Coherent optical control of spin-spin interaction in doped semiconductors

C. Piermarocchi and G. F. Quinteiro
Department of Physics and Astronomy, Michigan State University, East Lansing, Michigan 48824-2320, USA
(Received 10 August 2004; published 17 December 2004)

We provide a theory of laser-induced interaction between spins localized by impurity centers in a semiconductor host. By solving exactly the problem of two localized spins interacting with one itinerant exciton, an analytical expression for the induced spin-spin interaction is given as a function of the spin separation, laser energy, and intensity. We apply the theory to shallow neutral donors (Si) and deep rare-earth magnetic impurities (Yb) in III-V semiconductors. When the photon energy approaches a resonance related to excitons bound to the impurities, the coupling between the localized spins increases, and may change from ferromagnetic to antiferromagnetic. This light-controlled spin interaction provides a mechanism for the quantum control of spins in semiconductors for quantum information processing; it suggests the realization of spin systems whose magnetic properties can be controlled by changing the strength and the sign of the spin-spin interaction.

DOI: 10.1103/PhysRevB.70.235210 PACS number(s): 71.35.Gg, 03.67.Lx

I. INTRODUCTION

The possibility of creating semiconductor systems that can work simultaneously as electronic, photonic, and magnetic devices has boosted recently the research on light-spin interaction in semiconductors. These efforts extend both in the direction of an optical control of macroscopic magnetic properties and toward the quantum control of single spins.[1] Examples belonging to the first class include investigations on light-induced paramagnetic to ferromagnetic transitions in magnetic semiconductors, using coherent[2] or incoherent[3] processes, and attempts to control the macroscopic spin polarization of carriers using polarized light.[4] More recently, schemes for optical control at the level of a few spins have been proposed. Bao *et al.*[5] have demonstrated quantum spin entanglement of a few donors and magnetic impurities. The strong potentialities of this optical quantum control of spins for quantum information processing using quantum dots[6,7] and impurities[8] have been emphasized, and recent advances in the optical control at the nanoscale in semiconductor nanostructures[9,10] are particularly encouraging in this perspective.

It was pointed out in Ref. 6 that itinerant excitons, i.e., optical excitations free to move in the host material that embeds the localized spins, can induce an effective spin-spin interaction between localized spins. This mechanism has been dubbed the *optical Ruderman-Kittel-Kasuya-Yosida* (ORKKY) mechanism, in analogy to the mechanism in the theory of magnetism,[11] where electrons are involved. In the coherent optical case virtual excitons are created, and the ORKKY coupling is obtained from a second-order perturbation theory in the exchange coupling between the itinerant exciton and the localized spin. The ORKKY result predicts that the coupling between the localized spins is always ferromagnetic, independently of the sign of the coupling with the excitons. In this paper we show that higher-order terms in the exciton-impurity coupling can modify the strength and sign of the interaction, and affect its dependence as a function of the spin separation. The calculation of the spin-spin interaction can be reduced to a spin-dependent scattering problem that can be solved including exactly all the multiple-scattering terms between the two localized spins. We follow here an approach similar to that used to calculate multiple-scattering effects of $\pi^{\pm}$ mesons by deuterons.[12] The higher-order terms in the exciton-impurity coupling describe bound states which affect strongly the optically induced spin-spin interaction. In particular, a controlled antiferromagnetic (AF) coupling can be realized when the laser energy is tuned in the bonding-antibonding gap for the exciton localized by two impurities. This laser controlled switching of sign of the spin-spin interaction opens interesting directions in the investigation of competing interactions in spin systems.

The exchange between nuclear spins through excited electronic states has been discussed in the past in the case of molecules[13] and insulators.[14] In particular, Bloembergen and Rowland predicted in Ref. 14 an exponential decay of the spin-spin interaction with a characteristic length $\kappa = \hbar/\sqrt{2mE_g}$. This length depends on the energy gap E_g of the insulator and on the mass of the virtual electron-hole pairs across the gap. In the optically induced RKKY mechanism, the energy gap is effectively reduced by the laser field which increases the effective length to $\kappa = \hbar/\sqrt{2m(E_g - \hbar\omega_P)}$, with $\hbar\omega_P$ being the energy of the laser. Also, the density of electrons in the occupied bands in the insulator is replaced in the optical case by the density of photons in the field. The innovative strength of the optically induced case resides in the control potentialities since both the intensity and the frequency of the laser can be controlled in an experiment. Going beyond second-order perturbation theory presents intrinsic difficulties in the case of metals.[15] These difficulties are not present in the optical coherent case since there is no Fermi sea of electrons. The presence of a Fermi sea simultaneously with the laser would produce light-induced Kondo effects,[16] which we do not consider here.

The paper is organized as follows: In Sec. II we derive the expression for the effective Hamiltonian of two localized

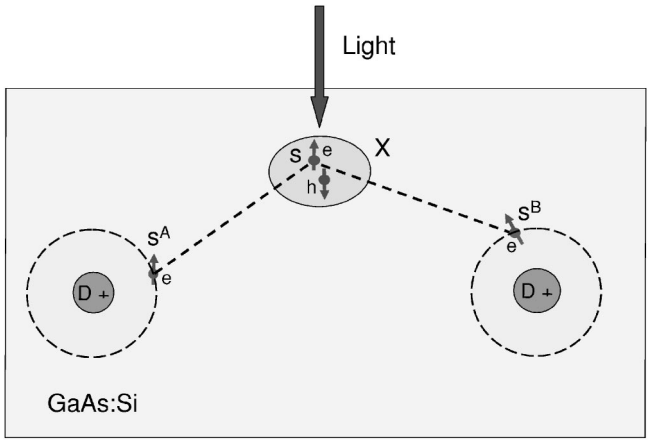

FIG. 1. Scheme of the light induced spin-spin interaction in the case of two shallow donors.

spins in the presence of a light field, relating it to the spin-dependent *T*-matrix operator of a two-center scattering problem. We study first in Sec. III the scattering of one exciton with one center. By generalizing a result from scattering theory[17] to the spin-dependent case, we show in Sec. IV how the *T*-matrix operator for the exciton scattering on two centers can be expressed in terms of the *T* operator for the one center scattering. We also study in this section the effects of the polarization of the light and we show that a circularly polarized field will induce an additional term representing a magnetic field. The theory is applied in Sec. V to two systems: shallow donors, and deep rare-earth magnetic impurities. We discuss implications for quantum computing implementations and for the optical control of macroscopic magnetic properties in Sec. VI.

II. EFFECTIVE HAMILTONIAN FOR LOCALIZED SPINS COUPLED BY THE LIGHT

A scheme showing the realization of a light induced spin coupling in the case of two shallow donors is given in Fig. 1. We are not interested in calculating the optical properties of the whole system, but we want to consider the effect of a coherent field on the dynamics of the two initially noninteracting localized spins s^A and s^B. The light creates virtual/real excitons in the semiconductor host and couples the localized spins. We want to study the behavior of the two localized spins in the coherent optical regime. This implies that the laser is always off resonance with respect to the free exciton band to avoid strong energy absorption. We therefore consider only single exciton processes in the presence of a monochromatic laser field. The system of two localized spins coupled to one itinerant exciton is described by the Hamiltonian

$$H_{XS} = H_0 + H_1, \tag{1}$$

where H_0 describes a free exciton of mass M with dispersion $\epsilon_k = \epsilon_0 + \hbar^2 k^2/2M$. The term H_1 describing the spin dynamics can be written in the form

$$H_1 = \frac{1}{V}\sum_{kk'\alpha\alpha'\beta} J_{k,k'}(s^A \cdot s_{\alpha'\alpha} + e^{-i(k'-k)R} s^B \cdot s_{\alpha'\alpha}) b^\dagger_{k'\alpha'\beta} b_{k\alpha\beta}, \tag{2}$$

where the two localized spins $\frac{1}{2}$ are described by s^A and s^B. V is the volume and s is the electronic spin of the itinerant exciton. $b^\dagger_{k\alpha\beta}$ creates an exciton with center-of-mass momentum k, electron spin α, and hole spin β. R is the separation between the two impurities. $J_{k,k'}$ is the exciton-spin exchange interaction. The strength and the sign of this term depend strongly on the nature of the localized spin. The sign, for instance, is determined by the competition between the ferromagnetic potential exchange and the antiferromagnetic kinetic exchange which is due to the hybridization of the itinerant exciton state with the localized state.[18] We will keep for the moment a general approach independent of the nature of the $J_{k,k'}$, and we will discuss two specific examples in Sec. V. A spin-independent term corresponding to a direct Coulomb interaction between the exciton and the impurity is also present. This term is small for shallow impurities, where kinetic exchange effects dominate, but becomes important for deep impurities. We will include this term in the case of rare-earth impurities discussed in Sec. V B, and we disregard it in the general discussion since it only introduces spin-independent energy shifts. We assume that the 1*s* excitons dominate the light induced effect, as discussed in Ref. 6. Moreover, we focus on systems where the localized states interact only with the electron in the exciton: the full Hamiltonian in Eq. (1) is diagonal in the hole spin index β. This is a good approximation for electrons in neutral donors, since it is equivalent to neglecting the electron-hole exchange interaction which in most semiconductors is much smaller than the electron-electron exchange. Concerning the second example we will consider, i.e., the case of the Yb^{3+} ions in III–V; it is known that these ions act as strong isoelectronic traps for electrons and the *s-f* exchange in the conduction band dominates.

The interaction of the excitons with an external time-dependent optical field provides the mechanism for the control of the two localized spins and is described by the Hamiltonian

$$H_{XL} = \sqrt{V}\sum_\sigma \Omega_\sigma e^{i\omega_L t}\phi_{1s} b_{k=0,\alpha+\beta=\sigma} + \text{H.c.}, \tag{3}$$

where Ω_σ is the Rabi energy of the interband optical transition and $\hbar\omega_L$ is the energy of the laser, σ is the polarization of the light. We have used the rotating wave approximation in Eq. (3). ϕ_{1s} is the envelope function of the electron hole pair taken at $\rho = r_e - r_h = 0$. In the case of a cw laser field, the time dependence can be eliminated by moving to the rotating frame with frequency ω_L, thus replacing ϵ_k by $\epsilon_k - \hbar\omega_L$ in H_0.

We are deriving an effective Hamiltonian for the two localized spins in the presence of the laser field. For a fixed value of the polarization σ, this Hamiltonian is four dimensional, corresponding to the degenerate ground state described by $|\lambda\rangle = \{|\uparrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$ for the two localized spins, and can be written to second order in H_{XL} as

$$H^{eff}_{\lambda\lambda'} = \langle\lambda|H_{XL}\frac{1}{\varepsilon^0_\lambda - PH_{XS}P}H_{XL}|\lambda'\rangle, \tag{4}$$

where P is the projector operator onto the subspace of one exciton plus two spins and ε^0_λ is the ground-state energy of two spins with no exciton. Operating H_{XL} onto $|\lambda\rangle$ generates states with one exciton at $k=0$ with polarization σ, states written as $\{|\lambda 0\sigma\rangle\}$. The ground-state energy can be chosen to be $\varepsilon^0_\lambda = 0$, and the expression above simplifies to

$$H^{eff}_{\lambda\lambda'} = V\sum_\sigma |\Omega_\sigma|^2|\phi_{1s}|^2 G_{\lambda 0\sigma,\lambda' 0\sigma}(\omega_L), \tag{5}$$

where

$$G_{\lambda 0\sigma,\lambda' 0\sigma}(\omega_L) = \left\langle \lambda 0\sigma \left| \frac{1}{[G^0(\omega_L)]^{-1} - H_1} \right| \lambda' 0\sigma \right\rangle \tag{6}$$

is the Green's-function operator for the system composed by the exciton and two spins, and

$$G^0(\omega_L) = \frac{\delta_{kk'}}{\hbar\omega_L - \left(\epsilon_0 + \dfrac{\hbar^2k^2}{2M}\right) + i\eta}. \tag{7}$$

We remark that we work in the off-resonance regime for which $\omega_L < \epsilon_0$, thus making the real part of G^0 always negative. Since we are dealing with only single-exciton processes, the Lippman-Schwinger equation for G can be rewritten in terms of an equation for the T matrix defined by the relation $G = G_0 + G_0 T G_0$. We solve the problem in two steps: (i) the T^A and T^B operators representing the scattering of the exciton with only one impurity (identified by the index A or B) is solved. (ii) The T matrix for the exciton interacting with two impurities is explicitly rewritten in terms of T^A and T^B using[17]

$$T = \frac{1}{1 - T^A G^0 T^B G^0} T^A[1 + G^0 T^B] + (A \rightleftharpoons B), \tag{8}$$

where $(A \rightleftharpoons B)$ stands for repeating the previous term with interchange of superscripts A and B. Equation (8) takes into account exactly all the multiple-scattering processes between the exciton and the two localized spins. We will focus in the next section on the interaction of the exciton with a single localized spin. Multiple-scattering effects in the two spins case are addressed in Sec. IV.

III. EXCITON-SINGLE IMPURITY SCATTERING

This section focuses on the solution of the T-matrix equation for one scattering center (named A). Due to the short-range nature of the exchange interaction, the exchange integral $J_{k,k'}$ in Eq. (2) is often reduced to a constant, corresponding to a deltalike interaction in space. Here we consider a more realistic form of the interaction using the separable potential approximation[19,20] where $J_{k,k'} = J v_k v_{k'}$, with v_k being a dimensionless form factor that depends only on $k = |\vec{k}|$. v_k describes the effect of the finite size of the nonlocal exchange interaction. The separable form of $J_{k,k'}$ will allow us to obtain analytical expressions for the T matrix, and provide a flexible theoretical framework with parameters that can be taken for the experiments. On the other hand, this potential can support at most one s-like bound state. The integral equation for the T matrix, $T = H_1 + H_1 G_0 T$, can then be written explicitly as

$$T^A_{kk'\alpha\alpha'} = \frac{J}{V} v_k v_{k'} s^A \cdot s_{\alpha\alpha'} + \frac{J}{V}\sum_{k''\alpha''} v_k v_{k''} s^A \cdot s_{\alpha\alpha''} G^0_{k''} T^A_{k''k'\alpha''\alpha'}. \tag{9}$$

We can write the T matrix as a sum of a scalar and vector part

$$T^A_{kk'\alpha\alpha'} = \frac{v_k v_{k'}}{V}[T_0 \delta_{\alpha\alpha'} + T_1 s^A \cdot s_{\alpha\alpha'}], \tag{10}$$

and, using the identity

$$(s^A \cdot s)^2 = \frac{3}{16} - \frac{s^A \cdot s}{2}, \tag{11}$$

we rewrite Eq. (9) in the form of two coupled equations,

$$T_1 = J + JF_0(T_0 - T_1/2), \tag{12a}$$

$$T_0 = \tfrac{3}{16} JF_0 T_1, \tag{12b}$$

where

$$F_0(\omega_L) = \frac{1}{V}\sum_{k''} v^2_{k''} G^0_{k''}. \tag{13}$$

The reduction of the integral equation to two algebraic equations is a consequence of the form of the interaction. The two coupled equations in Eq. (12) are solved and give

$$T_0 = \frac{3J}{16}\frac{JF_0}{1 + \dfrac{JF_0}{2} - \dfrac{3(JF_0)^2}{16}}, \tag{14a}$$

$$T_1 = \frac{J}{1 + \dfrac{JF_0}{2} - \dfrac{3(JF_0)^2}{16}}. \tag{14b}$$

This analytical solution allows us to investigate the strong-coupling regime in which the quantity JF_0 is not small. The most interesting feature of the strong-coupling regime is the formation of bound states of the exciton with the impurities, identified by the poles in the T matrix. Varying the frequency of the laser ω_L, which will modify the F_0, we can scan the spectrum to obtain the energies of those bound states. We remark that under the condition $\omega_L < \epsilon_0$, and assuming that the potential $v(k)$ is an analytic function of k, no singularities or branch cuts exist for the function F_0. Therefore the only source of poles is given by the zeros of the function $1 + JF_0/2 - 3(JF_0)^2/16$, appearing in T_1 and T_0. Considering separately the singlet and triplet channels we find

$$T^S = -\frac{3/4J}{1 + 3/4JF_0}, \tag{15a}$$

$$T^T = \frac{J/4}{1 - 1/4JF_0}. \tag{15b}$$

In the singlet and triplet channels only one of the two poles in Eq. (14) is present. Due to the fact that $F_0(\omega_L)$ is negative for all allowed values of ω_L, we also remark that, as expected, the exciton binds in a singlet spin state if $J>0$ (antiferromagnetic coupling), while the bound state is a triplet if the exciton-electron exchange is ferromagnetic (i.e., $J<0$).

IV. EXCITON-TWO IMPURITIES SCATTERING

Starting from the results obtained in the previous sections, we construct in this section the solution for the exciton-two impurities T matrix and the corresponding H_{eff} for the localized spins. Equation (8) can be expanded in terms of T operators as

$$T = T^A + T^A G^0 T^B + T^A G^0 T^B G^0 T^A + \cdots. \tag{16}$$

The matrix for T^B can be obtained from a simple phase shift: if $T^A_{kk'}$ is the T matrix for a scattering center with potential $V(r)$ then $e^{-i(k'-k)\cdot R}T^A_{kk'}$ is the corresponding one for a potential $V(r-R)$, i.e., $T^B_{kk'}$.[17] We can take the matrix elements of Eq. (16) in the k representation. To illustrate how this series can be summed let us consider as an example the third term in Eq. (16),

$$\langle k|T^A G^0 T^B G^0 T^A|k'\rangle = \frac{1}{V^3}\sum_{k'',k'''} v_k v_{k''} \Upsilon^A G^0_{k''} e^{-i(k'''-k'')\cdot R} v_{k''} v_{k'''} \times \Upsilon^B G^0_{k'''} v_{k'''} v_{k'} \Upsilon^A, \tag{17}$$

where we have defined

$$\Upsilon^{A(B)} = T_0 + T_1 s^{A(B)} \cdot s. \tag{18}$$

Reordering factors and defining the function

$$F_R(\omega_L) = \frac{1}{V}\sum_k e^{ik\cdot R} v_k^2 G^0_k, \tag{19}$$

this term takes the form

$$\frac{v_k v_{k'}}{V} F_R^2(\omega_L) \Upsilon^A \Upsilon^B \Upsilon^A. \tag{20}$$

Following the same procedure, the full series can be summed and we obtain for $k=k'=0$

$$T_{0,0} = \frac{v_0 v_0/V}{1 - F_R^2 \Upsilon^A \Upsilon^B} \Upsilon^A [1 + F_R \Upsilon^B] + (A \rightleftharpoons B). \tag{21}$$

The T matrix is now expressed as an operator in an eight-dimensional space generated by three spins $1/2$: one electron in the exciton and two localized electron states. By direct inversion and products of 8×8 matrices, Eq. (21) can be rewritten in terms of a combination of spin products (see the Appendix), and using $G=G_0+G_0TG_0$ we obtain the spin-dependent effective Hamiltonian

$$H_{eff} = B_{eff} \cdot (s^A + s^B) + J_{eff} s^A \cdot s^B. \tag{22}$$

B_{eff} represents an effective magnetic field acting on both spins and J_{eff} is an effective isotropic Heisenberg exchange. The effective magnetic field and exchange constant can be written as

$$B_{eff} = \frac{|\Omega_{\sigma+}|^2 - |\Omega_{\sigma-}|^2}{\delta^2} \frac{|\phi_{1s}|^2 v_0^2 J(1 - JF_R^-)}{(1 - JF_R^+)[1 - JF_R^+(3JF_R^- - 2)]} \frac{\hat{z}}{2} \tag{23}$$

and

$$J_{eff} = \frac{|\Omega_{\sigma+}|^2 + |\Omega_{\sigma-}|^2}{\delta^2} \times \frac{|\phi_{1s}|^2 v_0^2 J^2 F_R/2(1 - JF_R^-)}{(1 - JF_R^+)[1 - JF_R^+(3JF_R^- - 2)][1 - JF_R^-(3JF_R^+ - 2)]}, \tag{24}$$

where we have defined

$$F_R^{\pm}(\omega_L) = \frac{1}{4V}\sum_k (1 \pm e^{ik\cdot R}) v_k^2 G^0_k. \tag{25}$$

$\hat{z}$ identifies the direction of propagation of the light, $\delta=\epsilon_0-\hbar\omega_L$ is the optical detuning, and $\Omega_{\sigma\pm}$ correspond to the contributions to the Rabi energy from the two circularly polarized components of the light. From Eq. (21) a spin-independent term is also derived which is not shown in Eq. (22) since it is irrelevant for our purposes. If we want to include the effect of the degenerate light hole band the two expressions in Eqs. (23) and (24) should be multiplied by $2/3$ and $4/3$, respectively. By keeping the lowest order in J in Eqs. (23) and (24) we obtain

$$B_{eff} = \frac{|\Omega_{\sigma+}|^2 - |\Omega_{\sigma-}|^2}{\delta^2} |\phi_{1s}|^2 v_0^2 J \frac{\hat{z}}{2} + O(J^2), \tag{26}$$

and

$$J_{eff} = \frac{|\Omega_{\sigma+}|^2 + |\Omega_{\sigma-}|^2}{\delta^2} |\phi_{1s}|^2 v_0^2 J^2 F_R/2 + O(J^3) \tag{27}$$

which recovers the optical RKKY result of Ref. 6. The magnetic field induced by virtual excitons has recently been analyzed using a more fundamental approach in the case of a single impurity by Combescot and Betbeder-Matibet in Ref. 21. In this reference the spin-independent term that provides a correction to the optical Stark shift is also discussed.

V. SPIN-SPIN COUPLING

In this section we apply the results obtained above to (i) excitons mediating the interaction between two electronic spins localized in shallow donors (e.g., GaAs:Si) and (ii) excitons mediating the interaction between two magnetic ions with spin $1/2$ (two rare-earth ion Yb^{3+} in InP). Yb in InP is one of the most investigated rare-earth-doped III-V materials. In principle Yb^{3+} in GaAs could be used but it is technically more challenging to obtain samples where only

substitutional Yb is present.[22] We will focus on the effect of the binding of excitons on the spin-spin coupling. The parameters J and the range of the potential v_k can be fixed in such a way that the single-spin exciton T matrix reproduces the binding energy and the spin configuration of the bound exciton obtained from the experiment.

A. Shallow donors

For a scheme of the system we can refer again to Fig. 1. For excitons interacting with a shallow neutral donor the effective mass approximation can be used. The problem of excitons bound to neutral donors and acceptors has been heavily investigated both experimentally and theoretically.[23] In the case of GaAs it is known that the exciton binds to a neutral donor with a binding energy of about 1 meV. It is also clear from the magnetic-field dependence of the bound exciton resonance that the two electrons are paired in a singlet around the donor ion and the hole is bound by Coulomb interaction. The picture is very similar to the one of a positive charge bound to an H^- ion. As in the H^- ion case, the dominant term responsible for the binding of the two electrons is a kinetic exchange term and we can therefore disregard the effect of a spin-independent term in the Hamiltonian. The range of the kinetic exchange is determined by the hybridization between the localized electron state in the neutral donor and the electron state in the free exciton. We therefore assume that the v_k is of the form

$$v_k^2 = \frac{1}{1+(\Lambda k)^2}, \tag{28}$$

where the parameter Λ determines the range of the potential. In the following we will use the excitonic atomic units where energy is given in excitonic Ry^* and lengths are in Bohr radius a_B^*. $Ry^* = e^4\mu/\varepsilon_0^2 2\hbar^2$ where μ is the reduced mass of the electron hole system (in electronic mass units) and ε_0 is the static dielectric constant in the semiconductor. $a_B^* = \varepsilon_0\hbar^2/e^2\mu$, and the relation $Ry^* = \hbar^2/2\mu a_B^{*\,2}$ holds. For GaAs these units give $1Ry^* \sim 5$ meV and $a_B^* \sim 100$ Å. Using these units we can calculate the functions F_0 and F_R in Eqs. (13) and (19) as

$$F_0 = -\frac{1}{4\pi\Lambda}\frac{1}{(\Lambda\sqrt{\delta\nu}+\nu)}, \tag{29}$$

$$F_R = -\frac{1}{4\pi R}\frac{e^{-R/\Lambda}-e^{-R\sqrt{\delta/\nu}}}{\Lambda^2\delta-\nu}, \tag{30}$$

where $\nu=\mu/M$ is the reduced total mass ratio of the excitonic system which is about $1/5$ in GaAs taking $m_e=0.08$ and $m_h=0.17$. Notice that F_R can be rewritten as

$$F_R = F_0\Lambda\nu\frac{e^{-R/\Lambda}-e^{-R\sqrt{\delta/\nu}}}{R(\Lambda\sqrt{\delta\nu}-\nu)} \tag{31}$$

and has no poles for positive detuning δ; for $R \gg \lambda$, F_R has a Yukawa form with a detuning-related decay length $a_B^*\sqrt{\nu/\delta}$ as found in Ref. 6, while at short R the finite range of the potential regularizes the $1/R$ divergence. In v_k we take

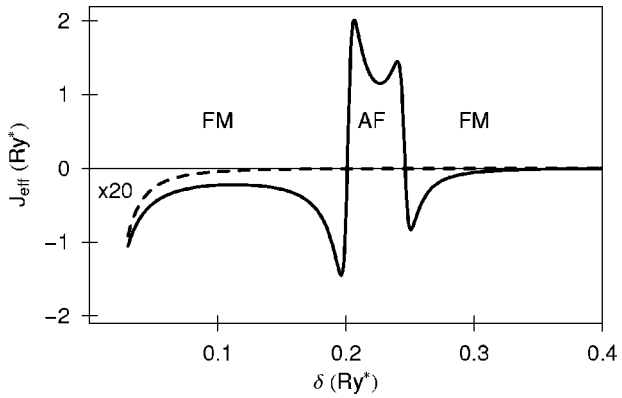

FIG. 2. Coupling constant J_{eff} between the two electronic spins localized in a shallow neutral donor embedded in GaAs as a function of the laser energy, measured from the bottom of the free exciton band. The intensity of the laser corresponds to a Rabi energy of $\Omega_{\sigma+}=0.05$ meV. The dashed line gives the result predicted with the same parameters using second-order perturbation theory in the coupling constant J.

$\Lambda=0.25$. Using the fact that the exciton binds to the donor only in the singlet channel, we can determine the value of J in the T^S in Eq. (15a) in such a way to have a pole at the experimental binding energy. The J is positive, as expected from the fact that the kinetic exchange is antiferromagnetic, and we take its value to be $J=1Ry^*(a_B^*)^3$ which gives a binding energy for the singlet of $0.23Ry^*$, in accordance with the experimental value of 1 meV. The triplet is unbound.

We plot in Fig. 2 the coupling constant J_{eff} obtained from Eq. (24) as a function of the energy of the laser measured from the bottom of the free exciton band, $\delta=\epsilon_0-\hbar\omega_L$. A small imaginary contribution to the energy, $\eta=0.0001Ry^*$, has been added in all the plots. The Rabi energy is $\Omega_{\sigma+}=0.05$ meV. The $\sigma-$ component of the Rabi energy is zero. The separation between the two neutral donors R is $2a_B^*$. In the region of large detuning we have a ferromagnetic coupling in agreement with the results obtained in the ORKKY limit. When we approach the energy corresponding to the binding of the exciton to the impurity, at $\delta=0.23$, we observe that the interaction is strongly enhanced and there is a region with an antiferromagnetic (AF) coupling. Multiple scattering between the two impurities results in the formation of bonding-antibonding states for the exciton. When the light has a frequency in the bonding-antibonding gap the effective interaction changes sign. This is analogous to the antiferromagnetic coupling generated by superexchange in magnetic materials.[18] When the laser is tuned above the resonances we recover again the ferromagnetic coupling. In the same plot we also show the effective coupling that would result by keeping the lowest order in J (ORKKY). In this case no resonances due to the binding of the excitons are present and, in order to obtain a sizeable coupling, the laser has to be tuned close the bottom of the excitonic band. In Fig. 3 we show a contour plot of the effective spin-spin coupling as a function of the detuning and impurity separation. Red corresponds to strong negative and violet to strong positive coupling, the green tone in the upper-right corner corresponds to zero. At large R the coupling is mostly ferromagnetic and

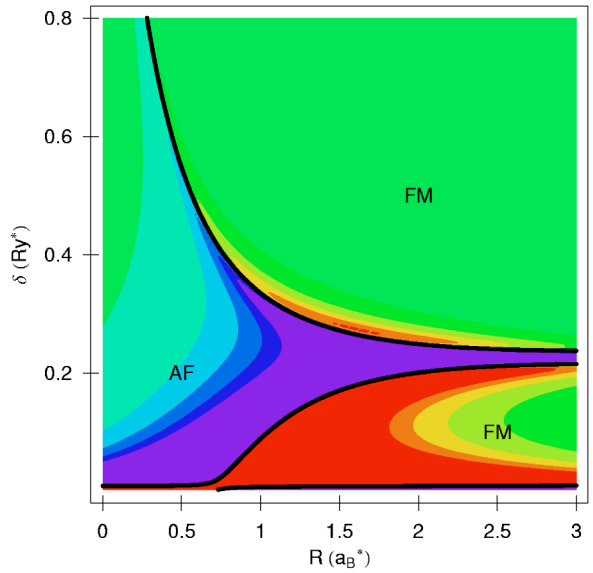

FIG. 3. (Color online). J_{eff} as a function of the donors separation R and detuning δ. The contour plot identifies the regions where the coupling is ferromagnetic (FM) or antiferromagnetic (AF). The thick lines indicate a change of sign of J_{eff}. The intensity of the laser corresponds to a Rabi energy of $\Omega=0.1$ meV. In the color figure, green corresponds to $J_{eff}=0$. Colors from green to red correspond to negative values (FM). Colors from green to violet correspond to positive values (AF).

there is only a small region close to the exciton binding energy where the coupling can be AF (violet region in the plot). When the distance between the two impurities decreases, the bonding-antibonding gap and the region corresponding to the antiferromagnetic coupling is wider. The thick line indicates a change of sign of J_{eff}. Notice also the different decay of the interaction as a function of R for different values of the detuning. At $\delta=0.4$ the maximum strength is at $R=0.8$ and decays quickly within a quarter of a_B^* to the minimum value in the plot. At $\delta=0.1$ the same minimum is reached within a much larger interval of about $2a_B^*$. This is consistent with the fact that at a small detuning there is a contribution from the free exciton band which can give a longer range for the effective interaction.

B. Rare-earth impurities

The magnetic properties of the Yb^{3+} ion in III-V (Ref. 24) arise from its partially filled $4f$ shell, possessing 13 electrons. In III-V materials, for a substitutional impurity, the crystal fields split the ground manifold of the ion into two doublets $\left(\text{spin}=\frac{1}{2}\right)$, Γ_6 and Γ_7, and a quadruplet $\left(\text{spin}=\frac{3}{2}\right)$, Γ_8. The lowest-lying state is the Kramers doublet Γ_6, which behaves like a spin $\frac{1}{2}$ with an effective isotropic $g=24/7$.[25] Yb in InP replaces indium and acts as an isoelectronic trap. From electrical[26] and optical[27] measurements it is known that the exciton binds to this isoelectronic impurity with a binding energy of 30 meV. The binding is due to a short-range potential that arises from the difference in the core pseudopotential between the impurity and the host ion it replaces.[28] It is reasonable to assume that this short-range potential is spin-independent and we take it into account by adding to the exciton-impurity Hamiltonian of Eq. (1) the term

$$H_2 = \frac{1}{V}\sum_{k,k'} \Delta_{k,k'}(1 + e^{-i(k'-k)R}) b^\dagger_{k'\alpha\beta} b_{k\alpha\beta}. \tag{32}$$

We use the separable potential approximation also for the spin-independent short-range potential and we parametrize it in the form $\Delta_{k,k'}=\Delta v_k v_{k'}$, i.e., it has the same k dependence of $J_{k,k'}$. A more general analytical result can be obtained using a separable form for $\Delta_{k,k'}$ with different coefficients, but we expect the range of the s-f exchange and that of the impurity potential to be very similar. The value of Δ is determined by imposing that the exciton-single impurity T matrix has a pole for both singlet and triplet channels at 30 meV. Following the same procedure used in Sec. III we obtain for the T operators in the singlet and triplet channels

$$T^S = \frac{-3/4J + \Delta}{1 + 3/4JF_0 - \Delta F_0}, \tag{33a}$$

$$T^T = \frac{J/4 + \Delta}{1 - 1/4JF_0 - \Delta F_0}. \tag{33b}$$

The expressions for the J_{eff} and B_{eff} modified by the presence of Δ can be obtained by plugging the Eqs. (33) in the general expressions of Eqs. (A3) in the Appendix. The quantity J is the s-f exchange interaction between the impurity and the electron in the exciton. In typical rare-earth ferromagnetic semiconductors the s-f exchange is ferromagnetic and is of the order of few eV $Å^3$,[29] comparable to the s-d exchange in Mn based diluted magnetic semiconductors.[30] We are using $J=-10^{-4}$ in our units which corresponds to a conservative estimate of 0.7 eV $Å^3$ in InP. In InP the value of the Ry^* is about the same as that of GaAs (5 meV), while the Bohr radius is about 120 Å. For Λ in v_k we take $\Lambda=0.01$ which is of the order of the ionic radius of Yb^{3+}.

We show in Fig. 4 the contour plot of J_{eff} as a function of the laser detuning $\delta=\epsilon_0-\hbar\omega_L$ and of the separation between the impurities R. At large distances we observe two resonances related to the binding of the exciton in the singlet and triplet channels. Figure 5(a) shows in detail the J_{eff} for a distance $R=1a_B^*$. The two peaks in Fig. 5(a) correspond to the exciton bound to the impurity in the triplet and singlet channel. The peak at larger detuning corresponds to the triplet since the s-f exchange is ferromagnetic. For shorter distances we see from Fig. 4 that each of the two peaks starts to split. The singlet (at smaller detuning) follows a behavior similar to the one of the shallow donors described above: the bonding and antibondig states identify a region where the coupling becomes antiferromagnetic. The triplet state splits

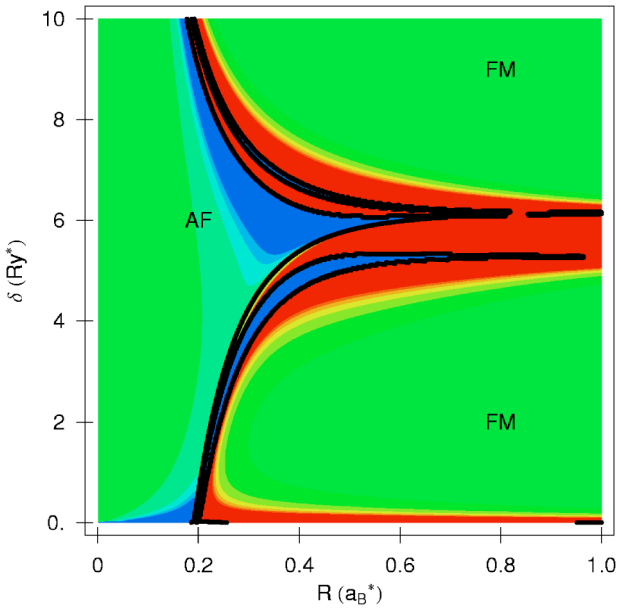

FIG. 4. (Color online). Coupling constant J_{eff} between two magnetic Yb^{3+} localized in InP as a function of the laser detuning $\delta = \epsilon - \hbar\omega_L$ and separation between the ions. The thick lines indicate $J_{eff}=0$ and a change form ferromagnetic to antiferromagnetic coupling. The intensity of the laser corresponds to a Rabi energy of $\Omega=0.1$ meV. In the color figure, green corresponds to $J_{eff}=0$. Colors from green to red correspond to negative values (FM). Colors from green to violet correspond to positive values (AF).

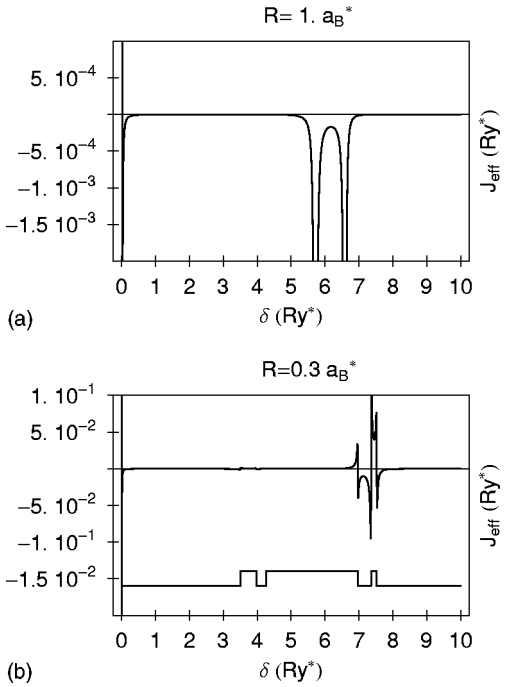

FIG. 5. Coupling constant J_{eff} between two Yb^{3+} ions in InP as a function of the detuning δ. (a) Large distance. The coupling is ferromagnetic and the resonances in the interaction are close to the energy of the exciton bound to the Yb. (b) Short distance. The triplet channel splits in many different peaks producing many changes of the spin-spin coupling sign. The lower curve shows the sign of the coupling constant.

in many different peaks as can be seen from Fig. 5(b). The sign of the interaction can change many times as a function of the detuning in this short distance region. This is indicated by the sign of J_{eff} plotted in the lower part of Fig. 5(b). Overall the antiferromagnetic coupling dominates at short distances while the interaction is ferromagnetic at large R.

VI. DISCUSSION

The spin-spin interaction control discussed in this paper has potential applications in quantum computing implementations. In fact, an optical control of the spin of electrons localized in quantum dots or impurities has several advantages with respect to approaches where electrodes are needed. Ultrafast lasers are available, promising the realization of quantum gates in time scales that are hard to achieve with an electrical control. Lasers are also very flexible for quantum control since pulse shaping can be used to increase accuracy and speed.[31] Finally, metallic electrodes necessarily add a source of noise for the quantum system, and they are not needed in an optical scheme. The possibility of changing the sign of the spin-spin interaction can add flexibility to many control schemes for the qubits, like, e.g., in the exchange-only scheme.[32] We have seen that resonances in the spin-spin coupling induced by the binding of the excitons can increase the magnitude of the interaction for distances that are reasonable from a nanofabrication point of view. This will imply that lasers with lower intensities can be employed in the control. The polarization of the light represents an additional control parameter that can be used to selectively address qubits with an optically induced magnetic field. This is also an advantage from a practical point of view since it could simplify the experimental setup by eliminating the need of an external magnetic field.

Although the feasibility of single impurity spectroscopy in semiconductors has been proven,[33,34] little attention has been paid to optical properties of impurity-bound excitons for information storage and processing. Impurities deserve at least the same attention as quantum dots for such applications. Their homogeneous character and the variety of properties that one can obtain combining different hosts and ions are indeed special advantages. An exciton bound to an impurity has optical properties very similar to an exciton trapped in a shallow quantum dot. Most of the ideas involving excitons in quantum dots as a main ingredient for quantum information and communication can be reformulated for excitons bound to impurities. We have provided only two examples here, but our phenomenological theory, being based on inputs from the experiments, is very flexible and many other combinations of host and ions can be used to explore a large range of confinement energy and different optical properties. We also have seen that the spin-spin coupling has a resonant behavior at frequencies depending on the separation between the impurities. By organizing the impurities in chains with different separation this can be used to

selectively address a single pair of impurities and it allows for scalability.

A very special case is represented by impurities in silicon. This material has obvious technological advantages and many proposals for using impurities in Si for quantum computing have been suggested.[35–37] In particular, the optical control of electronic spins localized by deep donors in Si using a *control* impurity has been proposed.[8] In the scheme we are suggesting here, the exciton bound to the impurity plays the role of the *control* impurity and it takes advantage of the host material for mediating the interaction. Even if Si is an indirect gap material, there is a finite optical coupling to the exciton bound to the impurity due to symmetry breaking. Additional complications in the use of excitons bound to donors for mediating spin-spin coupling arise from the valley degeneracy in Si.[38] We will address the optical spin control of impurities in Si and the role of valley degeneracy in a future publication.

Excitons bound to rare-earth magnetic ions can be controlled very rapidly and efficiently due to their strong dipole moment. Their dipole moment is mainly determined by the optical properties of the host material, since it involves the creation of electron-hole pairs across the semiconductor gap. At the same time, they interact with the internal degrees of freedom in the core *f* states. Schemes involving excitons bound to rare-earth impurities in III-V materials bring in the advantages of the optical properties of the host and the stability of the internal degrees of freedom of the *f* orbitals in the rare-earth ion where the qubit is stored. This hybrid system is thus extremely powerful, providing both reliable storage and fast processing of information.

Finally, the light controlled spin-spin coupling in a semiconductor matrix is also appealing for the coherent control of macroscopic properties of materials. This was the idea behind the coherently induced ferromagnetism in Ref. 2. There, a finite critical temperature for a paramagnetic to ferromagnetic transition in diluted magnetic semiconductors was found when the material is coupled to a strong laser field. The results presented in this paper suggest that the presence of bound states could enhance the effect. Also, the same idea could be used in other systems where the light can induce antiferromagnetic or glassy phases starting from a paramagnetic system. This represents a unique opportunity to study phase transitions in a solid where the coupling is controlled by an external field and may lead to a different class of *controlled* materials to be investigated.

In conclusion, we have studied the problem of two spins $\frac{1}{2}$ localized by impurities in semiconductor in the presence of an intense light field. The light induces a frequency-dependent spin-spin coupling and a magnetic field that can be controlled by the polarization of the light. The effects are enhanced by the presence of impurity bound excitons which may split into bonding and antibonding states in the case of two impurities. The sign of the spin-spin coupling is generally ferromagnetic, but it can switch to antiferromagnetic when the laser is tuned to the bonding-antibondig gap. We have developed a flexible theoretical approach based on scattering theory where the parameters from the experiment can be used to estimate the size of the effect. We have discussed explicitly the case of two neutral donors in GaAs and two rare-earth magnetic ions (Yb^{3+}) in InP.

ACKNOWLEDGMENTS

This work was supported by NSF Grant No. DMR-0312491. We thank T. A. Kaplan for enlightening discussions on the theory of magnetism, and S. D. Mahanti for suggesting useful references on RKKY.

APPENDIX: MATRIX REPRESENTATION OF THE *T* OPERATOR

Using the basis set $|s_z^A, s_z^B, s_z\rangle$ we obtain for the spin products $s^A \cdot s$, $s^B \cdot s$, $s^A \cdot s^B$ the matrices

$$s^A \cdot s = \begin{bmatrix} \frac{1}{4} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -\frac{1}{4} & 0 & 0 & \frac{1}{2} & 0 & 0 & 0 \\ 0 & 0 & \frac{1}{4} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -\frac{1}{4} & 0 & 0 & \frac{1}{2} & 0 \\ 0 & \frac{1}{2} & 0 & 0 & -\frac{1}{4} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{1}{4} & 0 & 0 \\ 0 & 0 & 0 & \frac{1}{2} & 0 & 0 & -\frac{1}{4} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{4} \end{bmatrix} \tag{A1a}$$

$$s^B \cdot s = \begin{bmatrix} \frac{1}{4} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -\frac{1}{4} & \frac{1}{2} & 0 & 0 & 0 & 0 & 0 \\ 0 & \frac{1}{2} & -\frac{1}{4} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{1}{4} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{1}{4} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -\frac{1}{4} & \frac{1}{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{1}{2} & -\frac{1}{4} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{4} \end{bmatrix} \tag{A1b}$$

$$s^A \cdot s^B = \begin{bmatrix} \frac{1}{4} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \frac{1}{4} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -\frac{1}{4} & 0 & \frac{1}{2} & 0 & 0 & 0 \\ 0 & 0 & 0 & -\frac{1}{4} & 0 & \frac{1}{2} & 0 & 0 \\ 0 & 0 & \frac{1}{2} & 0 & -\frac{1}{4} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{1}{2} & 0 & -\frac{1}{4} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{4} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{4} \end{bmatrix}. \tag{A1c}$$

By substituting these expressions in Υ^A and Υ^B and then in Eq. (21), we obtain after matrix inversions and multiplications an expression for $T=(1-F_R^2\Upsilon^A\Upsilon^B)^{-1}\Upsilon^A[1+F_R\Upsilon^B] + (A \rightleftharpoons B)$. The traceless part of this matrix is

$$\begin{bmatrix} \frac{\beta+2\alpha}{4} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \frac{\beta-2\alpha}{4} & \frac{\alpha}{2} & 0 & \frac{\alpha}{2} & 0 & 0 & 0 \\ 0 & \frac{\alpha}{2} & -\frac{\beta}{4} & 0 & \frac{\beta}{2} & 0 & 0 & 0 \\ 0 & 0 & 0 & -\frac{\beta}{4} & 0 & \frac{\beta}{2} & \frac{\alpha}{2} & 0 \\ 0 & \frac{\alpha}{2} & \frac{\beta}{2} & 0 & -\frac{\beta}{4} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{\beta}{2} & 0 & -\frac{\beta}{4} & \frac{\alpha}{2} & 0 \\ 0 & 0 & 0 & \frac{\alpha}{2} & 0 & \frac{\alpha}{2} & \frac{\beta-2\alpha}{4} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{\beta+2\alpha}{4} \end{bmatrix} \tag{A2}$$

where α and β can be conveniently expressed as a function of the single impurity T^T and T^S operators in Eqs. (15) or Eqs. (33) as

$$\alpha = \frac{2(T^T - T^S)(T^T F_R + 1)}{(T^T F_R - 1)[F_R(T^S - T^T + 2F_R T^T T^S) - 2]}, \tag{A3a}$$

$$\beta = -\alpha \frac{(T^T - T^S)F_R}{(F_R(T^T - T^S + 2F_R T^T T^S) - 2)}, \tag{A3b}$$

where we have dropped the $\{A,B\}$ index since we are considering two identical centers. Notice that the matrix in Eq. (A2) can be rewritten as

$$\alpha(s^A + s^B) \cdot s + \beta s^A \cdot s^B. \tag{A4}$$

[1] *Semiconductor Spintronics and Quantum Computation*, edited by D. D. Awschalom, D. Loss, and N. Samarth (Springer, New York, 2002).

[2] J. Fernández-Rossier, C. Piermarocchi, P. Chen, A. H. MacDonald, and L. J. Sham, Phys. Rev. Lett. **93**, 127201 (2004).

[3] H. Boukari, P. Kossacki, M. Bertolini, D. Ferrand, J. Cibert, S. Tatarenko, A. Wasiela, J. A. Gaj, and T. Dietl, Phys. Rev. Lett. **88**, 207204 (2002).

[4] H. Ohno, Science **281**, 951 (1998).

[5] J. Bao, A. V. Bragas, J. K. Furdyna, and R. Merlin, Nat. Mater. **2**, 175 (2003); Solid State Commun. **127**, 771 (2003); cond-mat/0406672, Phys. Rev. B (to be published).

[6] C. Piermarocchi, P. Chen, L. J. Sham, and D. G. Steel, Phys. Rev. Lett. **89**, 167402 (2002).

[7] E. Pazy, E. Biolatti, T. Calarco, I. D'Amico, P. Zanardi, F. Rossi, and P. Zoller, Europhys. Lett. **62**, 175 (2003).

[8] A. M. Stoneham, A. J. Fisher, and P. T. Greenland, J. Phys.: Condens. Matter **15**, L447 (2003).

[9] X. Li, Y. Wu, D. G. Steel, D. Gammon, T. H. Stievater, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Science **301**, 809 (2003).

[10] P. Bianucci, A. Muller, C. K. Shih, Q. Q. Wang, Q. K. Xue, and C. Piermarocchi, Phys. Rev. B **69**, 161303 (2004).

[11] M. A. Ruderman and C. Kittel, Phys. Rev. **96**, 99 (1954); T. Kasuya, Prog. Theor. Phys. **16**, 45 (1956); K. Yoshida, Phys. Rev. **106**, 893 (1957).

[12] S. D. Drell and L. Verlet, Phys. Rev. **99**, 849 (1955).

[13] N. F. Ramsey, Phys. Rev. **91**, 303 (1953).

[14] N. Bloembergen and T. J. Rowland, Phys. Rev. **97**, 1679 (1955).

[15] G. Vertogen and W. J. Caspers, Z. Phys. **198**, 37 (1967).

[16] T. V. Shahbazyan, I. E. Perakis, and M. E. Raikh, Phys. Rev. Lett. **84**, 5896 (2000).

[17] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford University Press, London, 1965).

[18] P. W. Anderson, Solid State Phys. **14**, 99 (1963).

[19] Y. Yamaguchi, Phys. Rev. **95**, 1628 (1954).

[20] L. H. Schick, Rev. Mod. Phys. **33**, 608 (1961).

[21] M. Combescot and O. Betbeder-Matibet, Solid State Commun. **132**, 129 (2004).

[22] A. Taguchi, H. Nakagome, and K. Takahei, J. Appl. Phys. **68**, 3390 (1990).

[23] P. J. Dean and D. C. Herbert, *Bound Excitons in Semiconductors*, in *Excitons*, edited by K. Cho (Springer-Verlag, Berlin, 1979).

[24] V. F. Masterov and L. F. Zakharenkov, Fiz. Tekh. Poluprovodn. (S.-Peterburg) **24**, 610 (1990) [Sov. Phys. Semicond. **24**, 383 (1990)].

[25] G. Aszodi, J. Weber, Ch. Uihlein, L. Pu-lin, H. Ennen, U. Kaufmann, J. Schneider, and J. Windscheif, Phys. Rev. B **31**, 7767 (1985).
[26] P. S. Whitney, K. Uwai, H. Nakagome, and K. Takahei, Appl. Phys. Lett. **53**, 2074 (1988).
[27] B. Lambert, A. Le Corre, Y. Toudic, C. Lhomer, G. Grandpierre, and M. Gauneau, J. Phys.: Condens. Matter **2**, 479 (1990); A. Kozanecki and A. Szcerbakow, Appl. Phys. Lett. **66**, 3630 (1995); M. A. J. Klik, T. Gregorkiewicz, I. V. Bradley and J.-P. R. Wells, Phys. Rev. Lett. **89**, 227401 (2002).
[28] A. Baldereschi and J. J. Hopfield, Phys. Rev. Lett. **28**, 171 (1972).
[29] W. Nolting and A. M. Olés, Phys. Rev. B **22**, 6184 (1980).
[30] J. K. Furdyna, J. Appl. Phys. **64**, R29 (1988).
[31] P. Chen, C. Piermarocchi, and L. J. Sham, Phys. Rev. Lett. **87**, 067401 (2001).
[32] D. Bacon, J. Kempe, D. A. Lidar, and K. B. Whaley, Phys. Rev. Lett. **85**, 1758 (2000).
[33] U. Woggon, E. Luthgens, H. Wenisch, and D. Hommel, Phys. Rev. B **63**, 073205 (2001).
[34] S. Strauf, P. Michler, M. Klude, D. Hommel, G. Bacher, and A. Forchel, Phys. Rev. Lett. **89**, 177403 (2002).
[35] B. Golding and M. I. Dykman, cond-mat/0309147 (unpublished).
[36] R. Vrijen, E. Yablonovitch, K. Wang, H. W. Jiang, A. Balandin, V. Roychowdhury, T. Mor, and D. DiVincenzo, Phys. Rev. A **62**, 012306 (2000).
[37] B. E. Kane, Nature (London) **393**, 133 (1998).
[38] Y. C. Chang and T. C. McGill, Phys. Rev. B **25**, 3945 (1982).