Indirect coupling between spins in semiconductor quantum dots

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

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Control of spins in semiconductors has been intensively investigated in recent years due to its potential for applications in spintronics and quantum computation. Coherent coupling between localized spins is particularly sought after because it is a key requirement in any proposal for spin-based implementation of quantum computation. Several coupling mechanisms have been proposed to construct quantum gates between spins in quantum dots (QDs). These include direct wavefunction overlap using electric gates where the interdot separation is small and exchange of a cavity photon mode between spins in QDs for a large interdot separation.

Recently Piermarocchi et al. proposed to use an indirect mechanism to couple the two QD spins at intermediate interdot separations. 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 and serves to reduce the bandgap energy, thus extending the interaction range. The proposed scheme has the advantages of ultrafast optical control and long spin coherence times due to the virtual nature of the excitations. Combined with the proposal to use Raman optical transitions via intermediate trion states for single qubit operations, this mechanism provides the necessary set of universal gates for quantum computing. This optically induced indirect spin exchange is a variant of several analogous mechanisms. These include the RKKY interaction in metals and Bloembergen-Roundland coupling in direct-gap semiconductors, the superexchange mediated by two holes in diluted magnetic semiconductors (DMS), the magnetic exchange mediated by bound correlated states (excitons), and ferromagnetism induced by virtual Mn acceptor level-valence band transitions in DMS materials.

A key ingredient in all of these indirect spin coupling mechanisms is the exchange interaction of a localized spin with the mediating itinerant excitation. The purpose of the present work is to introduce a microscopic formulation that provides a quantitative description of this exchange interaction by taking into account the effects of hybridization of continuum and dot states and the double occupancy in the dot.

For the case of the optically induced indirect interaction between spins in quantum dots, the spin-spin coupling is obtained by considering 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. The result is a Heisenberg Hamiltonian involving the localized spins, with an effective positive exchange constant that is given by

$$J_{12}(R) = \frac{[\Omega]^2}{16} \int \frac{d^2k d^2k'}{(2\pi)^2} \frac{J(k, k')^2 e^{-i(k-k') \cdot R}}{\left(\delta + \frac{k^2}{2m_e} + \frac{k'^2}{2m_e}\right)^2}$$

(1)

where \( R \) is the distance between the dot centers, \( \delta \) is the detuning of the laser with respect to the electron-hole continuum, \( \Omega \) is the Rabi energy for the light coupling to the electron-hole pair and \( \mu \) is its reduced mass. \( J(k, k') \) is the exchange interaction between the spin in the quantum dot and the itinerant electron.

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

$$H = H_0 + H_M + H_1$$

(2)

where

$$H_0 = \sum_{\sigma} E_d n_\sigma + \sum_{k, \sigma} E_k c_{k\sigma}^\dagger c_{k\sigma} + Un_\uparrow n_\downarrow$$

(3a)

$$H_M = \sum_{k, \sigma} \left[ V_k c_{k\sigma}^\dagger c_{d\sigma} + T_k c_{k\sigma}^\dagger c_{d\sigma} n_{-\sigma} + h.c. \right]$$

(3b)

$$H_1 = \sum_{kk', \sigma\sigma'} C_{\sigma\sigma'}^{\text{dir}} c_{k\sigma}^\dagger c_{k'\sigma'} n_{\sigma'} + \sum_{kk', \sigma} C_{\sigma\sigma'}^{\text{exx}} c_{k\sigma}^\dagger c_{d\sigma}^\dagger c_{d-\sigma}^\dagger c_{k'\sigma'} - \sum_{kk', \sigma} C_{\sigma\sigma'}^{\text{mix}} c_{k\sigma}^\dagger c_{d\sigma}^\dagger c_{k'\sigma'} n_{\sigma'} c_{d-\sigma} + h.c.$$  

(3c)

Here \( c_{d\sigma}^\dagger \) (\( c_{d\sigma} \)) is the creation operator for a localized (itinerant) electron, \( n_{\sigma} = c_{d\sigma}^\dagger c_{d\sigma} \), and the last term in Eq. \( 3c \) is the on-site Coulomb repulsion. \( H_M \) represents the hybridization of the localized and itinerant electrons, where we include a population dependent hybridization (2nd term in Eq. \( 3b \)), which was absent...
in previous works on coupling of localized and itinerant spins. We show below that this latter term 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 contributions. The last term in Eq. (3k) describes the effect of localized and continuum state mixing. We have neglected scattering processes between carriers in the continuum since the corresponding effects are not relevant to the problem we wish to solve. \(V_k = \int \frac{d\mathbf{r}d\mathbf{r}'}\nu_{\mathbf{r}} \nu_{\mathbf{r}'} (\mathbf{r}) \nu_{\mathbf{r}'} (\mathbf{r})\) is the tunnelling amplitude, where \(\nu_{\mathbf{r}} (\mathbf{r})\) is the itinerant (localized) electron wave function, also used to calculate various Coulomb integrals in Eq. (2).

We aim at bringing the Hamiltonian \(\mathcal{H}' = \mathcal{H}_0 + \mathcal{H}_M\) to a form similar to that of an s-d spin exchange Hamiltonian by applying a canonical transformation
\[
\tilde{\mathcal{H}}' = e^S \mathcal{H}' e^{-S}. \tag{4}
\]
The unitary operator \(S\) is constructed to eliminate \(\mathcal{H}_M\) to first order by requiring \(\mathcal{H}_M = -[S, \mathcal{H}_0]\) and is given by
\[
S = \sum_{k} \left[ \beta_k + (\alpha_k - \beta_k) n_{-\sigma} \right] c_{d\sigma}^\dagger c_{k\sigma} - h.c. \tag{5}
\]
where
\[
\alpha_k = \frac{V_k + T_k}{U + E_d - E_k} : \beta_k = \frac{V_k}{E_d - E_k}. \tag{6}
\]
This is a generalized form of the Schrieffer-Wolff transformation, which was first used to establish the connection between the Anderson and Kondo models. 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
\[
J^{(1)} (k, k') = \frac{1}{2} \left[ \beta_k V_k^* - \alpha_k (V_k + T_k)^* \right] + [k \leftrightarrow k']^* . \tag{7}
\]
This contribution vanishes when correlation effects are neglected \((U, T_k \rightarrow 0)\). We find that the first order result, given in Eq. (7), is inadequate because it requires that \(\mathcal{H}_M\) would 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
\[
\tilde{\mathcal{H}}' = \mathcal{H}_0 + \sum_{n=1}^\infty \left( \frac{1}{n!} - \frac{1}{(n+1)!} \right) [S, \mathcal{H}_M]_n, \tag{8}
\]
where \([S, \mathcal{H}_M]_n = [S, [S, ..., [S, \mathcal{H}_M] ...]]\). To this end we use a method suggested by Chan and Gulicsits but employ a different strategy to solve the problem. The first term in the series in Eq. (8) is
\[
[S, \mathcal{H}_M]_1 = \sum_{kk'\sigma} \left[ J_1 (k, k') \left( c_{k\sigma}^\dagger c_{d\sigma} c_{k'\sigma} + c_{d\sigma} c_{k\sigma} c_{k'\sigma} + h.c. \right) - K_1 (k, k') c_{k\sigma}^\dagger c_{k'\sigma} + \sum_{\sigma} [G_1 n_{\sigma} + I_1 n_{\sigma} n_{-\sigma}] \right] \tag{9}
\]
where \(J_1 (k, k') = 2J^{(1)} (k, k')\) is given in Eq. (7), and the rest of the coefficients in Eq. (9) are
\[
\begin{align*}
J_1 (k, k') &= \frac{1}{2} \left[ \alpha_k V_k^* - \beta_k (V_k + T_k)^* \right] + [k \leftrightarrow k']^*, \\
K_1 (k, k') &= \beta_k V_k^* + \beta_k V_k, \\
G_1 &= 2 \sum_k \beta_k V_k^*, \\
I_1 &= 2 \sum_k \left[ \alpha_k (V_k + T_k)^* - \beta_k V_k^* \right].
\end{align*}
\]
Calculating the second order term we find that it has the same form of \(\mathcal{H}_M\) 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 on the occupation numbers. 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
\[
\begin{align*}
J_{m+1} (k, k') &= 2G_m (\alpha_k \alpha_{k'}^* - \beta_k \beta_{k'}^*) + 4I_m \alpha_k \alpha_{k'}^* - \\
& \quad \sum_{k''} \left\{ 2J_m (k, k'') \alpha_{k''} \alpha_{k'}^* + 2P_m (k, k'') \beta_{k''} \alpha_{k'}^* - \\
& \quad K_m (k, k'') (\alpha_{k''} \alpha_{k'}^* - \beta_{k''} \beta_{k'}^*) + [k \leftrightarrow k']^* \right\}, \\
P_{m+1} (k, k') &= I_m (\beta_k \alpha_{k'}^* + \alpha_k \beta_{k'}^*) - \\
& \quad \sum_{k''} \left\{ J_m (k, k'') \alpha_{k''} \beta_{k'}^* + P_m (k, k'') \beta_{k''} \beta_{k'}^* - \\
& \quad \frac{1}{2} K_m (k, k'') (\alpha_{k''} \beta_{k'}^* - \beta_{k''} \alpha_{k'}^*) + [k \leftrightarrow k']^* \right\}, \\
K_{m+1} (k, k') &= -2G_m \beta_k \beta_{k'}^* - \\
& \quad \sum_{k''} \left[ K_m (k, k'') \beta_{k''} \beta_{k'}^* + (k \leftrightarrow k')^* \right].
\end{align*}
\tag{10}
\]
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 case.

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

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 one that results from the hybridization terms in Eq. (3b). As seen in Figure 1a, $J(k, k')$ is ferromagnetic for one transformation, which would differ from other results for this kinetic exchange contribution, e.g., in a renormalization group approach. Only after a set of transformations (typically 10-20) are the renormalized hybridization terms eliminated and the antiferromagnetic nature of 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. In order to address the possibility that the off-diagonal contributions from the continuum scattering terms might alter our results, we have estimated them from limits of the off diagonal density factors. The results lie between the lower and upper bounds given in figure 1a; thus we believe that our summation represents the complete Schrieffer-Wolff transformation with a good accuracy.

Since the kinetic exchange interaction that we calculate from the transformed $\hat{H}$ is antiferromagnetic, it competes with the ferromagnetic exchange given by the second term in Eq. (3c). Thus, 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. A full transformation is also valuable in the case where $U + E_d > 0$, leading to a divergence of $\alpha_s$ in Eq. (3d). Here, the kinetic exchange contribution is dominant and cannot be obtained via a perturbative approach. This regime corresponds to dots with small size ($R_D \leq 5nm$) and shallow potential (barrier $\leq 80meV$), which are not typical for physical systems and are not considered here.

In figure 2 we show the results for the spin-spin coupling $J_{12}$ [Eq. (11)], incorporating all the exchange contributions. Figure 2a 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 figure 2b. Here we used $m_e = 0.07m$, $m_h = 0.5m$ and $\Omega = 0.1meV$. The localized electron wave function was taken to be a combination of Bessel functions in the lateral direction and a combination of Cosine and Exponential functions in the $z$ direction. 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 the optical and spin vertices due to the exciton wave functions. We have evaluated the dominant contribution of the electron-hole interaction to
radii. The laser detuning is $\delta$ the coupling values after excitonic corrections; (b) Same as a $R$ for a quasi 1D host and cylindrical dots with $J$ several dot heights.

Thus, the excitonic effects reduce the difference of magnitude in the one-dimensional case (right axes in figure 2). 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 figure 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 necessitate 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.

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. Placing the active layer at the antinode of the microcavity increases 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. Our formulation is also applicable to other systems of localized spins coupled by carriers, such as electrons bound to donors $^{12}$, magnetic impurities $^{13}$, and nuclear spins $^{14}$.

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