrow 0$  line dominates, while it becomes negligible at  $\sim 10$  K where the system is virtually nonmagnetic and the double occupied state ( $2 \rightarrow 1$  line) is dominant. At higher  $T$ , due to the shift of  $\mu$  toward CB, the probability of single occupancy increases, and for  $T > 100$  K, the probability of zero occupancy increases. The resulting  $T$  dependencies are remarkably nonmonotonic for both  $I_{\text{tot}}$  peak position (red- and blue shifts) and  $I_{\text{tot}}$  peak intensity; a signature of reentrant MPs, consistent with  $E_{\text{MP}}(T)$  in Fig. 3.

What are the semiconductor systems where the reentrant MP could be found? Recently, a nonmonotonic PL redshift was observed in type-II (Zn,Mn)Te/ZnSe QDs,<sup>4</sup> which have partially guided our choice of parameters. However, the reentrant magnetism should not be limited to type-II systems. The necessary condition is the  $T$ -dependent multiple occupancy, readily seen in both type-II<sup>16,17</sup> and type-I QDs.<sup>15,38</sup> Multiple occupancy can be reached by raising photo-excitation intensity, which may first lead to weakening of MPs (blue shift) through Mn-spin heating.<sup>22,39-41</sup> Nevertheless, an increased blue shift was attributed to double occupancy in type-I magnetic QDs.<sup>39</sup>

Considering only a reduced  $T$  range could conceal the presence of reentrant MP. An initial steep decline in  $E_{\text{MP}}(T)$  (see Fig. 3) is similar to conventional MPs, while a slightly higher  $T$  region (e.g., 5 to 30 K for solid line in Fig. 3) could be misinterpreted as a final thermal breakup of MPs. Thus further experimental studies of power and  $T$  dependence are important. Specifically, it would be desirable to consider single

self-assembled QDs<sup>8,42</sup> to reduce uncertainties due to inhomogeneous averaging, and focus on moderate  $x_{\text{Mn}}$  to suppress the Mn-Mn antiferromagnetic interactions.<sup>4</sup> Colloidal (II,Mn)VI QDs<sup>3,10</sup> showing a robust MP formation are also promising candidates to test some of our predictions.

Even though in this work we have only focused on reentrant magnetism, we expect that further studies of the implications of multiple occupancies will lead to additional surprises in

both epitaxial and colloidal QDs. Since prior findings in QDs were successfully applied to different finite fermion systems,<sup>43</sup> it may be possible to seek other promising paths for observing reentrant magnetism.

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- <sup>1</sup>T. Gurung, S. Mackowski, G. Karczewski, H. E. Jackson, and L. M. Smith, *Appl. Phys. Lett.* **93**, 153114 (2008).
- <sup>2</sup>A. G. Petukhov and M. Foygel, *Phys. Rev. B* **62**, 520 (2000).
- <sup>3</sup>R. Beaulac, L. Schneider, P. I. Archer, G. Bacher, and D. R. Gamelin, *Science* **325**, 973 (2009).
- <sup>4</sup>I. R. Sellers, R. Oszwaldowski, V. R. Whiteside, M. Eginligil, A. Petrou, I. Žutić, W.-C. Chou, W. C. Fan, A. G. Petukhov, S. J. Kim, A. N. Cartwright, and B. D. McCombe, *Phys. Rev. B* **82**, 195320 (2010).
- <sup>5</sup>F. Xiu, Y. Wang, J. Kim, P. Upadhyaya, Yi Zhou, X. Kou, W. Han, R. K. Kawakami, J. Zou, and K. L. Wang, *ACS Nano* **4**, 4948 (2010), and references therein.
- <sup>6</sup>J. Fernández-Rossier and L. Brey, *Phys. Rev. Lett.* **93**, 117201 (2004).
- <sup>7</sup>A. O. Govorov, *Phys. Rev. B* **72**, 075359 (2005); W. Zhang, T. Dong, and A. O. Govorov, *ibid.* **76**, 075319 (2007).
- <sup>8</sup>C. Le Gall, A. Brunetti, H. Boukari, and L. Besombes, *Phys. Rev. Lett.* **107**, 057401 (2011).
- <sup>9</sup>R. M. Abolfath, P. Hawrylak, and I. Žutić, *Phys. Rev. Lett.* **98**, 207203 (2007); R. M. Abolfath, A. G. Petukhov, and I. Žutić, *ibid.* **101**, 207202 (2008); N. T. T. Nguyen and F. M. Peeters, *Phys. Rev. B* **78**, 045321 (2008); Y. G. Semenov and K. W. Kim, *ibid.* **72**, 195303 (2005); N. Lebedeva, A. Varpula, S. Novikov, and P. Kuivalainen, *ibid.* **81**, 235307 (2010); L. Villegas-Lelovsky, F. Qu, L. O. Massa, V. Lopez-Richard, and G. E. Marques, *ibid.* **84**, 075319 (2011); S.-J. Cheng, *ibid.* **77**, 115310 (2008).
- <sup>10</sup>D. A. Bussian, S. A. Crooker, M. Yin, M. Brynda, A. L. Efros, and V. I. Klimov, *Nat. Mater.* **8**, 35 (2009).
- <sup>11</sup>D. R. Yakovlev and W. Ossau, in *Introduction to the Physics of Diluted Magnetic Semiconductors*, edited by J. Kossut and J. A. Gaj (Springer, Berlin, 2010).
- <sup>12</sup>T. Dietl and J. Spáček, *Phys. Rev. B* **28**, 1548 (1983).
- <sup>13</sup>P. A. Wolff, in *Semiconductors and Semimetals*, edited by J. K. Furdyna and J. Kossut (Academic Press, San Diego, 1988), Vol. 25.
- <sup>14</sup>E. L. Nagaev, *Physics of Magnetic Semiconductors* (MIR Publishers, Moscow, 1983).
- <sup>15</sup>A. V. Chernenko, A. S. Brichkin, S. V. Sokolov, and S. V. Ivanov, *Phys. Status Solidi B* **247**, 1514 (2010).
- <sup>16</sup>K. Matsuda, S. V. Nair, H. E. Ruda, Y. Sugimoto, T. Saiki, and K. Yamaguchi, *Appl. Phys. Lett.* **90**, 013101 (2007).
- <sup>17</sup>B. Bansal, S. Godefroy, M. Hayne, G. Medeiros-Ribeiro, and V. V. Moshchalkov, *Phys. Rev. B* **80**, 205317 (2009).
- <sup>18</sup>B. Fisher, J. M. Caruge, D. Zehnder, and M. Bawendi, *Phys. Rev. Lett.* **94**, 087403 (2005).
- <sup>19</sup>L. Besombes, Y. Leger, L. Maingault, D. Ferrand, H. Mariette, and J. Cibert, *Phys. Rev. B* **71**, 161307(R) (2005).
- <sup>20</sup>A. G. Petukhov, I. Žutić, and S. C. Erwin, *Phys. Rev. Lett.* **99**, 257202 (2007).
- <sup>21</sup>In the bulk, reentrant magnetism could be possible due to large thermally excited carrier density [Ref. 20, M. J. Calderon and S. Das Sarma, *Phys. Rev. B* **75**, 235203 (2007), and experiments of V. Krivoruchko, V. Tarenkov, D. Varyukhin, A. Dyachenko, O. Pashkova, and V. Ivanov, *J. Magn. Magn. Mater.* **322**, 915 (2010)] or spin glass formation [C. K. Thomas, D. A. Huse, and A. A. Middleton, *Phys. Rev. Lett.* **107**, 047203 (2011)].
- <sup>22</sup>A. A. Maksimov, G. Bacher, A. McDonald, V. D. Kulakovskii, A. Forchel, C. R. Becker, G. Landwehr, and L. W. Molenkamp, *Phys. Rev. B* **62**, R7767 (2000).
- <sup>23</sup>J. Seufert, G. Bacher, M. Scheibner, A. Forchel, S. Lee, M. Dobrowolska, and J. K. Furdyna, *Phys. Rev. Lett.* **88**, 027402 (2001).
- <sup>24</sup>S. Takeyama, S. Adachi, Y. Takagi, and V. F. Aguekian, *Phys. Rev. B* **51**, 4858 (1995).
- <sup>25</sup>I. Žutić, J. Fabian, and S. Das Sarma, *Rev. Mod. Phys.* **76**, 323 (2004); J. Fabian, A. Matos-Abiague, Ch. Ertler, P. Stano, and I. Žutić, *Acta Phys. Slov.* **57**, 565 (2007); *Concepts in Spin Electronics*, edited by S. Maekawa (Oxford University Press, Oxford, 2006).
- <sup>26</sup>For simplicity, we consider here only heavy holes.
- <sup>27</sup>R. Heitz, M. Veit, N. N. Ledentsov, A. Hoffmann, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, and Zh. I. Alferov, *Phys. Rev. B* **56**, 10435 (1997); I. V. Ignatiev, I. E. Kozin, S. V. Nair, H. W. Ren, S. Sugou, and Y. Masumoto, *ibid.* **61**, 15633 (2000); M. Funato, K. Omae, Y. Kawakami, S. Fujita, C. Bradford, A. Balocchi, K. A. Prior, and B. C. Cavenett, *ibid.* **73**, 245308 (2006); S. Mackowski, G. Prechtel, W. Heiss, F. V. Kyrychenko, G. Karczewski, and J. Kossut, *ibid.* **69**, 205325 (2004); The recombination time is even longer in type-II QDs.<sup>4</sup> F. Hatami, M. Grundmann, N. N. Ledentsov, F. Heinrichsdorff, R. Heitz, J. Böhrer, D. Bimberg, S. S. Ruvimov, P. Werner, V. M. Ustinov, P. S. Kop'ev, and Z. I. Alferov, *ibid.* **57**, 4635 (1998).
- <sup>28</sup>J. K. Furdyna, *J. Appl. Phys.* **64**, R29 (1988).
- <sup>29</sup>A. Golnik, J. Ginter, and J. A. Gaj, *J. Phys. C* **16**, 6073 (1983). A uniform wave function is a good approximation. When carrier localization effects are considered, using a different wave function and the canonical ensemble, we find at most a 15 % change in  $E_{\text{MP}}$  at  $T = 5$  K.
- <sup>30</sup>R. Kubo, *Statistical Mechanics* (North-Holland, Amsterdam, 1960).
- <sup>31</sup>We have determined that the effects of magnetic bipolarons, described in R. Oszwaldowski, I. Žutić, and A. G. Petukhov, *Phys. Rev. Lett.* **106**, 177201 (2011), are only important at very low  $T$ , much below the nonmagnetic doubly occupied regime. All our main findings here are thus not altered.

- <sup>32</sup>P. Wojnar, J. Suffczyński, K. Kowalik, A. Golnik, G. Karczewski, and J. Kossut, *Phys. Rev. B* **75**, 155301 (2007).
- <sup>33</sup>A much larger  $U$  would allow only one carrier to be confined and would require a simpler form of  $F_h$  in Eq. (4).
- <sup>34</sup>A. Bhattacharjee, *Solid State Commun.* **57**, 31 (1986).
- <sup>35</sup>M. Umehara, *Phys. Rev. B* **61**, 12209 (2000).
- <sup>36</sup>This quantity may be  $T$  dependent owing to the Varshni shift; G. Bacher, A. A. Maksimov, H. Schömig, V. D. Kulakovskii, M. K. Welsch, A. Forchel, P. S. Dorozhkin, A. V. Chernenko, S. Lee, M. Dobrowolska, and J. K. Furdyna, *Phys. Rev. Lett.* **89**, 127201 (2002).
- <sup>37</sup>This order of peaks is typical for type-II QDs.<sup>16,17</sup>
- <sup>38</sup>In type-I QDs, CB and VB extrema coincide spatially.
- <sup>39</sup>H. Schömig, G. Bacher, A. Forchel, S. Lee, M. Dobrowolska, and J. K. Furdyna, *J. Supercond.* **16**, 379 (2003).
- <sup>40</sup>G. Bacher, H. Schömig, M. Scheibner, A. Forchel, A. A. Maksimov, A. V. Chernenko, P. S. Dorozhkin, V. D. Kulakovskii, T. Kennedy, and T. L. Reinecke, *Physica E* **26**, 37 (2005).
- <sup>41</sup>L. Worschech, T. Schmidt, A. Forchel, T. Slobodskyy, G. Schmidt, and L. W. Molenkamp, *Semicond. Sci. Technol.* **23**, 114018 (2008).
- <sup>42</sup>Ł. Kłopotowski, Ł. Cywiński, P. Wojnar, V. Voliotis, K. Fronc, T. Kazimierczuk, A. Golnik, M. Ravaro, R. Grousson, G. Karczewski, and T. Wojtowicz, *Phys. Rev. B* **83**, 081306(R) (2011).
- <sup>43</sup>S. M. Reimann and M. Manninen, *Rev. Mod. Phys.* **74**, 1283 (2002); C. Yannouleas and U. Landman, *Rep. Prog. Phys.* **70**, 2067 (2007).