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Spin and energy transfer in nanocrystals without tunneling

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We describe a mechanism of spin transfer between individual quantum dots without tunneling. Incident circularly polarized photons create interband excitons with nonzero electron spin in the first quantum dot. When the quantum-dot pair is properly designed, this excitation can be transferred to the neighboring dot via the Coulomb interaction with *conservation* or *flipping* of electron spin. The second dot can radiate circularly polarized photons at smaller energy. Selection rules for spin transfer are determined by the resonant conditions and by the strong spin-orbit interaction in the valence band of nanocrystals. Coulomb-induced energy and spin transfer in pairs and chains of dots can become very efficient under resonant conditions. The electron can preserve its spin orientation even in randomly oriented nanocrystals.

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Manipulation of spins in nanostructures is presently attracting a tremendous amount of interest.[1–3] Since spins in solids have relatively long lifetimes, they can be exploited as qubits—basic elements of quantum computers.[4] Spin-polarized states of electrons in crystals can be generated optically[5–7] by driving current through spin-dependent barriers or by injecting electrons from ferromagnetic materials.[1,2] In most cases, spin transport across a crystal occurs either via tunneling or injection. This would not be the case for so-called colloidal quantum dots (QD's), where individual nanocrystals strongly confine carriers and do not permit efficient tunnel coupling.[8–10] However, instead of direct tunnel coupling, the colloidal QD's permit long-range Coulomb-induced transfer of optically excited excitons.[8–10] Such transport has been observed in several recent experiments and is often referred to as Förster energy transfer.[11] Theoretically, Förster-like transfer in nanocrystals has been discussed in connection with exciton dynamics in QD arrays and quantum computing.[12]

Here we develop a theory of electron spin transfer between individual nanocrystals without tunneling, involving optically excited excitons and the Coulomb interaction. So far, spin transport in nanostructures has been considered almost exclusively in relation to direct transport of charge.[1,2] Since the spin orientation in the conduction band of semiconductors can be efficiently created with the circularly polarized light pumping,[5,6] it is interesting to study the possibility of spin transfer between individual dots without transfer of charge. In such a transfer process, the optical and spin selection rules would be dictated by the strong spin-orbit interaction in the valence band. The typical experimental scheme related to Förster transport involves pairs of quantum dots with different sizes [Figs. 1(a)–1(c)]. An incident photon creates an exciton in the small dot 1 with a larger optical gap [Figs. 1(a)–1(c)]. Then, the exciton is transferred via the Förster-like mechanism into the large dot 2 with a smaller optical gap. Due to fast energy relaxation in dot 2, the exciton becomes trapped and contribute to the photoluminescence (PL) at the dot-2 energy. If electrons in dot 1 are created by circularly polarized light, they become spin polarized due to the spin-orbit interaction in the valence band.[5,6] Here we will focus on the dynamics of excitons generated by circularly polarized photons and develop principles for electron-spin transport in QD pairs without tunneling. We will show that the spin orientation can be efficiently transported between QD's via the Coulomb interaction. This becomes possible thanks to the strong spin-orbit interaction in the valence bands of QD's. The spin-transfer selection rules strongly depend on geometry and resonance conditions. In the resonance regime, the transfer can lead to either conservation or flipping of spin.

Pairs of semiconductor QD's can be grown by using self-organization technology.[13] In such stacked QD's, the sizes of dots and interdot separation are well controlled. Another method to fabricate a system with QD pairs is colloidal synthesis.[8–10] In a solid of colloidal QD's with two distinct sizes, QD pairs are randomly oriented.[8,9] In monolayers of QD's [Fig. 3(a)], the orientation of pairs is directional.[9,10] Another possibility to avoid a randomness is to study a single QD pair bound to a surface.[10,14]

In what follows, we will use several simplifications related to the time scales. In particular, we will assume that $\tau_{e\text{-}spin}, \tau_{exc} \gg \tau_{energy}, \tau_{h\text{-}spin}$, where τ_{exc} is the exciton lifetime in a single QD related to radiational and nonradiational transitions, τ_{energy} is the energy relaxation time of excitons within a dot, $\tau_{e\text{-}spin}$ is the electron spin lifetime, and $\tau_{h\text{-}spin}$ is the momentum relaxation time of holes. In other words, we suppose (1) fast intradot relaxation of the angular momentum of holes and (2) fast energy relaxation to the ground state in the dots.

Disk-shaped dots with a cubic lattice. First we consider a pair of oblate (disk-shaped) quantum dots (Fig. 1) with dimensions $a_i \ll b_i$, where a_i is the QD size in the z direction, b_i is the in-plane diameter, and i is the dot index ($i=1,2$). For simplicity, we assume that the QD potential has infinite walls. In such a model, a single QD is quasi two dimensional (2D) and its valence-band structure is similar to that in a 2D quantum well.[15] To find the wave functions, we first quantize the motion of heavy and light holes in the z direction; it provides us with the Bloch functions. Then we can introduce weak quantization in the x-y plane involving effective masses of holes. The wave functions in the conduction and valence bands for the dots 1 and 2 take a form

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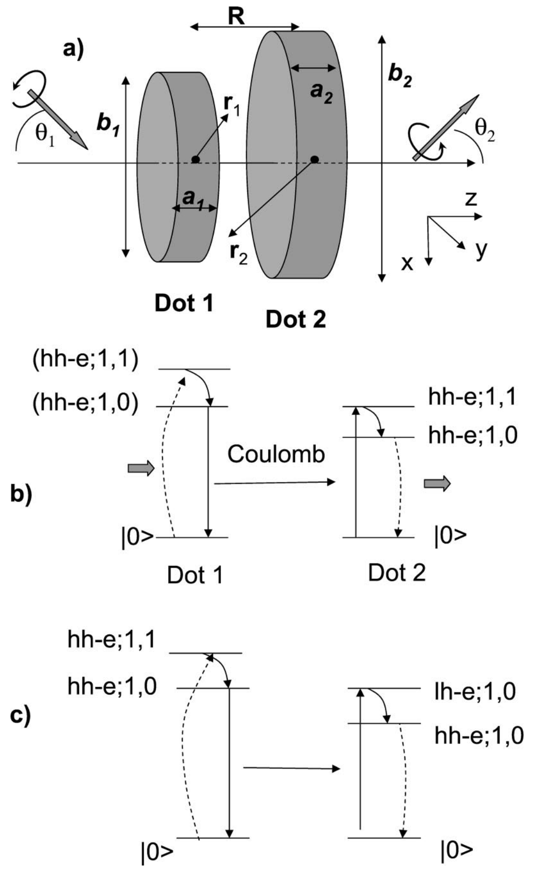

FIG. 1. Sketch of a quantum-dot molecule (a). Energy diagrams of intradot and interdot transitions in the transfer process (b), (c); the label $(\gamma-e;n,l)$ denotes the exciton composed of hole γ and electron, where γ can be *hh* or *lh*, and (n,l) are envelope-function indices.

$$\Psi_i^{e,\uparrow(\downarrow),n,l}=u_{\uparrow(\downarrow)}\Phi_{n,l}^{(i)}(r_{||},z),$$

$$\Psi_i^{hh,\pm3/2,n,l}=u_{\pm3/2}\Phi_{n,l}^{(i)}(r_{||},z),$$

$$\Psi_i^{lh,\pm1/2,n.l}=u_{\pm1/2}\Phi_{n,l}^{(i)}(r_{||},z), \tag{1}$$

where $i=1,2$ and $\mathbf{r}_{||}=(x,y)$; $u_{\uparrow(\downarrow)}$, $u_{\pm3/2}$, and $u_{\pm1/2}$ are the Bloch functions of electrons, heavy holes (*hh*), and light holes (*lh*), respectively; $\Phi_{n,l}^{(i)}(r_{||},z)=f_0^{(i)}(z)R_{n,l}^{(i)}(r_{||})$ are the envelope functions, where the $f_0^{(i)}(z)$ is the ground-state function for motion in the *z* direction, $R_{n,l}^{(i)}(r_{||})$ are Bessel's functions describing the in-plane motion, and (n,l) are the radial and azimuthal quantum numbers of the in-plane motion, respectively; $n=1,2,\ldots$ and $l=0,\mp1,\pm2,\ldots$. In our simplified approach all types of carriers are described with the same set of envelope wave functions $\Phi_{n,l}(r_{||},z)$.

In the geometry shown in Fig. 1, the optical operator for the exciton in dot 1 can be written as follows: $\hat{V}_{1,+}^{opt}=\mathbf{e}_1\hat{\mathbf{p}}=\cos(\theta_1)\hat{p}_x+i\hat{p}_y+\sin(\theta_1)\hat{p}_z$, where $\hat{\mathbf{p}}$ and $\mathbf{e}_1$ are the momentum operator and polarization vector, respectively. Using this operator, the probability of interband optical transitions for dot 1 takes a form $P_1^{s,\mu}=|\langle\Psi_1^{s,n,l}|\hat{V}_{1,+}^{opt}|\Psi_1^{\mu,n,l}\rangle|^2$, where $s=\uparrow(\downarrow)$ and $\mu=\pm3/2,\pm1/2$. Emission of dot 2 is described in a similar way with the operator $\hat{V}_{2_\pm}^{opt}=\cos(\theta_2)\hat{p}_x\pm i\hat{p}_y-\sin(\theta_2)\hat{p}_z$, where the signs ± relate to the different polarizations of the secondary photon $(\mp\hbar)$. For simplicity, we consider the case when the linear momenta of both photons lie in the *x*-*z* plane.

The interdot transfer is described by the Coulomb operator which can be expanded into an infinite series of multipole terms. However, it is natural to assume that the dipole-dipole interaction will provide the leading term,

$$\hat{V}_{Coul}=\frac{e^2}{\epsilon R^3}(\mathbf{r}_1\mathbf{r}_2-3z_1z_2), \tag{2}$$

where $\mathbf{r}_{1(2)}$ are the radius vectors related to the dots [Fig. 1(a)], ϵ is the averaged dielectric constant, and R is the distance between the dots. Below, we will generalize our results including multipole interactions. The Förster-like probability of an interdot transition takes the form

$$W_{\beta_1}=\frac{2\pi}{\hbar}\sum_{\beta_2}|\langle\beta_1|\hat{V}_{Coul}|\beta_2\rangle|^2\delta(E_{\beta_1}-E_{\beta_2}), \tag{3}$$

where the indices $\beta_{1(2)}$ denote the exciton states in the dots: $\beta_1=(s_1,\mu_1,n_1,l_1)$ and $\beta_2=(\mu_2,s_2,n_2,l_2)$. Because of fast intradot energy relaxation, the function $|\beta_1\rangle$ in Eq. (3) describes the ground-state exciton in dot 1 with $s_1=\uparrow,\downarrow$ and $\mu_1=\pm3/2$, and $(n_1,l_1)=(1,0)$. In the spirit of Förster theory the δ function in Eq. (3) should be replaced by the spectral overlap integral $J_{\beta_1,\beta_2}=\int\rho_{\beta_1}(E)\rho_{\beta_2}(E)dE$ which involves normalized line shapes $\rho_{\beta_i}(E)=\pi^{-1}\Gamma_{\beta_i}/[(E-E_{\beta_i})^2+\Gamma_{\beta_i}^2]$, where Γ_{β_i} is the homogeneous broadening of the exciton β_i. Lorentzians were utilized for simplicity.

By using Eqs. (1)–(3), we now compute the mean spin in the dots and the degree of polarization of secondary photons. To be specific, we consider the resonant dipole-allowed absorption process of incident photons in dot 1 that involves a heavy-hole level [Fig. 1(b)]; in other words, the incident-photon energy is taken below the first interband transition related to the light hole. The mean *z* component of electron spin polarization in dot 1 is determined by the probabilities $P_1^{s,\mu}$ and is equal to $S_1=(P_1^{\uparrow}-P_1^{\downarrow})/(P_1^{\uparrow}+P_1^{\downarrow})=-2\cos(\theta_1)/[\cos(\theta_1)^2+1]$, where $P_i^{\uparrow(\downarrow)}\propto|P_{cv}|^2[\cos(\theta_1)\mp1]^2$ is the probability of the electron being in the state ↑(↓) and $P_{cv}=\langle S|\hat{p}_x|X\rangle$ is the interband optical matrix element. In the optical matrix elements, the operator $\hat{\mathbf{p}}$ was involved only in integrals with Bloch functions. For the next step, we calculate the Coulomb matrix elements under resonance conditions. In the regime of interdot resonance, the ground-state exciton energy of dot 1 is equal to the energy of the excited dipole-active exciton in dot 2. The latter state can be composed of either a heavy hole or light hole. We start with the resonance between heavy-hole states in the dots [Fig. 1(b)]. The probability to create an exciton with $s_2=\uparrow$ in dot 2 is given by $P_2^{\uparrow}=(1/2)P_1^{\uparrow}W_{\beta_1\rightarrow\beta_2}=P_1^{\uparrow}w_0J_{\beta_1,\beta_2}/2$, where $\beta_1=(\uparrow,3/2,1,0)$ and $\beta_2=(\uparrow,3/2,n_2,l_2)$; the factor of 1/2 is the probability to find a heavy hole in either state $(\pm3/2)$ in dot 1; this is due to fast momentum relaxation of holes.

Besides, $W_{\beta_1 \to \beta_2}$ is the probability of Förster-like transfer between the states β_1 and β_2. A coefficient $w_0 = 2\pi d_0^4 (e^4/\epsilon^2 R^6 \hbar)$, where $d_0 = \langle X|x|S\rangle$ is the atomic dipole moment. For spin ↓ we have a similar equation $P_2^\downarrow = P_1^\downarrow w_0 J_{\beta_1,\beta_2}/2$. Again, the operator **r** was involved only in integrals with Bloch functions. The spin polarization of dot 2 is given by

$$S_2' = \frac{P_2^\uparrow - P_2^\downarrow}{P_2^\uparrow + P_2^\downarrow} = S_1 = -\frac{2\cos(\theta_1)}{[\cos(\theta_1)^2 + 1]}. \tag{4}$$

If $\theta_1 = 0$, the system has axial symmetry, the transfer process conserves the total momentum, and therefore $S_2' = -1$. Thus, Förster transport *preserves spin polarization* in the regime of interdot resonance between heavy-hole levels. Now we assume that the parameters of dots are chosen to satisfy the condition of interdot resonance between heavy and light holes [Fig. 1(c)]. It is easy to see that Coulomb transfer results in *spin flipping*. For example, the probability $P_2^\uparrow$ is now expressed via $P_1^\downarrow$: $P_2^\uparrow = (1/2) P_1^\downarrow W_{\beta_1 \to \beta_2} = P_1^\downarrow w_0 J_{\beta_1,\beta_2}/6$, where $\beta_2 = (\uparrow, 1/2, n_2, l_2)$. Similarly, $P_2^\downarrow = P_1^\uparrow w_0 J_{\beta_1,\beta_2}/6$. Thus, we obtain the effect of spin flipping:

$$S_2'' = -S_1. \tag{5}$$

So far, we considered strongly resonant conditions. In the general case, the mean spin in dot 2 is calculated as

$$S_2 = \frac{\sum_{\beta_1,\beta_2} P_2^\uparrow(\beta_1 \to \beta_2) - P_2^\downarrow(\beta_1 \to \beta_2)}{\sum_{\beta_1,\beta_2} P_2^\uparrow(\beta_1 \to \beta_2) + P_2^\downarrow(\beta_1 \to \beta_2)}, \tag{6}$$

where the summation involves all pairs of states; the index β_1 is related to the *hh* ground state of dot 1: $\beta_1 = (\uparrow(\downarrow), \pm 3/2, 1, 0)$. The degree of circular polarization of secondary photons at the dot-2 ground-exciton energy is now written as

$$P_{circ} = \frac{I_+ - I_-}{I_+ + I_-} = -S_2 \frac{2\cos(\theta_2)}{[\cos(\theta_2)^2 + 1]}, \tag{7}$$

where $I_\pm$ are the light intensities given by $I_+ = P_2^\uparrow P_+(\uparrow) + P_2^\downarrow P_+(\downarrow)$ and $I_- = P_2^\uparrow P_-(\uparrow) + P_2^\downarrow P_-(\downarrow)$. Here, the optical transition rate $P_\sigma(s)$ describes the emission process in which an electron with spin s in dot 2 creates a photon with circular polarization σ, where σ can be + or −. The degree of circular polarization (7) strongly depends on the resonance conditions between the QD's [Fig. 2(a)]. If $\theta_{1(2)} = 0$, the system has axial symmetry and the electron spin is either conserved or flipped in the resonant-transfer process [Fig. 2(a)]. The latter comes from conservation of the total angular momentum in the Coulomb matrix elements. Besides, the rate of exciton transfer, $1/\tau_{trans} = W_{\beta_1}$, is strongly enhanced under the interdot resonance conditions [Fig. 2(b)]. Note that the total angular momentum is not conserved in the three-step process shown in Figs. 1(b) and 1(c) because of fast relaxation of angular momentum for the hole.

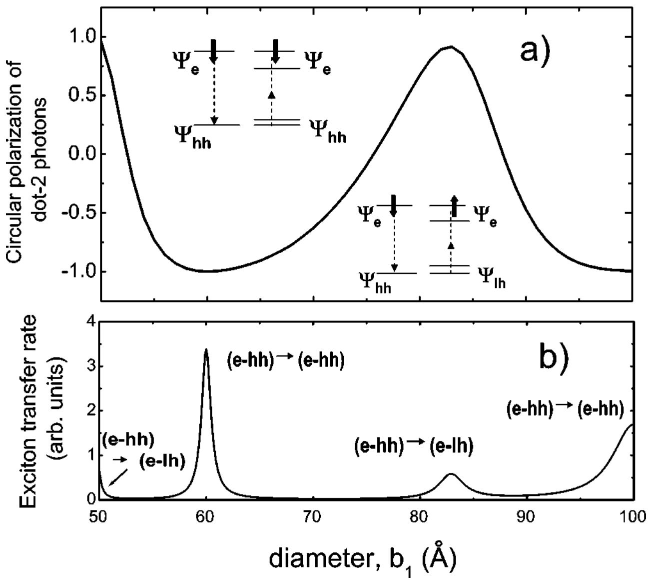

FIG. 2. (a) Calculated degree of circular polarization of photons emitted by dot 2 as a function of the dot-1 diameter; $\theta_{1(2)} = 0$. The sizes of dot 2 are kept constant, whereas the diameter of dot 1 is varied. (b) Calculated rate of exciton transfer from dot 1 into dot 2. Insets show diagrams of interband transitions. The crystal parameters correspond to InP quantum dots; effective masses: $m_e = 0.077 m_0$, $m_{lh} = 0.12 m_0$, $m_{hh} = 0.6 m_0$; $R = 80$ Å, $a_1 = a_2 = 25$ Å, $b_2 = 100$ Å, and $50 < b_1 < 100$ Å. The low-temperature broadening of the ground state of excitons in dot 1 is taken as 1 meV; the broadening of all excited states in dot 2 is assumed to be 5 meV.

Spherical quantum dots with a cubic lattice. In spherical dots, the symmetry of a single QD is high and both heavy and light holes will contribute to the transfer rate for the given interdot resonance. The multicomponent wave functions for the holes in a model with infinite walls are well known:[16]

$$\Psi_i^M = \sum_{l,m,\mu} C_{l,m,\mu,M} R_l^{(i)}(r) Y_{lm}^{(i)}(\Omega) u_\mu. \tag{8}$$

Here i is the QD number ($i = 1,2$), M is the z component of the total angular momentum, $Y_{lm}^{(i)}(\Omega)$ are spherical harmonic functions, $R_l^{(i)}(r)$ are functions of radial motion,[16] and $\mu = \pm 1/2, \pm 3/2$. Calculation of the spin orientation in dots 1 and 2 is straightforward. The mean z components of spin in dots are written as $S_1 = -\cos(\theta_1)/2$ and $S_2 = S_1/2$. The degree of circular polarization of emitted light takes the form $P_{circ} = -S_2 \cos(\theta_2)/2 = \cos(\theta_1)\cos(\theta_2)/8$. At the angles $\theta_{1(2)} = 0$, the polarization of emitted light is maximal and equal to $P_{circ} = 1/8$. The degree of polarization, 1/8, appears as a result of the three-step process. According to the theory of spin orientation in 3D crystals, the degree of polarization in the two-step process is 1/4.[5] Since the band structure of cubic spherical dots is isotropic, electron spin transfer does not depend on the type of interdot resonance and the electron spin is not flipped.

Oblate quantum dots with cubic and wurtzite lattices. Quantum dots can be anisotropic due to both shape and crys-

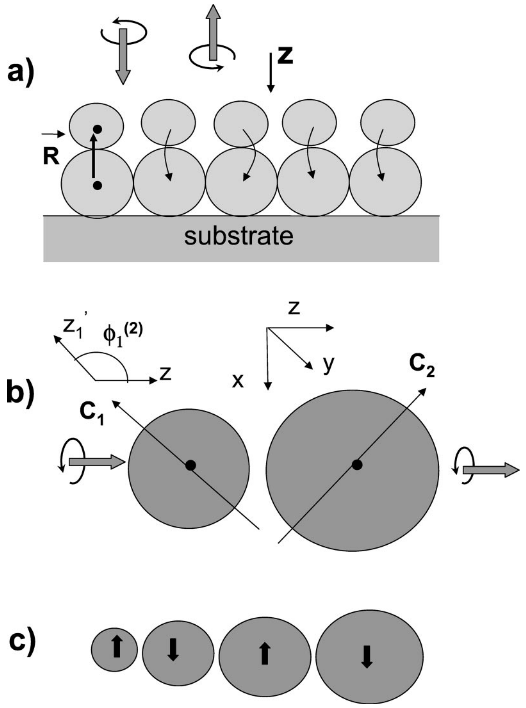

FIG. 3. (a) Schematic of a system with two monolayers of dots. Similar systems were studied experimentally in Refs. 9 and 10. (b), (c) Sketches of a pair of randomly oriented dots and a quantum-dot chain.

tal lattice. Such an anisotropy strongly affects the valence-band structure, giving rise to splitting between heavy- and light-hole levels. In nearly spherical crystals, anisotropy can be taken into account with perturbation theory.[16] The fourfold degeneracy of the hole states is split into 2 twofold degenerate states. The splitting can be written as $\Delta = \Delta_{cr} + \Delta_{shape}$, where Δ_{cr} is the crystal field splitting in a hexagonal lattice (like in CdSe) and Δ_{shape} is the splitting due to the shape. The Kramers doublet of hole states has quantum numbers $|M| = 1/2$ and $|M| = 3/2$. First, we consider two oblate dots forming a molecule with axial symmetry. In such a molecule, $\mathbf{c}_1 || \mathbf{c}_2 || \mathbf{z}$, where $\mathbf{c}_{1(2)}$ are the symmetry axes of dots [Fig. 3(b)]. To be more specific, we assume that the ground state of holes has the angular momenta $|M| = 3/2$, like in dots based on InP. Using the wave functions (8) it is easy to show that all results for disk-shaped QD's hold in the case of oblate dots with $\mathbf{c}_1 || \mathbf{c}_2 || \mathbf{z}$.

Randomly oriented QD pairs. It is natural to suppose that the randomness of nanocrystal axes in a QD solid will change the spin-transfer rates. To calculate the spin transport rates in a pair of arbitrary oriented dots, one can use the matrices of rotation for spin and spatial functions[17] and introduce Eulerian angles for the dots, $\phi_i^{(1)}, \phi_i^{(2)}, \phi_i^{(3)}$, where $i = 1,2$ [Fig. 3(b)]. By using the matrices of rotation, the coordinate system (x,y,z) is transformed into the systems (x_i', y_i', z_i') where individual dots have the symmetry of oblate ellipsoids. The spin-transfer probabilities for oblate dots depend only on the angles $\phi_i^{(1)}$ and $\phi_i^{(2)}$. Under resonance conditions the mean spins in the dots are connected by the equation

$$S_2 = S_1 \frac{W_a - W_b}{W_a + W_b}, \tag{9}$$

where $W_a = W_{\uparrow\to\uparrow} = W_{\downarrow\to\downarrow}$ and $W_b = W_{\uparrow\to\downarrow} = W_{\uparrow\to\downarrow}$. The coefficients $W_{a(b)}$ describe probabilities of interdot transitions with conservation (flipping) of spin and are complicated functions of $\phi_i^{(1)}$ and $\phi_i^{(2)}$. In a system with randomly oriented molecules, spin transfer does not vanish; it can seen by calculating the averaged probabilities $\overline{W} = \langle W \rangle_{\phi_1^{(1)}, \phi_1^{(2)}, \phi_2^{(1)}, \phi_2^{(2)}}$ and spins $\overline{S_i} = \langle S_i \rangle_{\phi_1^{(1)}, \phi_1^{(2)}, \phi_2^{(1)}, \phi_2^{(2)}}$. The ratio between averaged probabilities, $\overline{W}_a / \overline{W}_b$, depends on the type of interdot resonance: $\overline{W}_a / \overline{W}_b = 1.61$ for the *hh-hh* resonance and $\overline{W}_a / \overline{W}_b = 1/1.61$ for the resonance between *hh* and *lh* states. Thus, the *hh-hh* transfer conserves partially the spin orientation, whereas the *hh-lh* interdot coupling leads to a flipping of spin. For the case shown in Fig. 3(b), $\theta_1 = \theta_2 = 0$ and the calculated mean spin in dot 1 is given by $\overline{S}_1 = -0.586$. The dot-2 spin becomes $\overline{S}_2 = -0.22$ and 0.22 in the case of *hh-hh* and *hh-lh* resonances, respectively. Experimentally, the spin orientation in dot 2 can be observed by measuring the degree of circular polarization of secondary photons. We find that $\overline{P}_{circ}^{hh\text{-}hh} = 0.13$ and $\overline{P}_{circ}^{hh\text{-}lh} = -0.25$.

Quantum-dot chains. If cylindrical dots form an ideally oriented chain [like in self-assembled monolayers, Fig. 3(a)] and all dots are under resonance conditions, the spin can be transferred along the chain without losses, $S_N = \pm S_1$, where S_1 and S_N are the mean spins in the first and Nth dots, respectively [Fig. 3(c)]. The sign ± in the above relation depends on the types of interdot resonances. If an ideal chain is formed of spherical dots, the transferred spin rapidly decreases with the number of dots, $S_N = S_1/2^N$. In disordered chains, there is an additional mechanism of spin randomization. For oblate crystals with randomly oriented axes and under interdot resonance conditions, we can estimate the decay of spin using the averaged probabilities $\overline{W}_{a,b}$. This leads to $S_N \sim 0.2^N S_1$.

The dipole-dipole interaction (2) provides the main contribution to the transfer rate. At the same time, the higher multipole terms of the Coulomb operator can certainly affect the magnitude of transfer rate and lead to additional interdot resonances which should be consistent with symmetry. However, the spin-transfer selection rules established above will hold beyond the dipole-dipole approximation because these rules come from axial symmetry in a QD pair. Namely, the *hh-hh* and *hh-lh* interdot resonances will result in the conservation and flipping of spin, respectively.

Experimentally, the most preferable systems to observe spin transport are the system with QD monolayers[9,10] or a single QD pair[10] on a surface. In the first case, all QD pairs have the same orientation of the molecular axis **R** [Fig. 3(a)]. If QD's are spherical, the spin polarization in dots 2 will be $S_1/2$. In the case of oblate QD's with randomly oriented QD axes, the spin orientation will remain nonzero under interdot resonance conditions. Another suitable system is a single-QD molecule bound to a surface which can be studied by available methods of single-dot spectroscopy.[14]

Another important issue related to exciton transport is the strength of dipole transitions. In this paper, we assumed that the ground-state excitons in dots are optically active. This would not be the case for CdSe dots where a strong interband exchange interaction splits exciton levels. The resulting exciton ground state becomes dark. Our results are fully applicable to QD's with optically active excitons. For example, excitons in the ground state are optically active in InP nanocrystals where the exchange interaction is weak.[20] Besides, the exciton ground states are optically active in self-assembled QD's which are usually lens shaped.[13] The case with optically inactive ground states of excitons should be considered specially. In addition, we considered excitons within the single-particle approximation ignoring the intradot Coulomb interaction. This approximation is justified for our dot parameters since the typical energy of in-plane quantization is greater than the intradot Coulomb interaction.[21]

To observe spin transfer between QD's, one should have a sufficiently long spin-relaxation time. A moderate magnetic field can favor spin transport because it induces spin splitting and strongly enlarges the degree of circular polarization of emitted photons.[18,19] In the system with monolayers, the magnetic field can be applied parallel to the molecular axis **R**. The spin-relaxation times found in experiments on the bulk semiconductors and QD's range from 100 ps to 100 μs.[7,22,23] The exciton-transfer times in nanocrystals, recently measured in Refs. 8 and 9, are in the range from 700 ps to 10 ns. This tells us that suitable conditions to observe spin transport of electrons can be found experimentally. By analyzing the rate equations, one can see that the mean spin in dot 2 depends mostly on the ratio $\tau_{trans}/\tau_{e\text{-}spin}$, where $\tau_{e\text{-}spin}$ is the spin relaxation time for the electron and τ_{trans} is the interdot transfer time of excitons. At the same time, the emission intensity of dot 2 is determined by the ratio τ_{trans}/τ_{exc}. Spin and energy transfers become efficient if $\tau_{trans} \leqslant \tau_{e\text{-}spin}$ and $\tau_{trans} \leqslant \tau_{exc}$, respectively. The latter was satisfied in recent experiments.[8–10]

The rate of energy transfer between QD's can strongly depend on temperature and resonance conditions. We now assume that the QD pair is designed to satisfy the resonant condition $E_1^0 = E_2^{exc}$, where E_1^0 is the ground-state energy of excitons in dot 1 and E_2^{exc} is related to the excited exciton state in dot 2. In the case of *hh*-*hh* interdot resonance, the transfer time can be estimated as $\tau_{trans} = 1/(w_0 J)$. Here we will use the parameters of InP: $d_0 = 6$ Å and $\epsilon = 12.6$. At low temperatures, homogeneous broadenings of excitons are relatively small and $\Gamma_1^0 \ll \Gamma_2^{exc}$, where Γ_1^0 and Γ_2^{exc} are the broadenings of exciton levels in dots 1 and 2, respectively. We obtain $\tau_{trans} \sim 120$ ps, taking the parameters $\Gamma_1^0 = 1$ meV, $\Gamma_2^{exc} = 5$ meV, and $R = 70$ Å. At room temperature, we find $\tau_{trans} \sim 1$ ns with $\Gamma_1^0 \sim \Gamma_2^{exc} \sim 20$ meV.

To calculate the spin orientation in nanocrystals, we assumed that the time of angular momentum relaxation for the holes is much shorter that the spin-relaxation time for the electrons. This relation is typical for experiments. The momentum relaxation time of holes in solids and nanostructures is often short because of strong *hh*-*lh* mixing in the valence band and due to relatively weak quantization of energy levels of holes.[15]

To conclude, we have studied spin transfer in nanocrystals which does not involve the transport of charge. It has been demonstrated that the spins can be efficiently transferred between quantum dots via the Coulomb interaction. In the transfer process the electron spin can be conserved or flipped. The transferred spin polarization survives even in randomly oriented QD pairs and chains.

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