Optical RKKY Interaction between Charged Semiconductor Quantum Dots

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We show how a spin interaction between electrons localized in neighboring quantum dots can be induced and controlled optically. The coupling is generated via virtual excitation of delocalized excitons and provides an efficient coherent control of the spins. This quantum manipulation can be realized in the adiabatic limit and is robust against decoherence by spontaneous emission. Applications to the realization of quantum gates, scalable quantum computers, and to the control of magnetization in an array of charged dots are proposed.

Quantum control of an electron spin, either independent of other spins or condition on their states, in a semiconductor nanostructure is a central issue in the emerging fields of spintronics and quantum information processing. The spin of a single electron confined in a semiconductor quantum dot (QD) was proposed [1] as a qubit for the realization of scalable quantum computers. Quantum gates are designed using electric gates to control via overlap the exchange interaction between two electrons in neighboring dots. Optical control was also proposed, in which a cavity mode couples different dots [2], or a dipole-dipole interaction between charged excitons strongly polarized by an external dc field is exploited [3]. Optical control possesses several advantages compared with control by gate voltage. Ultrafast lasers can control quantum systems on the femtosecond time scale, and using shaping techniques the amplitude and phase of the pulses can be designed at will, offering a great deal of flexibility and efficiency [4].

In this Letter, we report a theory of an exchange interaction between two electron spins in separate dots in a typical semiconductor QD system by virtual excitation of delocalized exciton states in the host material which interact with the electrons in both dots. This time-dependent effective interaction is driven by the external laser field and is, thus, controllable. The virtual excitation by an off-resonant laser preserves the coherence of the spin dynamics. This indirect exchange mechanism is analogous to a RKKY interaction [5] between two magnetic impurities mediated by conduction electron or excitons [6], except that the intermediate electron-hole pair is produced by the external light. The optical quantum control of a single exciton in a semiconductor QD has been recently reported in GaAs QDs generated by monolayer fluctuations [7] and InGaAs self-assembled QDs [8,9]. The short radiative recombination lifetime of the exciton (of the order of 100 ps) gives a severe limitation for the application to quantum computation, even with the

help of shaping techniques [4]. This can be avoided by doping QDs each with a single conduction electron and by encoding the quantum information in the spin degrees of freedom. Optical control by virtual excitation avoids the fast optical decoherence. Thus, the advantages of a very long spin coherence time in QDs [10] and fast optical control can be combined.

Consider two electrons localized in two QDs at $\mathbf{R}_\ell$ ($\ell = 1, 2$) with wave functions $\phi_\ell(\mathbf{r} - \mathbf{R}_\ell)$ and a laser field used to generate exciton states in a continuum. This continuum is provided by states in the host material embedding the QDs with an energy gap ϵ_G. QDs can be embedded in bulk, quantum well, or quantum wire host structures. Fluctuation QDs embedded in a narrow quantum well [7] represent an example of a system with a two dimensional continuum. A promising system for the scheme which we propose is provided by pyramidal QDs [11], where localized states in growth-controlled QDs and delocalized states in the so-called vertical quantum wire are well separated and can be addressed selectively. In this case, the continuum states are in the vertical quantum wire. The Coulomb interaction between the photoexcited pairs and the localized states contains direct and exchange contributions. The direct term gives state renormalization. The attraction of the exciton to the dot is determined in the long range by the dipole moment of the exciton induced by the localized electron in the dot and in the short range by the dot potential. The binding of the exciton to the dot is then sensitive to the design of the dot, ranging from a very weakly bound state to one localized in the dot [12]. The former has a wave function overlap to the neighboring dot and contributes to the optical RKKY. The latter can be made far off resonance to the optical excitation. Here we focus on the spin structure of the Hamiltonian arising out of the exchange interaction between the localized and optically excited conduction electrons. The exchange interaction between the localized electron and the valence hole is negligible. For

convenience, $\hbar = 1$ throughout. Hence, the Hamiltonian of the system contains, besides the electron and hole energies in the host and in the dots, (1) the exchange interaction between electrons,

$$H_X = -\frac{1}{V} \sum_{\substack{\ell,\alpha,\alpha' \\ \mathbf{k},\mathbf{k}'}} J_\ell(\mathbf{k},\mathbf{k}')\mathbf{S}^\ell \cdot \mathbf{s}_{\alpha,\alpha'} c^\dagger_{\mathbf{k},\alpha} c_{\mathbf{k}',\alpha'}, \quad (1)$$

where $\mathbf{S}^j$ denotes the spin of the jth localized electron, $\mathbf{s}$ the spin of the electron in the photogenerated pair, and $c^\dagger_{\mathbf{k},\alpha}$ are the creation operators of free electron states; and (2) the time-dependent control Hamiltonian describing the creation of electron-hole pairs

$$H_C(t) = \sum_{\mathbf{k},\sigma} \frac{\Omega_{\mathbf{k},\sigma}(t)}{2} e^{-i\omega_{P\sigma}t} c^\dagger_{\mathbf{k},-\sigma} h^\dagger_{-\mathbf{k},\sigma} + \text{H.c.}, \quad (2)$$

where $h^\dagger_{\mathbf{k},\sigma}$ are hole creation operators. $\Omega_{\mathbf{k},\sigma}(t)$ is the time-dependent Rabi energy associated with the electric field of the optical pulse times the transition dipole matrix element of the electron-hole pair with momenta $\pm\mathbf{k}$, resulting from taking the wave vector of the photon to be zero. $\sigma = \pm$ denotes the $\sigma\pm$ circular polarization of light, which fixes the spin configuration of the photo-excited electron spin state $-\sigma(1/2)$ and hole state $\sigma(3/2)$. We consider only a single heavy hole band which is valid for GaAs confined heterostructure. For the exchange integral $J_\ell(\mathbf{k},\mathbf{k}')$, the Coulomb interaction is screened by the static dielectric constant [13]. We approximate the continuum electron by a plane wave orthogonalized (OPW) to the dot states and further simplify the exchange to the form

$$J_\ell(\mathbf{k},\mathbf{k}') \equiv j_\ell e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_\ell}, \quad (3)$$

with a constant prefactor given by

$$j_\ell^d \sim I\text{Ry}^* a_B^* \xi^{d-1}, \quad (4)$$

where ξ is the localization length within a dot, Ry^* and a_B^* denote the effective exciton Rydberg energy and Bohr radius in the host semiconductor, I is a dimensionless constant that depends only on the particular geometry of the dot, and d is the dimensionality of the host. The dependence of I on the wave vectors is removed by using a suitable average discussed below.

In the absence of the laser pulse, the system with two localized electrons is in a degenerate ground state with four spin states. In the presence of a laser field nearly resonant with the continuum, the ground state energy is shifted by $\Delta E_0 = -\sum_{\mathbf{k}}[\Omega_\sigma(t)/2](\delta + k^2/2\mu)^{-1}[\Omega_\sigma^*(t)/2]$ where $\delta = \epsilon_G - \omega_P$ is the detuning of the laser with respect to the electron-hole continuum and μ is the reduced mass of the electron-hole pair. ΔE_0 contributes to the well-known blueshift in the excitonic transition or dynamic Stark effect. This contribution, which is diagonal in the spin index of the localized states, is irrelevant for our spin control purposes. However, when

Coulomb interaction is taken into account, the spin configuration of the localized electrons does affect this shift. To calculate this effect, we break the pair propagator $(\delta + k^2/2\mu)^{-1}$ into its electron and hole parts and then consider the self-energy correction in the electron propagator due to the interaction with the localized states. These corrections give an effective Hamiltonian for the spins of the localized electrons. Figure 1 shows the lowest order contribution in Coulomb interaction to the effective exchange interaction between the two localized electrons mediated by the free electron in the photoexcited pair. The incoming photon (wavy line) with energy ω_P and polarization $\sigma+$ creates a pair of electron (solid line) and hole (dashed line). The electron in the pair interacts with the two electrons localized in the two neighboring dots (dotted lines) via H_X in Eq. (1) and then recombines with the hole. The indices β and γ refer to the spin states of the localized electrons. The loop contains the integral in the exchanged energy ω, and the sum in the momentum $\mathbf{k}'_e$ and spin α of the intermediate electron. If the free carriers' motion is spin independent, the interaction between two local spins associated with this diagram contains the term $(\mathbf{S}^1 \cdot \mathbf{s})(\mathbf{S}^2 \cdot \mathbf{s}) = (\mathbf{S}^1 \cdot \mathbf{S}^2)/4 + i\mathbf{s}\cdot(\mathbf{S}^1 \times \mathbf{S}^2)/2$. When it is summed with the diagram in which the order of the two local spins is reversed, the cross product terms cancel. Hence, the effective spin-spin Hamiltonian assumes the Heisenberg form

$$H_s = -2J_{12}\mathbf{S}^1 \cdot \mathbf{S}^2, \quad (5)$$

where the effective exchange constant J_{12} is always positive, given by
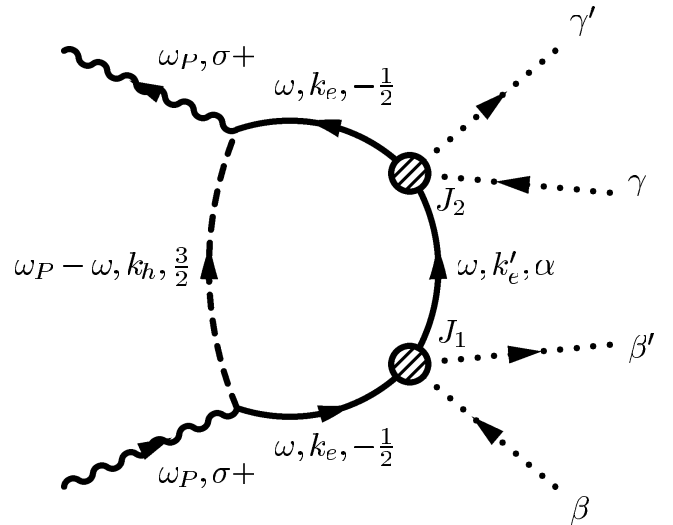

FIG. 1.   Effective spin-spin interaction for the localized electrons in the dots 1 and 2 (indicated by dotted lines) induced by a photoexcited electron-hole pair (the solid and dashed lines, respectively). The indices β and γ denote the spin states of the electrons localized in the dots. The photon propagator is depicted by a wavy line.

$$J_{12}(R) = \frac{1}{16} \sum_\sigma |\Omega_\sigma(t)|^2 j_1^{\mathrm{d}} j_2^{\mathrm{d}} \int \frac{d^d\mathbf{k}}{(2\pi)^d} \frac{d^d\mathbf{k}'}{(2\pi)^d} e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \left(\delta + \frac{k^2}{2m_{\mathrm{h}}} + \frac{k^2}{2m_{\mathrm{e}}}\right)^{-2} \left(\delta + \frac{k^2}{2m_{\mathrm{h}}} + \frac{k'^2}{2m_{\mathrm{e}}}\right)^{-1}, \tag{6}$$

where m_{e} (m_{h}) denotes the electron (hole) mass, and $\mathbf{R} = \mathbf{R}_1 - \mathbf{R}_2$. The Rabi energy can reasonably be assumed independent of $\mathbf{k}$. From Eq. (6) it can be seen that $J_{21} = J_{12}$. Parabolic dispersions for the electron and the hole energies are assumed. Unpolarized light is used to induce the ferromagnetic interaction between the local spins, so as to avoid the spin polarization effect of the first order process in H_X.

The most important correction to Fig. 1 is due to the Coulomb attraction between the electron and hole in the excited pair [6], giving rise to three exciton propagators. The electron-hole pair energies in Eq. (6) are replaced by the exciton energies. The exciton wave functions enhance the oscillator strength and, hence, the Rabi energies at the two optical vertices and the exchange constants at the spin vertices. To remain in the regime of virtual excitations, the laser energy must be adjusted to below the lowest discrete exciton state. Keeping only the $1s$ exciton contribution, we obtain

$$J_{12} = \left(\frac{\Omega}{4\delta}\right)^2 \frac{j_1^{\mathrm{d}} j_2^{\mathrm{d}} |\phi_{1s}^{\mathrm{d}}(0)|^2}{\delta} I_{\mathrm{d}}(R), \tag{7}$$

where

$$I_{\mathrm{d}}(R) = \int \frac{d^d\mathbf{q}}{(2\pi)^d} \frac{e^{i\mathbf{q}\cdot R} [\rho_{1s}^{\mathrm{d}}(\alpha q)]^2}{1 + (\kappa_{\mathrm{M}} q)^2}. \tag{8}$$

The term $\phi_{1s}^{\mathrm{d}}(0)$ is the $1s$ excitonic wave function in d dimensions, $\kappa_{\mathrm{M}} = 1/\sqrt{2M\delta}$ is a characteristic optical length related to the detuning, M is the total mass of the pair, and $\alpha = m_{\mathrm{h}}/M$. The integral in $\mathbf{q}$ represents the sum over the exciton center of mass wave vectors scattered by the localized electrons. The form factor is defined as $\rho_{1s}^{\mathrm{d}}(q) = \int d^d\mathbf{r} |\phi_{1s}^{\mathrm{d}}(\mathbf{r})|^2 e^{i\mathbf{q}\cdot\mathbf{r}}$. The dependence of the interaction on the interdot separation is contained in the integrals $I^{\mathrm{d}}(R)$. Their explicit analytical expression and the contributions from higher excitonic levels will be given in a long publication [12]. The spatial dependence in all three cases, $d = 1, 2, 3$, is dominated by two exponential terms with two characteristic lengths: the optical length κ_{M} and the Bohr radius.

In Fig. 2 we plot J_{12} as a function of the separation between the dots in the two dimensional case, for three different values of the detuning. The results when excitonic effects are not included are also shown for comparison. The excitonic enhancement can be more than 2 orders of magnitude. An analogous excitonic enhancement effects was found in the case of spin-flip Raman scattering of electrons trapped in neutral donors [14]. In the calculation we used $m_e = 0.07m$ and $m_h = 0.5m$, a Rabi energy of 0.1 meV at the peak of the pulse, 5 meV for the exciton Rydberg, and the dot dimension $\xi \sim 2a_B^*$. A

typical value for the Bohr radius is 150 Å. A simple estimate of $I = 5.76$, the magnitude of the exchange in Eq. (4), is obtained for one OPW from the average of exchange j_ℓ squared over k and k' for the wave vector cutoff equal to $1.1/\xi$. This value corresponds to the minimum value of the averages for all possible cutoffs. An orbital of Slater form is assumed for the localized electron. The exchange energy between two local spins is then of the order of 1 meV, which is comparable to the estimated values of the exchange coupling due to tunneling in coupled QDs [15]. The interaction can be considerably enhanced in systems where large QDs are vertically stacked with separations smaller that their lateral size.

The detuning δ has to be larger than the exciton linewidth so that there is no absorption of energy in the exciton spectrum from the pump. The dynamics of the localized spin is thus fully coherent, and the finite lifetime Γ of the photoexcited states is no longer a limitation for quantum information processing. The optical pulses has to be switched on and off in such a way that no real population is excited in the intermediate excitonic states. In the simple case of a two-level system G and E, this adiabaticity condition can be written in the form $|(\frac{d}{dt}\langle G'|)|E'\rangle| \ll |\epsilon_{G'} - \epsilon_{E'}|$, where $|G'\rangle$ and $|E'\rangle$ are the time-dependent dressed states in the rotating frame (or
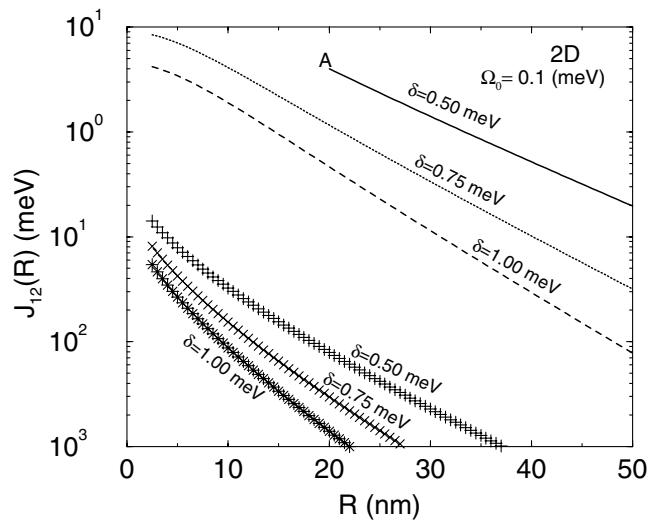

FIG. 2. Optically induced interaction in the two dimensional host as a function of the distance between the dots, with the excitonic corrections (upper three lines) and without (lower three curves), for three detuning values. The upper limit, if reached, of the adiabaticity condition in Eq. (9) for a swap gate is indicated by label A. Ω_0 represents the peak value of the Rabi energy in the pulse.

time-dependent Floquet states) of the ground and excited state [16]. This leads to $|\delta\dot{\Omega}(t)| \ll (\delta^2 + \Omega(t)^2)^{3/2}$. Assuming Gaussian optical pulses of the form $\Omega(t) = \Omega_0 e^{-(t/\sigma)^2}$, we find the condition for the length of the pulse to preserve the adiabaticity to be

$$\sigma \gg \frac{\Omega_0}{\delta^2}. \qquad (9)$$

If we fix the angle of rotation of the spin by, e.g., $\int dt J(t) = \pi/2$ (swap gate [1]), the condition above can be translated into an upper limit for J_{12}. This limit is indicated explicitly in Fig. 2 by the label "A" in the top curve. The other curves shown are all within the adiabatic limit. Our case differs from the ideal two-level case discussed above in two aspects: the presence of a continuum of excitonic states in addition to the discrete states and the interaction of the continuum with the localized states. Neither changes considerably the estimates which go into the adiabaticity condition, and Eq. (9) remains valid. The exchange Coulomb interaction between the photoexcited electron and the localized electrons in Eq. (1) is time independent and does not affect the adiabaticity. For the continuum, we have used a Fano approach and defined $|G'\rangle$ and $|E', \mathbf{k}\rangle$ as the ground state and continuum states dressed by the external laser field. Because of the detuning and the neglect of higher-order many-body effects, the Fano problem can be solved exactly. The dressed states can be explicitly written as a function of the density of states of the continuum and the coupling. The adiabaticity condition can then be put in the form $(\frac{d}{dt}\langle G'|)|E', \mathbf{k}\rangle \ll |\delta|; \forall \, \mathbf{k}$. It is possible to prove [12] that, for a reasonable density of states and energy dependence of the optical coupling, it is sufficient to satisfy this condition for $\mathbf{k} = 0$. This reduces to a two-level problem where Eq. (9) holds as a sufficient condition for adiabaticity.

A simple experimental setup with two dots of different sizes and a tunable laser could be used to check the spin entanglement from the interaction proposed here. The manipulation and measurements on a single dot can be realized selectively by exciting at the energy of the localized exciton. Single qubit operation on a selected dot can be carried out in a Raman configuration as proposed, for instance, in Ref. [2]. An external magnetic field has to be used to initialize the system. Quantum computation requires controllable spin-spin interaction between specific nearest neighbor pairs. Our proposed mechanism can induce spin-spin interaction. Near-field optical excitations can be used to ensure the interaction is between nearest neighbors. The dots in fact can be arranged in an array separated by distances of the order of the wavelength of light. Then quasidelocalized exciton states with subwavelength extension can be excited only between the two dots intended to be coupled. This possibility is well within the experimental state-of-the-art capabilities of near-field scanning optical microscopy [17]. In such a system, universal quantum computation can, in principle, be realized by using only the optically controlled exchange interaction, without resorting to single qubit operation [18]. As a possible extension to spintronics, we suggest that a regular array of charged dots may be magnetized by light which initializes the magnetization by exciting conduction electrons.

In conclusion, we have proposed an optical technique to generate and control the entanglement of the spin of two electrons localized in neighboring QDs. The control of the spin can be realized in the adiabatic regime and can lead to the realization of spin quantum gates useful for quantum information processing and to control of the magnetization of an ensemble of dots.

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