PHYSICAL REVIEW B **71**, 121305(R) (2005)

Indirect coupling between spins in semiconductor quantum dots

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(Received 3 December 2004; published 16 March 2005)

The optically induced indirect exchange interaction between spins in two quantum dots is investigated theoretically. We give a microscopic formulation of the interaction between the localized spin and the itinerant carriers, which is the basis of the indirect coupling, including the effects of correlation using a set of canonical transformations. Correlation effects are found to be of comparable magnitude to the direct exchange. We give quantitative results for the indirect spin-spin coupling for realistic quantum dot geometries and find the largest couplings for one-dimensional systems.

DOI: 10.1103/PhysRevB.71.121305 PACS number(s): 73.63.Kv, 78.67.Hc, 75.50.Pp, 03.67.Lx

Control of spins in semiconductors has been intensively investigated in recent years due to its potential for applications in spintronics and quantum computation.[1] Coherent coupling between localized spins is particularly sought after because it is a key requirement in proposals for spin-based implementations of quantum computation. Several coupling mechanisms have been proposed for quantum gates between spins in quantum dots (QDs). They include direct wave function overlap using electric gates at small dot separations[2] and exchange of a cavity photon mode between spins in QDs for large dot separations.[3]

Recently Piermarocchi *et al.* proposed an indirect mechanism to couple the two QD spins at intermediate interdot separations.[4] Here the interaction is mediated by virtual delocalized carrier excitations in the host material. The virtual excitations are driven by an interband off-resonance laser that provides optical control over the interaction. It effectively reduces the band-gap energy, thus increasing the interaction. This approach provides ultrafast optical control and long spin coherence times due to the virtual nature of the excitations. Combined with Raman optical transitions via intermediate trion states for single qubit operations,[5] this mechanism provides the necessary set of universal gates for quantum computing. This optically induced indirect spin exchange was considered in bulk semiconductors by Litvinov *et al.*[6] and is a variant of several analogous mechanisms, including the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction in metals,[7] Bloembergen-Rowland coupling in direct-gap semiconductors,[8] superexchange mediated by two holes in diluted magnetic semiconductors,[9] (DMS) and magnetic exchange mediated by bound correlated states (excitons).[10,11]

The proposal in Ref. 4 used qualitative estimates for the indirect coupling between spins. The purpose of the present work is to give quantitative results for this coupling for quantum dots. The key ingredient in all indirect spin coupling mechanisms is the exchange interaction of a localized spin with the mediating itinerant excitation. Here we give a microscopic formulation of this interaction, including the hybridization of continuum and dot states and the double occupancy of the dot, and we provide quantitative results for the spin coupling between QDs.

For the optically induced indirect interaction between spins in quantum dots, the spin-spin coupling is obtained from the self-energy correction in the continuum electron propagator due to its Coulomb interaction with each of the localized spins within second-order perturbation theory.[4] The result is a Heisenberg Hamiltonian for the localized spins, with an effective positive exchange constant[4]

$$J_{12}(R)=\frac{|\Omega|^2}{16}\int\frac{d^dk d^dk'}{(2\pi)^{2d}}\frac{|J(\mathbf{k},\mathbf{k}')|^2 e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}}}{\left(\delta+\frac{k^2}{2\mu}\right)^2\left(\delta+\frac{k^2}{2m_h}+\frac{k'^2}{2m_e}\right)} \tag{1}$$

where R is the distance between the dot centers, δ is the detuning of the laser from the electron-hole continuum, Ω is the Rabi energy for the light coupling to the electron-hole pair, and μ is its reduced mass. $J(\mathbf{k},\mathbf{k}')$ is the exchange interaction between the electron spin in the quantum dot and the itinerant electron.

To calculate $J(\mathbf{k},\mathbf{k}')$ we consider a Hamiltonian that includes the kinetic energy, the dot potential relative to the host material, and the electron-electron Coulomb interactions:

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_M+\mathcal{H}_1, \tag{2}$$

where

$$\mathcal{H}_0=\sum_\sigma E_d n_\sigma+\sum_{\mathbf{k},\sigma}E_k c^\dagger_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma}+U n_\uparrow n_\downarrow, \tag{3a}$$

$$\mathcal{H}_M=\sum_{\mathbf{k},\sigma}[V_k c^\dagger_{\mathbf{k}\sigma}c_{d\sigma}+T_k c^\dagger_{\mathbf{k}\sigma}c_{d\sigma}n_{-\sigma}+\text{H.c.}], \tag{3b}$$

$$\mathcal{H}_1=\sum_{\mathbf{k}\mathbf{k}',\sigma\sigma'}C^{\text{dir}}_{\mathbf{k}\mathbf{k}'}c^\dagger_{\mathbf{k}\sigma}c_{\mathbf{k}'\sigma}n_{\sigma'}+\sum_{\mathbf{k}\mathbf{k}'\sigma}C^{\text{ex}}_{\mathbf{k}\mathbf{k}'}c^\dagger_{\mathbf{k}\sigma}c_{d\sigma}c^\dagger_{d-\sigma}c_{\mathbf{k}'-\sigma}+\sum_{\mathbf{k}\mathbf{k}',\sigma}C^{\text{mix}}_{\mathbf{k}\mathbf{k}'}c^\dagger_{\mathbf{k}\sigma}c_{d\sigma}c^\dagger_{\mathbf{k}'-\sigma}c_{d-\sigma}+\text{H.c.} \tag{3c}$$

Here $c^\dagger_{d\sigma}(c^\dagger_{\mathbf{k}\sigma})$ is the creation operator for a localized (itinerant) electron, $n_\sigma=c^\dagger_{d\sigma}c_{d\sigma}$, and the last term in Eq. (3a) is the on-site Coulomb repulsion. $\mathcal{H}_M$ represents the hybridization of the localized and itinerant electrons where we include a population-dependent hybridization [second term in Eq.

(3b)]. This term was not included in previous work on coupling of localized and itinerant spins, and we find that it makes an important contribution to the spin exchange interaction. $\mathcal{H}_1$ contains the spin-independent and spin exchange Coulomb scattering; the latter gives rise to the Heitler-London exchange contribution.[12] The last term in Eq. (3c) describes localized and continuum state mixing. We have not included scattering between carriers in the continuum since the corresponding effects are not relevant to the problem of interest here. $V_k=\int d\mathbf{r}V_{\rm dot}\varphi_{\mathbf{k}}^*(\mathbf{r})\varphi_d(\mathbf{r})$ is the tunneling amplitude, where $\varphi_{\mathbf{k}}(r)[\varphi_d(r)]$ is the itinerant (localized) electron wave function, which are also used in the various Coulomb integrals in Eqs. (3a)–(3c).

We aim to bring the Hamiltonian $\mathcal{H}'=\mathcal{H}_0+\mathcal{H}_M$ to a form similar to that of an *s*-*d* spin exchange Hamiltonian by using a canonical transformation

$$\overline{\mathcal{H}'}=e^S\mathcal{H}'e^{-S}. \tag{4}$$

The unitary operator S is chosen to eliminate $\mathcal{H}_M$ to first order by requiring $\mathcal{H}_M=-[S,\mathcal{H}_0]$ and given by

$$S=\sum_{\mathbf{k}\sigma}[\beta_k+(\alpha_k-\beta_k)n_{-\sigma}]c_{d\sigma}^\dagger c_{\mathbf{k}\sigma}-\text{H.c.} \tag{5}$$

where

$$\alpha_k=\frac{V_k+T_k}{U+E_d-E_k},\quad \beta_k=\frac{V_k}{E_d-E_k}. \tag{6}$$

This is a generalized form of the Schrieffer-Wolff transformation, which was used to establish the connection between the Anderson and Kondo models.[13] It produces a contribution to the spin exchange arising from the correlation and hybridization terms in $\mathcal{H}'$, which is given to first order by

$$J^{(1)}(k,k')=\frac{1}{2}[\beta_kV_{k'}^*-\alpha_k(V_{k'}+T_{k'})^*]+[k\leftrightarrow k']^*. \tag{7}$$

This contribution vanishes when correlation effects are neglected ($U,T_k\to 0$). We find that the first-order result, given in Eq. (7), is inadequate because it requires that $\mathcal{H}_M$ be a small perturbation to $\mathcal{H}_0$, which is not the case generally. It is therefore necessary to sum up the infinite series in the transformed Hamiltonian

$$\overline{\mathcal{H}'}=\mathcal{H}_0+\sum_{n=1}^{\infty}\left(\frac{1}{n!}-\frac{1}{(n+1)!}\right)[S,\mathcal{H}_M]_n, \tag{8}$$

where $[S,\mathcal{H}_M]_n=[S,[S,\ldots,[S,\mathcal{H}_M]\ldots]]$. To this end we use a method suggested by Chan and Gulácsi[14] but employ a different strategy using a set of nested transformations. The first term in the series in Eq. (8) is

$$\begin{aligned}[S,\mathcal{H}_M]_1=\sum_{\mathbf{k}\mathbf{k}',\sigma}&[J_1(k,k')(c_{\mathbf{k}\sigma}^\dagger c_{d\sigma}c_{d-\sigma}^\dagger c_{\mathbf{k}'-\sigma}+n_{-\sigma}c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}'\sigma})\\&+P_1(k,k')(c_{\mathbf{k}\sigma}^\dagger c_{d\sigma}c_{\mathbf{k}'-\sigma}^\dagger c_{d-\sigma}+\text{H.c.})\\&-K_1(k,k')c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}'\sigma}]+\sum_\sigma[G_1n_\sigma+I_1n_\sigma n_{-\sigma}],\end{aligned} \tag{9}$$

where $J_1(k,k')=2J^{(1)}(k,k')$ is given in Eq. (7), and the rest of the coefficients in Eq. (9) are $P_1(k,k')=\frac{1}{2}[\alpha_kV_{k'}^*-\beta_k(V_{k'}+T_{k'})^*]+[k\leftrightarrow k']^*$, $K_1(k,k')=\beta_kV_{k'}^*+\beta_{k'}^*V_k$, $G_1=2\Sigma_{\mathbf{k}}\beta_kV_k^*$, and $I_1=2\Sigma_{\mathbf{k}}[\alpha_k(V_k+T_k)^*-\beta_kV_k^*]$. We find that the second-order term has the same form as $\mathcal{H}_M$ apart from higher-order correlation terms. We estimate the magnitude of these continuum scattering terms by neglecting off-diagonal contributions and placing lower and upper bounds of zero and one on the occupation numbers.[15] This procedure brings all higher odd orders in the series to the form of Eq. (9), and we are able to sum the series by solving the following set of recursion relations for the several coefficients:

$$\begin{aligned}J_{m+1}(k,k')=&2G_m(\alpha_k\alpha_{k'}^*-\beta_k\beta_{k'}^*)+4I_m\alpha_k\alpha_{k'}^*\\&-\sum_{\mathbf{k}''}\{[2J_m(k,k'')\alpha_{k''}\alpha_{k'}^*+2P_m(k,k'')\beta_{k''}\alpha_{k'}^*\\&-K_m(k,k'')(\alpha_{k''}\alpha_{k'}^*-\beta_{k''}\beta_{k'}^*)]+[k\leftrightarrow k']^*\},\end{aligned}$$

$$\begin{aligned}P_{m+1}(k,k')=&I_m(\beta_k\alpha_{k'}^*+\alpha_k\beta_{k'}^*)-\sum_{\mathbf{k}''}\left\{\left[J_m(k,k'')\alpha_{k''}\beta_{k'}^*\right.\right.\\&\left.+P_m(k,k'')\beta_{k''}\beta_{k'}^*-\frac{1}{2}K_m(k,k'')(\alpha_{k''}\beta_{k'}^*-\beta_{k''}\alpha_{k'}^*)\right]\\&\left.+[k\leftrightarrow k']^*\right\},\end{aligned}$$

$$K_{m+1}(k,k')=-2G_m\beta_k\beta_{k'}^*-\sum_{\mathbf{k}''}[K_m(k,k'')\beta_{k''}\beta_{k'}^*+(k\leftrightarrow k')^*],$$

$$G_{m+1}=-2G_m\sum_{\mathbf{k}}|\beta_k|^2-2\sum_{\mathbf{k},\mathbf{k}'}K_m(k,k')\beta_k\beta_{k'}^*,$$

$$\begin{aligned}I_{m+1}=&-2\sum_{\mathbf{k}}[G_m(|\alpha_k|^2-|\beta_k|^2)+2I_m|\alpha_k|^2]+2\sum_{\mathbf{k},\mathbf{k}'}[P_m(k,k')\\&\times(\beta_k\alpha_{k'}^*+\alpha_k\beta_{k'}^*)+2J_m(k,k')\alpha_k\alpha_{k'}^*\\&-K_m(k,k')(\alpha_k\alpha_{k'}^*-\beta_k\beta_{k'}^*)].\end{aligned} \tag{10}$$

Equations (10) are obtained from the lower bound in the higher-order contributions, and a second set of equations is obtained for the upper bound.

The exchange contribution is obtained from the odd orders of the series in Eq. (8). The odd orders also contain additional terms that renormalize the original Hamiltonian (3a). The even orders also are summed up, and they renormalize the hybridization Hamiltonian (3b). Figure 1(a) shows the result of the series summation for the diagonal part of the exchange, $J(k,k)$. We find that it differs appreciably from the first-order result of Eq. (7) and therefore the residual hybridization in the even orders need not be small, as shown in Fig. 1(b). Thus we need to perform a second canonical transformation by applying Eqs. (5) and (6) to the renormalized Hamiltonian. This second transformation eliminates the next order in the hybridization terms and further corrects the resulting exchange contribution. This process is reiterated until we fully eliminate the hybridization part of the Hamiltonian, as shown in Fig. 1(b).

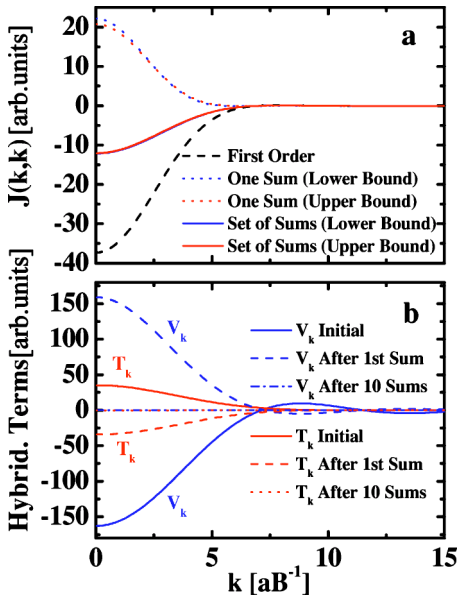

FIG. 1. (Color online) (a) Diagonal matrix elements of the spin exchange interaction between localized and itinerant electrons in a 2D host. The figure gives the first-order result from Eq. (7) (dashed line), intermediate results after the summation in Eq. (8) (dotted lines), and final results obtained by performing a set of transformations (solid lines). (b) The corresponding hybridization terms: tunneling amplitude V_k and population-dependent hybridization term T_k.

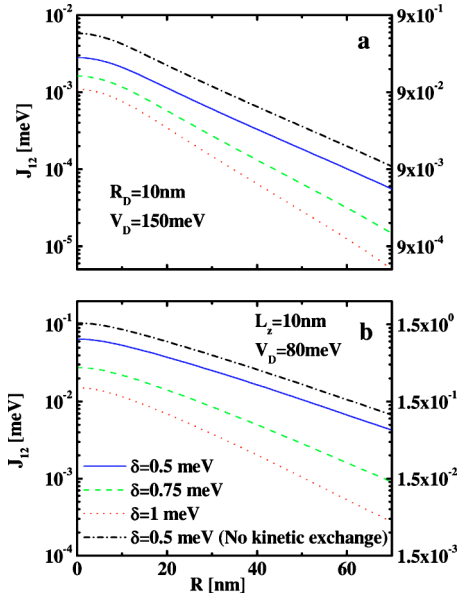

FIG. 2. (Color online) (a) Optically induced spin-spin exchange coupling in a 2D host vs the distance between the centers of the dots for dot radius $R_D=10$ nm, potential height $V_e=150$ meV and several values of laser detunings. The dashed-dotted line corresponds to the interaction without the kinetic exchange contribution and is provided for comparison. The right axis shows the coupling including excitonic effects discussed in the text. (b) The same as a for a quasi-1D host and cylindrical dots with $L_z=10$ nm, $R_D=5$ nm, and potential height $V_e=80$ meV.

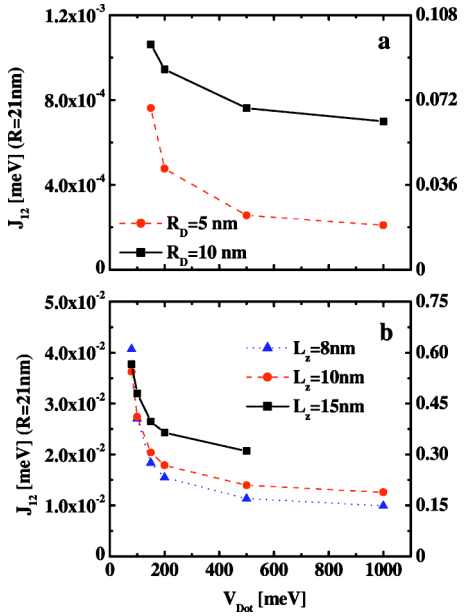

FIG. 3. (Color online) (a) Spin-spin coupling in a 2D host at a dot separation of 21 nm vs the dot potential for two dot radii. The laser detuning is $\delta=0.5$ meV. The right axis shows the coupling values after excitonic corrections. (b) The same as a for a quasi-1D host and cylindrical dots with $R_D=5$ nm and several dot heights.

This procedure of applying a set of nested Schrieffer-Wolff transformations is essential to obtain quantitative results for the "kinetic" exchange interaction, which is the part that results from the hybridization terms in Eq. (3b). As seen in Fig. 1(a), $J(\mathbf{k},\mathbf{k}')$ is ferromagnetic after one transformation, which differs from other results for this kinetic exchange contribution, e.g., those in a renormalization group approach.[16] Only after a set of transformations (typically 10–20) are the renormalized hybridization terms eliminated and the antiferromagnetic nature of the kinetic contribution to the interaction is restored, albeit with a modified magnitude compared to the first-order result. The results calculated with the lower and upper bounds discussed above are remarkably close to one another. We have verified that they coincide within 10% for a wide range of geometries and dot potentials; thus we believe that our summation represents the complete Schrieffer-Wolff transformation with a good accuracy.[17] In our calculations we used $m_e=0.07m$, and $m_h=0.5m$. The localized electron wave function was taken to be Bessel functions in the lateral direction and a combination of cosine and exponential functions in the z direction.

Since the kinetic exchange interaction that we calculate from the transformed $\mathcal{H}'$ is antiferromagnetic, it competes with the ferromagnetic exchange given by the second term in Eq. (3c). An accurate evaluation of the former is important as it can lead to an order of magnitude difference or even a change of sign in the total spin exchange coupling between localized and itinerant electrons. We point out that a full transformation is also valuable in the case where $U+E_d>0$, leading to a divergence of α_k in Eq. (6). Here, the kinetic exchange contribution is dominant and cannot be obtained via a perturbative approach. This regime corresponds to dots with small size ($R_D \lesssim 5$ nm) and shallow potential (barrier

$\leqslant$80 meV), which are not typical for physical systems and are not considered here.

In Fig. 2 we show the results for the spin-spin coupling J_{12} [Eq. (1)], including both the kinetic and Coulomb potential exchange contributions. J_{12} calculated with the potential exchange contribution alone is also shown in Fig. 2, and is a factor of 2 larger than J_{12} calculated with the kinetic exchange contribution.[18] Figure 2(a) shows the spin coupling for lateral cylindrical dots in a two-dimensional quantum well. The results for vertically stacked cylindrical dots in a quasi-one-dimensional wire are given in Fig. 2(b). Here we used $\Omega = 0.1$ meV for the optical vertices. It is seen that the spin coupling is more than an order of magnitude larger for the one-dimensional case than for the two-dimensional case.

The Coulomb interaction between the intermediate virtual electrons and holes results in an enhancement of the oscillator strength at both the optical and spin vertices due to the exciton wave functions.[4] We have evaluated the dominant contribution of the electron-hole interaction to J_{12}. It results in an increase of up to two orders of magnitude in the two-dimensional case and roughly one order of magnitude in the one-dimensional case right axes in Fig. 2). Thus, the excitonic effects reduce the difference in J_{12} between the two geometries.

Figure 3 shows the dependence of the spin-spin coupling on the dot potential and size. Larger dots give larger couplings but require larger separation to avoid overlap. The increase in the coupling as the dot potential decreases is mainly due to the reduction of the kinetic exchange contribution, particularly evident in the one-dimensional case.

A technologically viable way to increase J_{12} is by using a microcavity. This can be done by growing distributed Bragg reflector layers on the top and bottom of the active semiconductor layer containing the QDs. This can increase the electric field by orders of magnitude, and thus increases the Rabi energy at the optical vertices in Eq. (1).

We have shown that the effect of hybridization of continuum and dot states produces a sizable contribution to the exchange coupling between localized and itinerant electrons. For certain dot geometries this kinetic exchange can even lead to a change of sign in the spin exchange interaction. A set of canonical transformations with summations over higher-order terms provides a useful tool to evaluate the spin exchange interaction. Our transformation of the Hamiltonian (2) captures the multiple scattering processes involved in the interaction between the localized and itinerant carriers, and it provides the first microscopic description that accounts quantitatively for the exchange interaction.[19] Our formulation is also applicable to other systems of localized spins coupled by carriers, such as electrons bound to donors,[20] magnetic impurities,[21] and nuclear spins.[22]

This work was supported by ONR, DARPA, and the ONR Nanoscale Electronics Program. G. R. thanks A. K. Rajagopal for helpful discussions and acknowledges a NRL/NRC Research Associateship.

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[17] Applying our transformations to the total Hamiltonian, including $\mathcal{H}_1$, poses several computational difficulties. The spin couplings, including an estimate of the exchange contributions that result from this modification, do not differ significantly from our results given here.

[18] The main difference between the present results for the potential contribution to the exchange coupling and the estimates in Ref. 4 arise from the choice of much larger dots and the use of a contact approximation that replaces $J_{kk'}$ with a constant in that work.

[19] Multiple scattering effects were treated in Ref. 11, where the problem of two localized spins interacting via an itinerant exciton was solved utilizing scattering matrix formalism. This approach goes beyond the second-order perturbation result given in Eq. (1) but requires the use of a model separable potential.

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