Spin and energy transfer in nanocrystals without transport of charge

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ABSTRACT

We describe a mechanism of spin transfer between individual quantum dots that does not involve tunneling. Incident circularly-polarized photons create inter-band excitons with non-zero 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 either conservation or flipping of the electron spin. The second dot can radiate circularly-polarized photons at lower 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.
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 computer [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 (QDs), where individual nanocrystals strongly confine carriers and do not permit efficient tunnel coupling [8–10]. However, instead of direct tunnel coupling, the colloidal QDs 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 a 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 (fig. 1a-c). An incident photon creates an exciton in the small dot 1 with a larger optical gap (fig. 1a-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 the dot 2, the exciton becomes trapped and contribute to the photoluminescence (PL) at the dot-2 energy. If electrons in the 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 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 QDs via the Coulomb interaction. This becomes possible thanks to the strong spin-orbit interaction in the valence bands of QDs. 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 QDs can be grown by using self-organization technology [13]. In such stacked QDs, sizes of dots and inter-dot separation are well controlled. Another method to fabricate a system with QD pairs is colloidal synthesis [8–10]. In a solid of colloidal QDs with two distinct sizes, QD pairs are randomly oriented [8,9]. In monolayers of QDs (fig. 3a), 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 $\tau_{e^{-spin}}$, $\tau_{exc} \gg \tau_{energy}$, where $\tau_{exc}$ is the exciton lifetime in a single QD related to radiational and non-radiational transitions, $\tau_{energy}$ is the energy relaxation time of excitons within a dot, and $\tau_{e^{-spin}}$ is the electron spin lifetime, and $\tau_{h^{-spin}}$ is the momentum relaxation time of holes. In other words, we suppose: (1) fast intra-dot relaxation of 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:

$$
\Psi_{e, \uparrow (\downarrow), n,l} = u_{\uparrow (\downarrow)} \Phi_{n,l}^{(i)}(r||, z), \quad \Psi_{h, \pm 3/2, n,l} = u_{\pm 3/2} \Phi_{n,l}^{(i)}(r||, z), \quad \Psi_{l, \pm 1/2, n,l} = u_{\pm 1/2} \Phi_{n,l}^{(i)}(r||, z), \quad (1)
$$

where $i = 1, 2$ and $r|| = (x, y)$; $u_{\uparrow (\downarrow)}$, $u_{\pm 3/2}$, and $u_{\pm 1/2}$ are the Bloch functions of electrons, heavy holes ($hh$) and light holes ($lh$), respectively; $\Phi_{n,l}^{(i)}(r||, z) = f_0^{(i)}(z) f_{n,l}^{(i)}(r||)$ are the envelope functions, where the $f_0^{(i)}(z)$ is the ground-state function for the motion in the $z$-
direction, $R_n^i(r_{||})$ are Bessel’s functions describing the in-plane motion, and $(n, l)$ are the radial and azimuthal quantum numbers of in-plane motion, respectively; $n = 1, 2, \ldots$ and $l = 0, \pm 1, \pm 2, \ldots$. 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 the dot 1 can be written as follows: $\hat{V}^{opt}_{1,+} = e_1 \hat{p} = \cos(\theta_1) \hat{p}_x + i \hat{p}_y + \sin(\theta_1) \hat{p}_z$, where $\hat{p}$ and $e_1$ are the momentum operator and polarization vector, respectively. Using this operator, the probability of inter-band optical transitions for the dot 1 takes a form $P_{s,\mu}^{1} = |\langle \psi_{s,n,l}^{1} | \hat{V}^{opt}_{1,+} | \psi_{\mu,n,l}^{1} \rangle|^2$, where $s = \uparrow, \downarrow$ and $\mu = \pm 3/2, \pm 1/2$. Emission of the dot 2 is described in a similar way with the operator $\hat{V}^{opt}_{2,\pm} = \cos(\theta_2) \hat{p}_x \pm i \hat{p}_y - \sin(\theta_2) \hat{p}_z$, where signs $\pm$ 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 inter-dot 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,

$$\hat{V}_{Coul} = \frac{e^2}{\epsilon R^3} (r_1 r_2 - 3 z_1 z_2),$$

where $r_{1(2)}$ are the radius vectors related to the dots (fig. 1a), $\epsilon$ 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 inter-dot transition takes the form

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

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 intra-dot energy relaxation, the function $|\beta_1 >$ in eq. 3 describes the ground-state exciton in the dot 1 with $s_1 = \uparrow, \downarrow$ and $\mu_1 = \pm 3/2$, and $(n_1, l_1) = (1, 0)$. In the spirit of the Förster theory the delta 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 $\Gamma_{\beta_i}$ is the homogeneous broadening of the exciton $\beta_i$. 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 photon in the dot 1 that involves a heavy-hole level (fig. 1b); in other words, the incident-photon energy is taken below the first inter-band transition related to the light hole. The mean $z$-component of electron spin polarization in the dot 1 is determined by the probabilities $P_{1\mu}$ and is equal to

$$S_1 = \frac{P_{1\uparrow} - P_{1\downarrow}}{P_{1\uparrow} + P_{1\downarrow}} = -\frac{2 \cos(\theta_1)}{[\cos(\theta_1)^2 + 1]},$$

where $P_{1\uparrow,\downarrow} \propto |P_{cv}|^2(\cos(\theta_1) \mp 1)^2$ is the probability of the electron being in the state $\uparrow(\downarrow)$ and $P_{cv} = \langle S|\hat{p}_x|X \rangle$ is the inter-band optical matrix element. In the optical matrix elements, the operator $\hat{p}$ was involved only in the integrals with the Bloch functions. For the next step, we calculate the Coulomb matrix elements under resonance conditions. In the regime of inter-dot resonance, the ground-state exciton energy of the dot 1 is equal to the energy of excited dipole-active exciton in the dot 2. The latter state can be composed of either heavy-hole or light-hole. We start with the resonance between heavy-hole states in the dots (fig. 1b). The probability to create the exciton with $s_2 = \uparrow$ in the dot 2 is given by

$$P_{2\uparrow} = \frac{1}{2}P_{1\downarrow}W_{\beta_1 \rightarrow \beta_2} = \frac{1}{2}w_0J_{\beta_1,\beta_2}/2,$$

where $\beta_1 = (\uparrow, 3/2, 1, 0)$ and $\beta_2 = (\uparrow, 3/2, n_2, l_2)$; the factor $1/2$ is the probability to find the heavy hole in either state ($\pm 3/2$) in the dot 1; this is due to fast momentum relaxation of holes. Besides, $W_{\beta_1 \rightarrow \beta_2}$ is the probability of Förster-like transfer between the states $\beta_1$ and $\beta_2$. A coefficient $w_0 = 2\pi d_0^4(e^4/e^2\hbar^2)$, where $d_0 = \langle X|x|S \rangle$ is the atomic dipole moment. For the spin $\downarrow$ we have a similar equation,

$$P_{2\downarrow} = \frac{1}{2}w_0J_{\beta_1,\beta_2}/6,$$

where $\beta_2 = (\uparrow, 1/2, n_2, l_2)$. Similarly, $P_{2\downarrow} = \frac{1}{2}w_0J_{\beta_1,\beta_2}/6$. Thus, we obtain the effect of spin flipping:
\[ S_2'' = -S_1. \] 

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

\[ S_2 = \frac{\sum_{\beta_1,\beta_2} P_2^\dagger(\beta_1 \rightarrow \beta_2) - P_2^\dagger(\beta_1 \rightarrow \beta_2)}{\sum_{\beta_1,\beta_2} P_2^\dagger(\beta_1 \rightarrow \beta_2) + P_2^\dagger(\beta_1 \rightarrow \beta_2)}, \]

where the summation involves all pairs of states; the index \( \beta_1 \) is related to the \( hh \) ground state of the 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

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

where \( I_\pm \) are the light intensities given by \( I_+ = P_2^\dagger P_+(\uparrow) + P_2^\dagger P_+(\downarrow) \) and \( I_- = P_2^\dagger P_-(\uparrow) + P_2^\dagger P_-(\downarrow) \). Here, the optical transition rate \( P_\sigma(s) \) describes the emission process in which an electron with the spin \( s \) in the dot 2 creates a photon with the circular polarization \( \sigma \), where \( \sigma \) can be + or −. The degree of circular polarization (7) strongly depends on the resonance conditions between the QDs (fig. 2a). 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. 2a). The latter comes from the conservation of the total angular momentum in the Coulomb matrix elements. Besides, the rate of exciton transfer, \( 1/\tau_{\text{trans}} = W_{\beta_1} \), is strongly enhanced under the inter-dot resonance conditions (fig. 2b). Note that the total angular momentum is not conserved in the three-step process shown in figs. 1b, c because of fast relaxation of angular momentum for the hole.

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

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

Here \( i \) is the QD number \( (i = 1, 2) \), \( M \) is the \( z \)-component of 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 the 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 a form $P_{\text{circ}} = -S_2 \cos(\theta_2)/2 = \cos(\theta_1) \cos(\theta_2)/8$. At the angles $\theta_{1(2)} = 0$, the polarization of emitted light is maximal and equal to $P_{\text{circ}} = 1/8$. 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 inter-dot 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 crystal lattice. Such 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 4-fold degeneracy of the hole states is split into two 2-fold degenerate states. The splitting can be written as $\Delta = \Delta_{\text{cr}} + \Delta_{\text{shape}}$, where $\Delta_{\text{cr}}$ is the crystal field splitting in a hexagonal lattice (like in CdSe) and $\Delta_{\text{shape}}$ is the splitting due to the shape. The Kramers doublet of hole states has the 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, $c_1||c_2||z$, where $c_{1(2)}$ are the symmetry axes of dots (fig. 3 b). To be more specific, we assume that the ground state of holes have the angular momenta $|M| = 3/2$, like in the dots based on InP. Using the wave functions (8) it is easy to show that all results for disk-shaped QDs hold in the case of oblate dots with $c_1||c_2||z$.

**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. 3b). 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 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 equation

$$S_2 = S_1 \frac{W_a - W_b}{W_a + W_b}, \quad (9)$$
where $W_a = W_{\uparrow\rightarrow\uparrow} = W_{\downarrow\rightarrow\downarrow}$ and $W_b = W_{\uparrow\rightarrow\downarrow} = W_{\downarrow\rightarrow\uparrow}$. The coefficients $W_{a(b)}$ describe probabilities of inter-dot 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 be seen by calculating averaged probabilities $\bar{W} = \langle W \rangle_{\phi_1^{(1)},\phi_1^{(2)},\phi_2^{(1)},\phi_2^{(2)}}$ and spins $\bar{S}_i = \langle S_i \rangle_{\phi_1^{(1)},\phi_1^{(2)},\phi_2^{(1)},\phi_2^{(2)}}$. The ratio between averaged probabilities $\bar{W}_a/\bar{W}_b$ depend on the type of inter-dot resonance: $\bar{W}_a/\bar{W}_b = 1.61$ for the $hh - hh$ resonance and $\bar{W}_a/\bar{W}_b = 1/1.61$ for the resonance between $hh$ and $lh$ states. Thus, the $hh - hh$ transfer conserves partially the spin orientation, whereas the $hh - lh$ inter-dot coupling leads to the flipping of spin. For the case shown in fig. 3b, $\theta_1 = \theta_2 = 0$ and the calculated mean spin in the dot 1 is given by $\bar{S}_1 = -0.586$. The dot-2 spin becomes $\bar{S}_2 = -0.22$ and 0.22 in the case of $hh - hh$ and $hh - lh$ resonances, respectively. Experimentally, the spin orientation in the dot 2 can be observed by measuring the degree of circular polarization of secondary photons. We find that $P_{\text{circ}}^{hh-hh} = 0.13$ and $P_{\text{circ}}^{hh-lh} = -0.25$.

Quantum-dot chains. If cylindrical dots form an ideally-oriented chain (like in self-assembled monolayers, fig. 3a) and all of the 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 $N^{th}$ dots, respectively (fig. 3c). The sign $\pm$ in the above relation depends on the types of inter-dot 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 inter-dot resonance conditions, we can estimate the decay of spin using averaged probabilities, $\bar{W}_{a,b}$. 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 inter-dot 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. Specifically, the $hh - hh$ and $hh - lh$ inter-dot resonances will result in 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. 3a). If QDs are spherical, the spin polarization in the dots 2 will be \( S_1/2 \). In the case of oblate QDs with randomly-oriented QD axes, the spin orientation will remain non-zero under inter-dot 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 inter-band exchange interaction splits exciton levels. The resulting exciton ground state turns to be dark. Our results are fully applicable to the QDs with optically-active excitons. For example, excitons in the ground state are optically-active in InP nanocrystals where the exchange interaction is weak [20]. In addition, the exciton ground states are optically active in self-assembled QDs [13]. Such self-assembled QDs are usually lens-shaped. 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 intra-dot Coulomb interaction. This approximation is justified for our dot parameters since the typical energy of in-plane quantization is greater than the intra-dot Coulomb interaction [21].

A moderate magnetic field can favor the observation of spin transport because it induces the spin-splitting and strongly enlarges the degree of circular polarization of emitted light in QDs [18,19]. In the system with monolayers, the magnetic field can be applied parallel to the molecular axis \( R \). To observe spin transfer between QDs, one should have a sufficiently long spin-relaxation time. The spin-relaxation times found in experiments on the bulk semiconductors and QDs range from 100 ps to 100 \( \mu \)s [7,22,23]. The exciton-transfer times in nanocrystals, recently measured in refs. [8,9], 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 the dot 2 depends mostly on the ratio \( \tau_{\text{trans}}/\tau_{\text{e-spin}} \), where \( \tau_{\text{e-spin}} \) is the spin relaxation time for the electron and \( \tau_{\text{trans}} \) is the inter-dot transfer time of excitons. At the same time, the emission intensity of the dot-2 is determined by the ratio \( \tau_{\text{trans}}/\tau_{\text{exc}} \). Spin and energy
transfers become efficient if $\tau_{\text{trans}} \leq \tau_{\text{e-spin}}$ and $\tau_{\text{trans}} \leq \tau_{\text{exc}}$, respectively. The latter was satisfied in recent experiments [8–10].

The rate of energy transfer between QDs can strongly depend on temperature and resonance conditions. We now assume that the QD pair is designed to satisfy the resonant condition, $E_{01}^0 = E_{2}^{\text{exc}}$, where $E_{01}^0$ is the ground-state energy of exciton in the dot 1 and $E_{2}^{\text{exc}}$ is related to the excited exciton state in the dot 2. In the case of $hh - hh$ inter-dot resonance, the transfer time can be estimated as $\tau_{\text{trans}} = 1/(\omega_0 J)$. Here we will use the parameters of InP: $d_0 = 6 \, \text{Å}$ and $\epsilon = 12.6$. At low temperatures, homogeneous broadenings of excitons are relatively small and $\Gamma_{01}^0 \ll \Gamma_{2}^{\text{exc}}$, where $\Gamma_{01}^0$ and $\Gamma_{2}^{\text{exc}}$ are the broadenings of exciton levels in the dots 1 and 2, respectively. We obtain $\tau_{\text{trans}} \sim 120 \, \text{ps}$ taking parameters $\Gamma_{01}^0 = 1 \, \text{meV}$, $\Gamma_{2}^{\text{exc}} = 5 \, \text{meV}$, and $R = 70 \, \text{Å}$. At room temperature, we find $\tau_{\text{trans}} \sim 1 \, \text{ns}$ with $\Gamma_{01}^0 \sim \Gamma_{2}^{\text{exc}} \sim 20 \, \text{meV}$.

To calculate the spin orientation in nanocrystals, we assumed that the time of 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 due to strong $hh - lh$ mixing in the valence band and relatively weak quantization of energy levels of holes [15].

To conclude, we have studied spin transfer in nanocrystals which does not involve 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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FIG. 1.
Sketch of the quantum-dot molecule (a). Energy diagrams of intra- and inter-dot transitions in the transfer process (b and c); the label $\gamma - e; n, l$ denotes the exciton composed of hole $\gamma$ and electron, where $\gamma$ can be $hh$ or $lh$, and $(n, l)$ are the envelope-function indices.
FIG. 2.

(a) Calculated degree of circular polarization of photons emitted by the dot 2 as a function of the dot-1 diameter; $\theta_{1(2)} = 0$. The sizes of the dot 2 are kept constant, whereas the diameter of the dot 1 is varied. (b) Calculated rate of exciton transfer from the dot 1 into dot 2. Inserts show diagrams of inter-band transitions. The crystal parameters correspond to InP quantum dots; effective masses: $m_e = 0.077m_0$, $m_{lh} = 0.12m_0$, $m_{hh} = 0.6m_0$; $R = 80$ 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 exciton in the dot 1 is taken as 1 $meV$; the broadening of all excited states in the dot 2 is assumed to be 5 $meV$. 
FIG. 3.

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