Magnetic exchange coupling mediated by bound states

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Usually indirect exchange coupling is mediated by unbound, noncorrelated intermediate states (Ruderman-Kittel-Kasuya-Yosida–like mechanisms) or by unbound, correlated intermediate states (superexchange-like mechanisms). Here, we investigate the possibility of indirect magnetic exchange coupling mediated by bound, correlated intermediate states. As a concrete example we study the magnetic coupling between two magnetic impurities embedded in a semiconductor matrix. The importance of long-ranged attractive Coulomb interactions between electrons and holes is emphasized. This attraction leads to exciton bound states which act as mediators of the effective exchange interaction between the two impurities. The resulting exchange interaction presents strong temperature dependence and can be analyzed in terms of the symmetry of the internal wave function of the exciton bound states. Possible applications of these results may include recent experimental results on ferromagnetic metal-semiconductor multilayers.

Typically, indirect magnetic exchange interactions are thought of as being mediated by unbound, noncorrelated intermediate states as it is the case for Ruderman-Kittel-Kasuga-Yosida (RKKY)-like mechanisms when applied to metals[1] or to semiconductors.[2] In addition, the indirect magnetic exchange interaction is also thought of as being mediated by unbound, correlated intermediate states as it is the case for the superexchange-like mechanisms when applied to insulating transition-metal oxides.[3] Our main aim in this paper is to describe a situation which is completely different from the two described above. Namely a case where the indirect magnetic exchange coupling is mediated by a bound, correlated intermediate state. The choice of a system to study the proposed effect is very important. We choose to study the concrete example of the indirect magnetic exchange between two magnetic impurities embedded in a semiconductor matrix, which may be extended to the case of a magnetic impurity lattice. Our analysis is not only of academic interest but also may be brought in contact with recent experiments on ferromagnetic metal-semiconductor multilayers like Fe/FeSi,[4] Fe/Si,[5] MnTe/CdTe, and EuTe/PdTe.[6]

Now let us briefly discuss the central physical picture of the mechanism. Consider two magnetic impurities inside of a direct gap semiconductor and allow an attractive Coulomb interaction between an electron in the conduction band and a hole in the valence band. The Coulomb interaction binds electron-hole pairs into excitons, which exist in the gap of the semiconductor (excitons are stable bound states with relatively long lifetimes). As a result, these bound states provide intermediate states inside the gap which can mediate the exchange of spin information between the two magnetic impurities. This mediation is possible since excitons, although chargeless, can carry spin information, i.e., can be singlet or triplet. It is also important to emphasize that these bound states inside the gap can mediate the indirect magnetic coupling more effectively than the unbound electron-hole pairs, which have energies greater than the gap size.

As a result of the existence of these bound states inside the gap of the semiconductor, the magnetic exchange coupling increases with temperature. This increase in the indirect coupling with increasing temperature is a natural consequence of the population increase of these bound states caused by thermal excitations. In addition to this thermal effect, we shall see that the finite extension of the excitons internal wave functions and their symmetries play an important role in the determination of the *magnitude* and *sign* of the effective exchange coupling $J_{\rm eff}$.

Let us begin our analysis with the choice of the model and the geometry. First, let us study the situation of two magnetic impurities embedded in an isotropic and direct gap semiconductor matrix in order to establish the basic mechanism of the effective magnetic exchange. The starting Hamiltonian is of the following form:

$$H = H_a + H_c + H_v + H_{cv} + H_{acv} \ , \tag{1}$$

where $H_a = \sum_{i,\sigma} \epsilon_\sigma C^\dagger_\sigma(R_i) C_\sigma(R_i)$ is the kinetic energy term of the magnetic impurities located at positions R_i, with $i=1,2$; $H_c = \sum_{k_e,\sigma_e} \varepsilon_c(k_e) f^\dagger_{c,\sigma_e}(k_e) f_{c,\sigma_e}(k_e)$ is the kinetic energy for the conduction electrons in the semiconductor matrix, with $\varepsilon_c(k_e) = E_c + k_e^2/2m_e - \mu$; $H_v = \sum_{k_h,\sigma_h} \varepsilon_v(k_h) f^\dagger_{v,\sigma_h}(k_h) f_{v,\sigma_h}(k_h)$ is the kinetic energy of the valence electrons with dispersion $\varepsilon_v(k_h) = E_v - k_h^2/2m_h - \mu$. Here the gap of the semiconductor is $E_g = E_c - E_v$. The Coulomb interaction between an electron in the conduction band and a hole in the valence band is

$$H_{cv} = \sum{}' V_{\sigma_e,\sigma_h}(q) f^\dagger_{c,\sigma_e}(k_e+q) f_{c,\sigma_e}(k_e) \times f^\dagger_{v,\sigma_h}(k_h-q) f_{v,\sigma_h}(k_h) \ ,$$

with the summation (′) being over $(k_e, k_h, q, \sigma_e, \sigma_h)$, and finally

$$H_{acv}=-\sum{}''\tilde{J}\tilde{\delta}f^{\dagger}_{c,\sigma_e}(k_e)f_{v,\sigma_h}(k_h)C^{\dagger}_{\sigma'}(R_i)C_{\sigma}(R_i)+\text{H.c.}$$

is the *exchange* interaction between the local magnetic impurity and an electron in the conduction band and a hole in the valence band. Here, the summation ('') is over $(k_e,k_h,\sigma,\sigma',\sigma_e,\sigma_h)$;

$$\tilde{J}=J^{\sigma,\sigma'}_{\sigma_e,\sigma_h}(k_e,k_h)\exp[i(k_h-k_e)\cdot R_i]$$

is the *exchange* coupling and the spin conservation function $\tilde{\delta}=\delta_{\sigma_e,\sigma}\cdot\delta_{\sigma_h,\sigma'}$.

We continue our approach by constructing the action of the system

$$S=\int_0^{\beta}d\tau[K(\tau)-H(\tau)]\ ,$$

where $K(\tau)=K_a+K_c+K_v$, with the first term being $K_a=\sum_{i,\sigma}C^{\dagger}_{\sigma}(R_i,\tau)\partial_{\tau}C_{\sigma}(R_i,\tau)$, the second being $K_c=\sum_{k_e,\sigma_e}f^{\dagger}_{c,\sigma_e}(k_e,\tau)\partial_{\tau}f_{c,\sigma_e}(k_e,\tau)$, and the last being

$$K_v=\sum_{k_h,\sigma_h}f^{\dagger}_{v,\sigma_h}(k_h,\tau)\partial_{\tau}f_{v,\sigma_h}(k_h,\tau)\ .$$

Now we use the functional integral formalism to derive the effective action which involves only the fermion variables of the magnetic atoms. Upon functional integration of the valence and conduction fermions with actions $S_c=\int_0^{\beta}d\tau[K_c-H_c]$, $S_v=\int_0^{\beta}d\tau[K_v-H_v]$ and interaction actions $S_{cv}=-\int_0^{\beta}d\tau H_{cv}(\tau)$ and $S_{lcv}=-\int_0^{\beta}d\tau H_{lcv}(\tau)$ we obtain

$$\int Df^{\dagger}Df\,[S_c+S_v+S_{cv}+S_{acv}]\approx\exp[S_{\text{eff}}]$$

and arrive at the effective action $S_{\text{eff}}=-H_{\text{eff}}$.[7] The effective interaction is given by

$$H_{\text{eff}}(\mathbf{R}_{12})=-\sum_{\sigma,\sigma',i\omega,\nu_1,\nu_2}J^{\sigma,\sigma'}_{\text{eff}}(\mathbf{R}_{12},i\omega)C^{\dagger}_{\sigma}(R_1,i\nu_1-i\omega)C_{\sigma'}(R_1,i\nu_1)C^{\dagger}_{\sigma'}(R_2,i\nu_2)C_{\sigma}(R_2,i\nu_2-i\omega)\ , \tag{2}$$

where $\mathbf{R}_{12}=\mathbf{R}_2-\mathbf{R}_1$ is the relative separation between the two magnetic atoms and

$$J^{\sigma,\sigma'}_{\text{eff}}(\mathbf{R}_{12},i\omega)=\sum_q J^{\sigma,\sigma'}_{\text{eff}}(q,i\omega)\exp[-i\mathbf{q}\cdot(\mathbf{R}_2-\mathbf{R}_1)]$$

with $J^{\sigma,\sigma'}_{\text{eff}}(q,i\omega)$ being just the generalized exchange-coupling tensor due to the exchange of an electron-hole pair between the two magnetic impurity atoms. The spin dependence of $J^{\sigma,\sigma'}_{\text{eff}}$ is just a consequence of the spin-dependent interaction $V_{\sigma_e,\sigma_h}(q)$ between the conduction-band electrons and the valence-band holes.

When the interaction $V_{\sigma_e,\sigma_h}(q)$ is spin independent, the singlet and triplet excitons have the same energy. Such a situation just reflects the nonexistence of the Pauli principle between conduction electrons and valence holes.[8] This is the case for the direct Coulomb interaction $V_{\sigma_e,\sigma_h}(q)=4\pi e^2/\epsilon q^2$, where ϵ is the dielectric constant of the semiconductor, that we consider here. Because of the static Coulomb interaction between the conduction electrons and the valence holes and determination of $J^{\sigma,\sigma'}_{\text{eff}}$ does not follow from the noninteracting susceptibility[2] but involves the solution of a Bethe-Salpeter (BS) equation for the vertex function Γ. Diagramatically $J^{\sigma,\sigma'}_{\text{eff}}(\mathbf{R}_{12},i\omega)$ corresponds to a bubble diagram with multiple static Coulomb line insertions. Using the techniques of functional integration[9] the resulting Bethe-Salpeter equation takes the well-known form[10]

$$\Gamma(k,k';q)=J(k,k';q)+\sum_{k''}V(k,k'';q)G_h(k''-q/2)G_e(k''+q/2)\times\Gamma(k'',k';q)\ , \tag{3}$$

where we have suppressed the spin indices. In the present situation we have no Pauli exclusion principle between conduction electrons and valence holes since the band index quantum number is different. In (3) the noninteracting single-particle Green's functions are $G_h(k)=1/[i\omega-\varepsilon_v(\mathbf{k})]$ and $G_e(k)=1/[i\omega-\varepsilon_c(\mathbf{k})]$.

To explicitly solve the BS equation defined in (3), we consider only the ladder diagrams since we will confine ourselves to the limit of low density of electrons and holes, i.e., $T\ll E_g$, where the crossed diagrams can be safely neglected. We focus our attention on the contribution to $J_{\text{eff}}(\mathbf{R}_{12},i\omega)$ coming from the bound states (excitons) only, which amounts to solving the homogeneous BS equation, given that in the vicinity of the bound-state energy E_b the bare interaction $J(k,k';q)$ is finite and therefore can be neglected. As a result of the summation over frequency and the redefinition $\psi(k)\equiv\Gamma(k,k';q)/\Lambda(k)$, where

$$\Lambda(k)=[E_g+\varepsilon_c(k+q/2)+\varepsilon_v(k-q/2)-i\omega]\ .$$

We can transform the homogeneous BS equation, i.e., Eq. (3) without $J(k,k';q)$, into a Schrödinger-like equation for the bound electron-hole pair (exciton)

$$[\varepsilon_c(k+q/2)+\varepsilon_v(k-q/2)+E_g-i\omega]\psi(k)=\sum_{k''}V(k-k'')\psi(k'')\ , \tag{4}$$

which is valid in the limit of low density of electrons and holes, i.e., $E_g\gg T$ so that the occupation factors of electrons in the conduction band and holes in the valence bands are negligible in comparison to unity. We will not be concerned with the scattering states of (4) given that they have energies larger than E_g. We focus our attention only on the bound states of (4) which have energies smaller than E_g and thus can provide a more effective magnetic coupling than the scattering states. From the eigenvalues $\varepsilon_\lambda(q)$ and eigenfunctions $\psi(k)=\phi_\lambda(k;q)$ of (4) we con-

struct the approximate Green's function for the bound state

$$K_\lambda(k,q,i\omega)\approx\frac{\phi^*_\lambda(k;q)\phi_\lambda(k;q)}{i\omega-\omega_\lambda(q)}\ , \tag{5}$$

from which we can obtain the effective interaction

$$J^{\sigma_e\sigma'}_{\rm eff}(q,i\omega)=\sum_{k,\lambda,\sigma_e,\sigma_h}|J^{\sigma\sigma'}_{\sigma_e,\sigma_h}(k,q)|^2\tilde{\delta}K_\lambda(k,q,i\omega)\ . \tag{6}$$

The explicit calculation of $J^{\sigma_e\sigma'}_{\rm eff}(q,i\omega)$ can be done with the help of (5) and (6). In addition, we need the expressions for the eigenfunctions of (4) given by

$$\phi_\lambda(k;q)=\int d\mathbf{r}\exp[i\mathbf{k}\cdot\mathbf{r}]\exp[i\gamma\mathbf{q}\cdot\mathbf{r}]\xi_\lambda(\mathbf{r})\ , \tag{7}$$

where $\xi_\lambda(\mathbf{r})$ are the hydrogenic wave functions for the relative coordinate of the electron-hole bound state (exciton) with quantum numbers $\lambda\equiv(n,l,m)$. The corresponding eigenenergies are $\varepsilon_\lambda(q)=E_g-\mathcal{R}/n^2+\hbar^2q^2/2M$, where $M=m_e+m_h$, $\mathcal{R}=\mu R_y/m_e\epsilon^2$ with μ being the reduced mass of the bound electron-hole pair and $R_y=13.6$ eV being the Rydberg. The characteristic size of the bound electron-hole pair in the state $\lambda\equiv(n,l,m)$ is $r_{\rm eh}=n^2a_0$, where $a_0=\epsilon(m/\mu)a_B$ is the Bohr radius of the electron-hole problem, while $a_B=0.529$ Å is just the Bohr radius for the hydrogen atom. The additional parameter appearing in (7) is expressed as $\gamma=\Delta m/2M$, where $\Delta m=m_e-m_h$.

We assume the simplest situation when $J^{\sigma,\sigma'}_{\sigma_e,\sigma_h}=J$ is independent of momenta and spins, i.e., the initial *exchange* interaction is taken to be local (zero ranged) and spin independent. Under this consideration the effective *exchange* coupling $J^{\sigma_e\sigma'}_{\rm eff}=\bar{J}_{\rm eff}$ also becomes spin independent and takes the form

$$\bar{J}_{\rm eff}(q,i\omega)=|J|^2P(q,i\omega)\ , \tag{8}$$

with the function

$$P(q,i\omega)=\sum_\lambda\frac{R_\lambda(q)}{i\omega-\omega_\lambda(q)}\ . \tag{9}$$

Notice that $P(q,i\omega)$ is independent of spins because of the absence of the Pauli principle between electrons and holes of different bands and $R_\lambda(q)$ plays the role of the residue of the pole at $i\omega=\omega_\lambda(q)$ and it is given by

$$R_\lambda(q)=\sum_k\phi^*_\lambda(k;q)\phi_\lambda(k;q)\ . \tag{10}$$

From $H_{\rm eff}(\mathbf{R}_{12})$ in (2) we can extract its *magnetic* part $H_{\rm mag}$, which has the Heisenberg form

$$H_{\rm mag}(\mathbf{R}_{12})=-\sum_{i\omega}\bar{J}_{\rm eff}(\mathbf{R}_{12},i\omega)\mathbf{S}_1(i\omega)\cdot\mathbf{S}_2(-i\omega)\ , \tag{11}$$

since $J^{\sigma_e\sigma'}_{\rm eff}=\bar{J}_{\rm eff}$ is independent of spin. The usual procedure in the evaluation of the interaction Hamiltonian $H_{\rm mag}$ is defined in (11) is to analyze the static limit $i\omega=0$ and thus obtain the interaction $\bar{J}_{\rm eff}(\mathbf{R}_{12},i\omega=0)$. In the approximation considered here this limit would mean the complete absence of temperature dependence in $\bar{J}_{\rm eff}(\mathbf{R}_{12},i\omega=0)$, which would be uninteresting from our point of view. Thus, we do not discuss this limit here. We would rather rewrite (11) in terms of imaginary times τ_1 and τ_2 producing

$$H_{\rm mag}=-\int_0^\beta d\tau_1d\tau_2\bar{J}_{\rm eff}(\mathbf{R}_{12},\tau_2-\tau_1)\mathbf{S}_1(\tau_1)\cdot\mathbf{S}_2(\tau_2)\ , \tag{12}$$

where the dynamical effects due to the presence of the exciton bound states become evident. Here, we will be concerned only with the *instantaneous* part $H_{\rm mag}$, therefore we need only to analyze

$$\bar{J}_{\rm eff}(\mathbf{R}_{12},\tau=0)=\sum_q\bar{J}_{\rm eff}(q,\tau=0)\exp[-i\mathbf{q}\cdot(\mathbf{R}_2-\mathbf{R}_1)]\ , \tag{13}$$

where $\tau=\tau_2-\tau_1$ is the relevant imaginary *time* difference and

$$\bar{J}_{\rm eff}(q,\tau=0)=\sum_{i\omega}\bar{J}_{\rm eff}(q,i\omega)\ , \tag{14}$$

is the *instantaneous* effective exchange interaction, which by means of an analytic continuation to real times would also correspond to the spin-spin correlation function evaluated at equal times. The frequency summation in (14) can be performed by picking up the poles of $\bar{J}_{\rm eff}(q,i\omega)$ as they appear in (8). These poles are the same poles of $P(q,i\omega)$, which are located at the boson frequencies $i\omega=\omega_\lambda(q)$. Their contribution results in

$$\bar{J}_{\rm eff}(q,\tau=0)=\frac{|J|^2}{T}\sum_\lambda N_B[\omega_\lambda(q)]R_\lambda(q)\ , \tag{15}$$

with N_B being the Bose function. To obtain $\bar{J}_{\rm eff}(\mathbf{R}_{12},\tau=0)$ we need to perform a sum over q which plays the role of the center of mass momentum of the bound state. Here we would like to eliminate first the center-of-mass coordinates and be left only with the internal coordinates in order to ellucidate the possible effects that the *symmetry* of the internal wave functions of the bound state might have on the effective exchange coupling. To do so we use the integral representation of $R_\lambda(q)$ in terms of the internal bound state wave functions as expressed in (10). For convenience, we concern ourselves here only with the temperature range where $\min[\omega_\lambda(q)]\gg T$, in which case N_B can be approximated by the Boltzmann distribution, i.e., $N_B[\omega_\lambda(q)]\approx\exp[-\omega_\lambda(q)/T]$. The Boltzmann distribution is then used when the integration over momentum $\mathbf{q}$ is performed to obtain

$$\bar{J}_{\rm eff}(\mathbf{R}_{12},\tau=0)=\sum_\lambda J_\lambda(\mathbf{R}_{12})\ , \tag{16}$$

where

$$J_\lambda(\mathbf{R}_{12})=\frac{|J|^2}{T}\exp[-E_\lambda/T]M_\lambda(T) \tag{17}$$

is an effective coupling that depends on the internal quantum numbers $\lambda\equiv(n,l,m)$ of the bound state via

$$M_\lambda(T)=\int d\mathbf{r}_1d\mathbf{r}_2\xi_\lambda(\mathbf{r}_1)U(\mathbf{R}_{12},\mathbf{r}_{12};a_T)\xi^*_\lambda(\mathbf{r}_2)\ , \tag{18}$$

which is a dimensionless matrix element between internal

exciton wave functions. Here U is a temperature-dependent potential (with dimensions of inverse volume) which has the form

$$U(\mathbf{R}_{12},\mathbf{r}_{12};a_T)=\frac{2^{3/2}}{a_T^3}\exp[-(\gamma x_{12})^2/a_T^2]\times\exp[-(\gamma y_{12})^2/a_T^2]\times\exp[-(Z_{12}-\gamma z_{12})^2/a_T^2]\,, \tag{19}$$

with $a_T=\hbar\sqrt{2/MT}$ being the characteristic thermal length. In (19), without loss of generality, we have chosen the z axis of our system to be along the line that connects the two impurities, thus we defined $\mathbf{R}_{12}\equiv Z_{12}$, i.e., $X_{12}=Y_{12}=0$. It is very important to notice in (16) that the coupling J_λ changes *sign* depending on the *sign* of M_λ.

The matrix element $M_\lambda(T)$ can be rewritten in a form where the characteristic Bohr radius a_0 of the electron-hole bound state and the characteristic thermal length a_T appear explicitly. The ratio a_T/a_0 as we shall see below is crucial to determine the strength of the effective potential U. Hence, it is convenient to express the dimensionless matrix element $M_\lambda(T)$ as a function of $\alpha=a_T/a_0$. This is done by scaling the internal variables $\mathbf{r}_1$ and $\mathbf{r}_2$ by a_0, which produces

$$M_\lambda(T)=\int d\tilde{\mathbf{r}}_1 d\tilde{\mathbf{r}}_2\tilde{\xi}_\lambda(\tilde{\mathbf{r}}_1)\tilde{U}(\tilde{\mathbf{R}}_{12},\tilde{\mathbf{r}}_{12},\alpha)\tilde{\xi}^*_\lambda(\tilde{\mathbf{r}}_2)\,, \tag{20}$$

with the effective dimensionless potential being

$$\tilde{U}(\tilde{\mathbf{R}}_{12},\tilde{\mathbf{r}}_{12};\alpha)=\frac{2^{3/2}}{\alpha^3}\exp[-(\gamma\tilde{x}_{12})^2/\alpha^2]\times\exp[-(\gamma\tilde{y}_{12})^2/\alpha^2]\times\exp[-(\tilde{Z}_{12}-\gamma\tilde{z}_{12})^2/\alpha^2] \tag{21}$$

and the dimensionless wave functions being $\tilde{\xi}_\lambda(\tilde{\mathbf{r}})=a_0^{3/2}\xi_\lambda(\tilde{\mathbf{r}})$. The dimensionless effective potential is of crucial importance in establishing the strength of the matrix element M_λ and thus determining $\bar{J}_{\text{eff}}$. Let us analyze $\tilde{U}$ in some particular limits. The most interesting limit occurs when $\gamma\neq 0$.[11] When the thermal length a_T is much larger than the characteristic Bohr radius a_0 ($\alpha\gg 1$) the potential $\tilde{U}\ll 1$ and M_λ is small. As a consequence, when $\alpha\gg 1$ the magnetic coupling J_λ is quite small and thus uninteresting. Notice that when $\alpha\to\infty$, the potential $\tilde{U}\to 0$ and the coupling $J_\lambda\to 0$.

On the other hand, the most interesting case occurs when $\alpha\ll 1$, i.e., when the thermal length a_T is much smaller than the characteristic Bohr radius a_0. In this case the potential $\tilde{U}\gg 1$ and the coupling J_λ can be appreciable. The most interesting limit occurs when $\alpha\to 0$, in which case

$$\tilde{U}=(2\pi)^{3/2}\delta(\gamma\tilde{x}_{12})\delta(\gamma\tilde{y}_{12})\delta(\tilde{Z}_{12}-\gamma\tilde{z}_{12}) \tag{22}$$

and the matrix element

$$M_\lambda=\frac{(2\pi)^{3/2}}{|\gamma|^3}\int d\tilde{\mathbf{r}}_1\tilde{\xi}_\lambda(\tilde{x}_1,\tilde{y}_1,\tilde{z}_1)\tilde{\xi}^*_\lambda(\tilde{x}_1,\tilde{y}_1,\tilde{z}_1-\tilde{Z}_{12}/\gamma) \tag{23}$$

is now just a simple overlap integral between the internal bound-state wave functions separated by the distance $\tilde{Z}_{12}/\gamma$. This expression for M_λ valid when $\alpha\ll 1$ combined with expression (17) valid when $\min[\omega_\lambda(q)\gg T]$ provides a simple expression for the effective exchange coupling $\bar{J}_{\text{eff}}$ defined in (16). The final expression for $\bar{J}_{\text{eff}}$ is valid under two conditions. The first corresponds to the Boltzmann approximation, which is valid when $T_0\equiv\min[\omega_\lambda(q)]\gg T$ is satisfied, i.e., $T_0\gg T$, with $T_0=E_g-\mathcal{R}$. It is required that T_0 be positive, otherwise there is no bound state inside the gap. The second condition corresponds to $\alpha\ll 1$, which implies $T\gg T_u$, where $T_u=4(\mu/M)\mathcal{R}$. As a result, the approximation used so far is valid in the temperature range $T_u\ll T\ll T_0$.

We shall now turn our attention to some qualitative aspects of the magnetic coupling J_λ. The first important qualitative aspect is the determination of the *sign* of the magnetic coupling J_λ which is in turn governed by the *sign* of M_λ in the temperature range $T_u\ll T\ll T_0$. It is not difficult to convince ourselves that for the $1s$ bound state ($n=1,l=0,m=0$) M_{1s} is always positive, thus J_{1s} has always the *ferromagnetic* sign. In the case $n=2$, the $2s$ ($l=0$) bound state also produces $M_{2s}>0$ and $J_{2s}>0$. A similar situation also occurs for the $2p_x$ and $2p_y$ ($l=1$) bound states. In contrast, a more interesting situation occurs for the $2p_z$ state ($l=1$): for small separations $\tilde{Z}_{12}/\gamma<,4$, the overlap M_{2p_z} is positive and thus J_{2p_z} has the *ferromagnetic* sign for large separation $\tilde{Z}_{12}/\gamma\gg 4$, the overlap $M_{2p_z}<0$ and J_{2p_z} has the *antiferromagnetic* sign. Similar analysis can be also performed for the higher n states, from which the qualitative behavior of M_λ for different orbital l and azimuthal m quantum num-

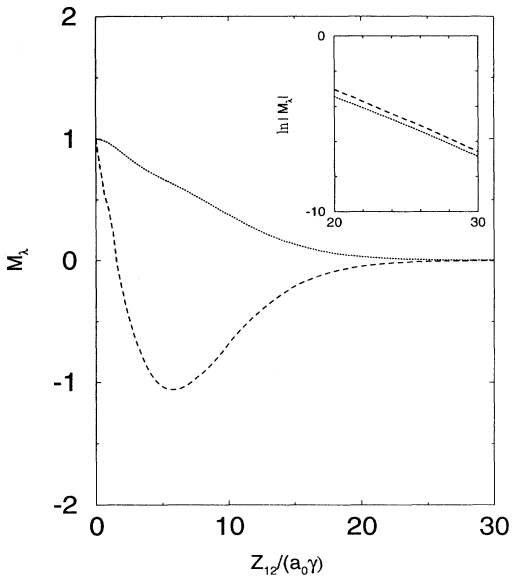

FIG. 1. Plot of M_{2s} (dotted curve) and M_{2p_z} (dashed curve) as a function of $\tilde{Z}_{12}=Z_{12}/(a_0\gamma)$, where Z_{12} is the separation between impurities. Notice the *sign* change in M_{2p_z} and recall that the indirect coupling J_λ is proportional to M_λ. In the inset, notice the exponential decay of M_λ for large $\tilde{Z}_{12}$.

bers can be easily extracted from the knowledge of the shapes of the bound-state wave functions. As an example, we plot M_λ for the $2s$ and $2p_z$ states in Fig. 1. Notice there the change in *sign* in M_{2p_z} and exponential decay at large distances of M_{2s} and M_{2p_z} shown in the inset.

The second important qualitative aspect of J_λ is that in the temperature range of interest ($T_u \ll T \ll T_0$) the matrix element M_λ is temperature independent and thus the magnetic exchange coupling has a simple activated form that can be read off from (17). The exact form of the activated behavior in (17) follows from the Boltzmann approximation used here, but physically it reflects the fact that when the temperature is increased more bound states (inside the gap) are populated and thus they can more effectively mediate the exchange interaction between the impurities. Conversely, when the temperature is lowered there are less bound states thermally populated and as a result the *magnitude* of the magnetic exchange J_λ is reduced.

Our results should be compared with the RKKY-like mechanisms (which neglect long-ranged Coulomb interactions) applied to metals[1] and semiconductors.[2] In the case of metals, where there is good screening, RKKY-like mechanisms have been quite successful in predicting the indirect exchange coupling between two magnetic impurities mediated by unbound particle-hole states, where the coupling oscillates (for large distances) between *ferromagnetic* and *antiferromagnetic* as a function of separation of the impurities.[1] In the case of semiconductors, where the screening is not so good, RKKY-like mechanisms (mediation via unbound electron-hole states) predict for a direct-gap semiconductor that the indirect exchange coupling (at large separations) decays exponentially with decay length $l_d=[2E_gM]^{-1/2}$ where the *sign* of the coupling is always *ferromagnetic*.[2] The inclusion of long-range attractive Coulomb interaction between conduction electrons and valence holes allows the formation of exciton bound states inside the semiconductor's gap, which in turn act as mediators of the indirect exchange interaction. As can be seen from Fig. 1 and Eq. (17), the decay of J_λ is still exponential at large distances, but its *sign* depends on the *symmetry* of the intermediate (correlated) bound state. This should be compared with the strong dependence of the superexchange interaction[3] on the *symmetry* of the intermediate (correlated) unbound states in the context of transition-metal oxides.[12]

Before concluding, we would like to mention briefly the case of a lattice of magnetic impurities. The qualitative aspects of the indirect coupling between two nearest-neighbor impurities does not change dramatically in comparison to the case of two magnetic impurities. This qualitative similarity occurs provided that the lattice impurity potential is not too large so that the bound states can still move around. This situation is still idealized but should be closer (quantitatively speaking) to the experimental situation of ferromagnetic-metal/semiconductor multilayers.[4–6] To compare our predictions qualitatively with experiments on ferromagnetic-metal/semiconductor multilayers, one must first establish the existence of stable excitons in the semiconductor spacer and secondly to identify the dominant *symmetry* of these stable intermediate states. If this is the case our results would then suggest that for semiconductor spacers with dominant $2s$ excitons the indirect exchange coupling is *ferromagnetic* for all spacer thicknesses, while for spacers with dominant $2p_z$ excitons the indirect exchange coupling is *ferromagnetic* at small thicknesses but *antiferromagnetic* at intermediate and larger thicknesses.

To conclude, we have proposed a new mechanism for indirect exchange coupling between two magnetic impurities embedded in a direct-gap semiconductor matrix and we briefly discussed the extension of the results for the impurity lattice case. Our mechanism is based on the existence of an attractive Coulomb potential between conduction electrons and valence holes. As a consequence of this Coulomb interaction bound states (excitons) exist inside the semiconductor's gap and serve as the intermediate states that transfer the spin information from one impurity to the other (or from one plane of impurities to the other). As a result of this theory the *magnitude* of the effective exchange coupling was strongly temperature dependent, always increasing with temperature, given that the effectiveness of the coupling was strongly dependent upon the thermal occupation of these bound states. The magnitude of the effect was mostly controlled by the binding energies of such states. In addition, the *sign* of the effective coupling was dependent on the *symmetry* of the internal wave functions of the bound states.

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[7]For useful tricks used to obtain effective actions see, e.g., V. N. Popov, *Functional Integrals and Collective Excitations* (Cambridge University Press, Cambridge, 1987).

[8]Rigorously speaking, a Coulomb exchange term between states where a spin-up electron is removed from the valence band and a spin-up electron is present at the conduction band (from which singlet excitons states are formed) must be added to the direct Coulomb interaction. This Coulomb exchange is absent in the case that leads to triplet excitons, and in the case that leads to singlet excitons it leads to a positive correction to the attractive Coulomb interaction between the electron-hole pair. This positive correction is just a consequence of Hund's rule where the total wave function of electron-hole pairs is more antisymmetric when the spins are aligned (triplet case) than when they are antialigned (singlet case), thus causing singlet exciton states to have higher energy than triplet exciton states. The size of this additional Coulomb exchange interaction is controlled by matrix elements appearing at the end of the interaction lines, which in turn are governed by the gap size. We assume here that this Coulomb exchange is negligible.

[9]See for instance, J. W. Negele and H. Orland, *Quantum Many-Particle Systems* (Addison-Wesley, Reading, MA, 1987), Chap. 2, 5, and 6.

[10]See, for instance, P. Nozières, *Theory of Interacting Fermi Systems* (Benjamin, New York, 1964).

[11]In the limit $\gamma=0$, $J_\lambda(\mathbf{R}_{12})$ either has always the *ferromagnetic* sign (for *s symmetry*) or vanishes identically (for other symmetries) as a function of $\mathbf{R}_{12}$.

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