Exciton-spin dephasing and relaxation due to symmetry breaking in two-band bulk semiconductors

M. Gallart, S. Cronenberger,* C. Brimont, B. Hönerlage, and P. Gilliot†
IPCMS-GONLO, UMR 7504 CNRS-ULP, Boite Postale 4323 rue du Læss, 67034 Strasbourg Cedex 2, France
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The full point group symmetry of a crystal can be broken due to internal or external effective fields. In the study of excitons, such symmetry breaking can lead to a coupling of different exciton states and if a system is prepared in an exciton state with a defined total angular momentum (pseudospin), spin beating is obtained. Looking at the fluctuations of these fields, we use the invariant expansion of an effective Hamiltonian to investigate exciton-spin-relaxation dynamics in a model two-band bulk semiconductor and discuss the respective importance of the different spin-flip processes. We find that interaction terms leading to an electron or hole spin flip give rise to a pure transverse dephasing. Terms where the electron and hole spins are simultaneously reversed lead to transitions between the spin states, which are characterized by the longitudinal relaxation time. Similar to motional narrowing in the case of free carriers, the latter process can lead to an increase in the exciton-spin-relaxation times if extrinsic electric, magnetic, or strain fields rapidly fluctuate in the sample. This effect is shown to be due to the electron-hole exchange interaction.

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I. INTRODUCTION

Free-carrier spin dynamics in bulk semiconductors has been largely studied both theoretically and experimentally[1–4] (for a detailed discussion of spin-relaxation processes and optical orientation, see Ref. 5). The Bir–Aronov–Pikus mechanism[2] considers electron scattering on free or bound holes with a spin flip. In the Elliott–Yafet (EY) mechanism[3] and the D'yakonov–Perel (DP) mechanism,[4] the spin flip is related to the crystal structure and its symmetry: Concerning the (EY) mechanism,[3] at a finite wave vector $\mathbf{k}$, conduction-band states with opposite spins become coupled because of a mixing of conduction- and valence-band states. By using the $\mathbf{k}\cdot\mathbf{p}$ perturbation theory, the spin-relaxation rate for this mechanism is predicted to be proportional to the carrier-momentum relaxation rate.

The main mechanism for electron-spin relaxation in semiconductor crystals lacking inversion symmetry is, however, the D'yakonov–Perel mechanism. In this mechanism, the anisotropic term in the conduction-band dispersion,[6] which is cubic in k, couples electron states of opposite spin. This interaction between conduction-band states may be looked upon as an internal effective magnetic field that is wave-vector dependent. At a given wave vector, the electron spin precesses around this field. This leads to spin beating if the spin states are coherently excited. If the wave vector of the quasiparticles rapidly changes, the effective magnetic field changes at the same time its direction and the resulting spin-relaxation rate is inversely proportional to the carrier-momentum relaxation rate. This effect is known as motional narrowing.

In semiconductors, excitons can be described as a bound state of an electron in the conduction band and a hole in a valence band, which interact via the Coulomb interaction. Excitons also have an internal structure, which is characterized by their total angular momentum (or pseudospin) and which combines the individual electron- and hole-spin states. Excitons or free electron-hole pairs can be optically excited in well-defined spin (or pseudospin states), depending on the excitation conditions. In spite of an intense activity concerning electrons or holes in semiconductors, exciton-spin relaxation has been less studied.[7–9] This is an interesting task, however, since the time evolution of the optically injected exciton-spin states can be followed by optical experiments, e.g., by time resolved pump and probe measurements.

We discussed such measurements in Ref. 10, considering dipole-active excitons in bulk material. We showed that in such a situation, the exciton pseudospin is not defined in a unique way at a finite center of mass wave vectors $\mathbf{Q}$. Because of the strong coupling between dipole-active excitons and the light field giving rise to polaritons as propagating quasiparticles, their wave functions have to be defined as being transverse or longitudinal with respect to $\mathbf{Q}$. The symmetry of the exciton wave function is, indeed, not defined only by the crystal symmetry but also by the propagation direction. This makes quite inappropriate the choice of a spin quantization axis that would be kept fixed along a crystal axis. As we showed, this has important consequences for the spin relaxation of dipole-active excitons and excludes motional narrowing,[10] which is a possible mechanism for free electrons and holes.

By using the well-established description of the exciton states,[11,12] we evidence here some specific properties of exciton-spin-relaxation processes, which depend on their coupling to external electric or magnetic perturbations that they encounter. We use an invariant expansion of the Hamiltonian for this discussion.[11,12] This enables us to identify the symmetry properties of the interactions, which give rise to well-identified relaxation and coupling schemes. We will discuss in this paper that, similar to the finite wave vector in the case of free carriers, several components of extrinsic perturbation fields may lead to an increase in the spin coherence time of excitons if they are rapidly fluctuating. The values of the resulting dephasing times can be determined from the strengths of the perturbing fields. A numerical calculation of these values is, however, beyond the scope of this paper.

II. INVARIANT EXPANSION OF THE HAMILTONIAN

Let us discuss the simplest case of a two-band bulk semiconductor, with a lowest conduction band that is only spin degenerate and an equally twofold degenerate highest valence band. The semiconductor is supposed to have a zinc blende structure (T_d point group symmetry) and a direct band gap at the center of the Brillouin zone. The spin degenerate lowest conduction band is assumed to have Γ_6^c symmetry and the highest valence-band Γ_7^v symmetry. All other bands are neglected. (This situation is realized in CuCl, to which our discussion fully applies. The model also gives, however, indications for the exciton relaxation dynamics if the highest valence band of the semiconductor has Γ_8^v symmetry and is split in the presence of a crystal field or if all couplings to the dark exciton states can be neglected.) We use the wave functions tabulated in Ref. 11. The electron states in the conduction band with $L^e=0$ are thus determined only by the electron spin with $S_z^e=\pm 1/2$. Here, α^e and β^e denote the electron spin up and down states, respectively. (In the following, we will denote excitons by X, electrons by e, and holes by h as superscripts on S, L, J, and σ.) Concerning the valence band, the hole wave function with $L^h=1$ contains an angular part (transforming as x, y, and z) as well as a hole spin contribution (labeled as α^h and β^h). Thus,

$$\phi_5 = (1/\sqrt{3})[z\alpha^h + (x+iy)\beta^h], \quad \phi_6(1\sqrt{3})[-z\beta^h + (x-iy)\alpha^h] \tag{1}$$

denote the valence-band states of Γ_7 symmetry, which are eigenfunctions of the total angular momentum $J^h=1/2$ of the hole, with eigenvalues for J_z^h equal to $+1/2$ and $-1/2$, respectively.

The conduction band and valence band are both twofold degenerate and the exciton ground state is thus only fourfold degenerate. The excitons transform like Γ_2^X and Γ_5^X, respectively. The triplet Γ_2^X exciton corresponds to an eigenstate of pseudospin $J^X=0$, which does not carry a dipole moment. We further consider that optical transitions between valence and conduction bands are dipole allowed: The Γ_5^X exciton state consists of two transverse excitons, which couple to the light field and a longitudinal state, in which the dipole moment is orientated along the wave vector $\mathbf{Q}$.

One obtains for the Γ_2 state in the electron-hole representation the exciton eigenvector,[11,12]

$$|0,0\rangle_z = (1/\sqrt{2})(\beta^e\phi_6 + \alpha^e\phi_5),$$

and for the three Γ_5 states,

$$|1,+1\rangle_z = \alpha^e\phi_6,$$

$$|1,0\rangle_z = (1/\sqrt{2})(\beta^e\phi_6 - \alpha^e\phi_5),$$

$$|1,-1\rangle_z = -\beta^e\phi_5. \tag{2}$$

The states $|1,\pm 1\rangle_z$ and $|1,0\rangle_z$ are eigenstates of the exciton pseudospin with $J^X=1$. They are circularly polarized along the z crystal axes with a pseudospin component $J_z^X=\pm 1$ or 0. The Γ_2 state $|0,0\rangle$ with zero pseudospin is a triplet state.

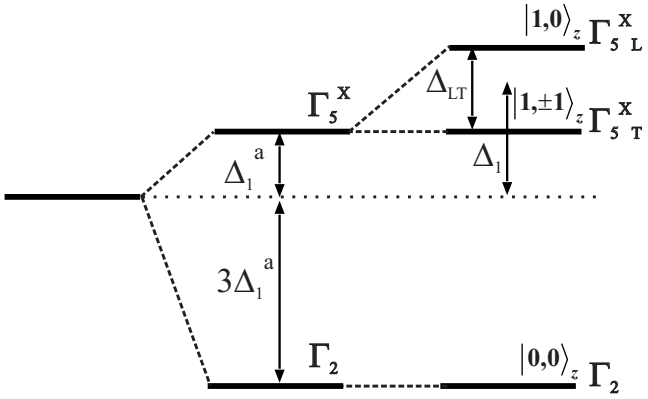

FIG. 1. Splitting of the exciton levels as induced by the analytic and nonanalytic exchange interaction. The z axis of quantization, which is used to identify the $|J^X,J_z^X\rangle_z$ states, is parallel to the wave vector $\mathbf{Q}$.

Due to the electron-hole exchange interaction,[11–14] the degeneracy of the exciton ground state is partially lifted at the Γ point (Fig. 1). While the energy of the triplet exciton decreases, the energies of transverse and longitudinal excitons increase. Furthermore, in the strong coupling regime, the energies of longitudinal and transverse excitons have different values because of the presence of the nonanalytical exchange interaction.

We now construct an effective exciton Hamiltonian that has the same symmetry properties as the full Hamilton operator,[11–14] i.e., which remains invariant under all symmetry operations (Γ_1) of the point group of the crystal and under time reversal (K^+). Since the conduction and valence bands are only twofold degenerate, we choose the Pauli spin matrices $\boldsymbol{\sigma}_x^e$, $\boldsymbol{\sigma}_y^e$, and $\boldsymbol{\sigma}_z^e$ and the unity matrix $\mathbf{1}^e$ and, likewise, $\boldsymbol{\sigma}_x^h$, $\boldsymbol{\sigma}_y^h$, $\boldsymbol{\sigma}_z^h$, and $\mathbf{1}^h$ as basis matrices to span the electron and hole subspaces. The transformation properties of these matrices are (Γ_1, K^+) in the case of the unit matrix and (Γ_4, K^-) for the Pauli matrices, respectively. The Hamiltonian of the exciton ground states now reads[11,12]

$$H_0 = \Delta_0\mathbf{1}^e \otimes \mathbf{1}^h + \Delta_1\boldsymbol{\sigma}^e \cdot \boldsymbol{\sigma}^h, \tag{3}$$

where the parameters Δ_0 and Δ_1 account for the exciton binding energy as well as for the different electron-hole exchange interactions.

In general, the Hamiltonian H depends on an additional set of physical quantities, which can be due to extrinsic perturbations (as magnetic fields $\mathbf{B}$, electric fields $\mathbf{E}$, strain ε, and so on) or to intrinsic perturbations that depend on the exciton center of mass wave vector $\mathbf{Q}$. Let us in the following consider their influence on the exciton-spin dynamics.

A. Magnetic field dependence

According to the transformation properties (Γ_4, K^-), the linear magnetic field dependence takes the form

$$H_{B^1} = g_e\boldsymbol{\sigma}^e \otimes \mathbf{1}^h \cdot \mathbf{B} + g_h\mathbf{1}^e \otimes \boldsymbol{\sigma}^h\mathbf{B}, \tag{4}$$

where g_e and g_h are the Landé factors of electrons and holes, respectively. The terms linear in B couple to the electron- or

hole-spin states and can lead to a spin flip of one of the carriers, while the other particle remains in its former state.

The terms quadratic in B contain direct and exchange interactions according to

$$H_{B^2} = \beta_0 B^2 \mathbf{1}^e \otimes \mathbf{1}^h + \beta_1 B^2 \boldsymbol{\sigma}^e \cdot \boldsymbol{\sigma}^h + \beta_2[3(\sigma_x^e \sigma_x^h - \sigma_y^e \sigma_y^h)(B_x^2 - B_y^2) + (2\sigma_z^e \sigma_z^h - \sigma_x^e \sigma_x^h - \sigma_y^e \sigma_y^h)(2B_z^2 - B_x^2 - B_y^2)] + \beta_3[(\sigma_y^e \sigma_z^h + \sigma_z^e \sigma_y^h)B_y B_z + \text{c.p.}], \quad (5)$$

where c.p. stands for cyclic permutation. The β_i are constants. $H_B^1 + H_B^2$ describe all interactions varying up to the second order in the magnetic field B. The term proportional to β_0 gives rise to the quadratic Zeeman effect; the terms proportional to β_1 to β_3 are due to the magnetic field dependence of the exchange interaction. While the term proportional to β_1 accounts for the full point group symmetry of the crystal, the symmetry is broken by the terms proportional to β_2 and β_3. These terms may mix different exciton states at finite magnetic fields and depend on their orientation with respect to the crystal axis. They then describe a simultaneous spin flip of electrons and holes, which is induced by the terms quadratic in B [Eq. (5)]. Similar to the discussion in Ref. 10 for $\mathbf{Q}^n$, all odd orders of $\mathbf{B}^n$ lead to one-particle spin flips and all even orders to simultaneous electron-hole spin flips. In addition, all higher order terms lead to the same coupling schemes between the states as those given in Eqs. (4) and (5).

B. Wave-vector dependent interactions

Concerning the wave-vector $\mathbf{Q}$ dependent terms [transforming as (Γ_5, K^-)], or its nth power, noted as $\mathbf{Q}^n$, their structure and importance is discussed in detail in Ref. 10. Here, we give it for completeness. A term linear in $\mathbf{Q}$ is absent in the Hamiltonian since it is forbidden by the symmetry. The term quadratic in $\mathbf{Q}$ reads

$$H_Q^2 = G_0 Q^2 \mathbf{1}^e \otimes \mathbf{1}^h + \delta_1 Q^2 \boldsymbol{\sigma}^e \cdot \boldsymbol{\sigma}^h + \delta_2[3(\sigma_x^e \sigma_x^h - \sigma_y^e \sigma_y^h)(Q_x^2 - Q_y^2) + (2\sigma_z^e \sigma_z^h - \sigma_x^e \sigma_x^h - \sigma_y^e \sigma_y^h)(2Q_z^2 - Q_x^2 - Q_y^2)] + \delta_3[(\sigma_y^e \sigma_z^h + \sigma_z^e \sigma_y^h)Q_y Q_z + \text{c.p.}]. \quad (6)$$

It is interesting to note that, although $\mathbf{B}$ and $\mathbf{Q}$ have different spatial transformation properties, the interaction terms given in Eqs. (5) and (6) have the same structure, leading therefore to the same coupling scheme of the states. Similar to β_2 and β_3, the point group symmetry is broken by the exchange interaction terms proportional to δ_2 and δ_3. These terms describe simultaneous spin flips of electrons and holes.

Similar to the electron or hole, g factor is the cubic term in $\mathbf{Q}$ which reads as

$$H_Q^3 = K_E\{[(Q_y^2 - Q_z^2)Q_x]\sigma_x^e + \text{p.c.}\}\mathbf{1}^h + K_H \mathbf{1}^e\{[(Q_y^2 - Q_z^2)Q_x]\sigma_x^h + \text{c.p.}\}, \quad (7)$$

where K_E and K_H are again arbitrary constants. As pointed out by Dresselhaus,[6] this term is due to the wave-vector dependence of the spin-orbit coupling in crystals with a zinc blende structure. For a fixed wave vector $\mathbf{Q}$, Eq. (7) has the same structure as Eq. (4). Therefore, the Q^2 and Q^3 terms in Eqs. (6) and (7) may be looked upon as an effective magnetic field. In contrast to $\mathbf{B}$, however, the Q^3 terms lead to an intrinsic coupling of the different hole (and therefore) exciton-spin states. Obviously, there also exist exchange terms bilinear in wave vector and magnetic field, in which we are not further interested here.

C. Electric field and strain dependent interactions

We denote by ε_{ij} the components of the strain tensor in the following: They transform as (Γ_5, K^+) in crystals with T_d point group symmetry. Since the Pauli spin matrices $\boldsymbol{\sigma}^e$ or $\boldsymbol{\sigma}^h$ are odd functions under time reversal (K^-), interactions involving $\mathbf{E}$ or ε_{ij} and being linear in only one of the $\boldsymbol{\sigma}^e$ or $\boldsymbol{\sigma}^h$ matrices cannot occur. Thus, single spin-flip processes cannot show up but the exciton spin has to be returned in a block by these perturbations. Again, the electron-hole exchange interaction leads to the terms that have the same structure as Eqs. (5) and (6). That is, in the case of strains,

$$H_\varepsilon = \gamma_0(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})\mathbf{1}^e \otimes \mathbf{1}^h + \gamma_1(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})\boldsymbol{\sigma}^e \cdot \boldsymbol{\sigma}^h + \gamma_2[3(\sigma_x^e \sigma_x^h - \sigma_y^e \sigma_y^h)(\varepsilon_{xx} - \varepsilon_{yy}) + (2\sigma_z^e \sigma_z^h - \sigma_x^e \sigma_x^h - \sigma_y^e \sigma_y^h) \times (2\varepsilon_{zz} - \varepsilon_{xx} - \varepsilon_{yy})] + \gamma_3[(\sigma_y^e \sigma_z^h + \sigma_z^e \sigma_y^h)\varepsilon_{yz} + \text{c.p.}]. \quad (8)$$

Since $\mathbf{E}^2$ and ε_{ij} have the same transformation properties, the interaction terms quadratic in the electric field $\mathbf{E}$ have the same structure as those given in Eq. (8). One only has to replace the strain tensor elements ε_{ij} by $E_i E_j$ and the corresponding constants are named as α. In addition, there exists a term linear in $\mathbf{E}$, which takes the form,

$$H_E = \kappa_3[(\sigma_y^e \sigma_z^h + \sigma_z^e \sigma_y^h)E_x + \text{c.p.}], \quad (9)$$

where Eqs. (8) and (9) give the complete set of perturbations linear in E and ε_{ij} and quadratic in $E_i E_j$.

III. ROLE OF SPIN: ORBIT COUPLING AND EXCHANGE INTERACTIONS

Electron and hole spins are coupled through the exchange interaction, which contains two contributions:[10–14] a short range (analytical) and a long range (nonanalytical) part. By choosing the cubic crystal axes as the coordinate system and using the exciton wave functions of Eq. (2), the matrix representation of the exciton Hamiltonian defined by Eq. (3) can be obtained. As shown in Fig. 1, the analytical exchange interaction lifts the degeneracy of the three pseudospin states with $J^X=1$ and the triplet exciton with $J^X=0$ by the energy $4\Delta_1^a$. Considering, for simplicity, the wave vector $\mathbf{Q}$ parallel to the cubic z axis, we can separate the three pseudospin states with $J^X=1$ into one longitudinal exciton state with $J_z^X=0$ and two circular polarization states with $J_z^X=\pm 1$. Then, in addition, the nonanalytic exchange interaction increases the energy of the longitudinal exciton with respect to the transverse (dipole-active) states, leading to a splitting of these states at $Q=0$. Δ_{LT} denotes the longitudinal-transverse splitting. As it was discussed in detail in Refs. 11 and 12,

TABLE I. Interaction terms between excitons, as given by Eq. (4) or (7), which are due to odd powers of magnetic fields or equivalent quantities. They are due to spin-orbit coupling of exciton or hole states and show up in the exciton energies.

	$\|0,0\rangle$	$\|1,+1\rangle$	$\|1,0\rangle$	$\|1,-1\rangle$
$\|0,0\rangle$	$\Delta_0-3\Delta_1^a$	$-(\zeta+i\eta)^-$	$-h_z+e_z$	$(\zeta-i\eta)^-$
$\|1,+1\rangle$	$-(\zeta-i\eta)^-$	$\Delta_0+\Delta_1^a+h_z+e_z$	$(\zeta-i\eta)^+$	0
$\|1,0\rangle$	$-h_z+e_z$	$(\zeta+i\eta)^+$	$\Delta_0+\Delta_1^a+\Delta_{\mathrm{LT}}$	$(\zeta-i\eta)^+$
$\|1,-1\rangle$	$(\zeta+i\eta)^-$	0	$(\zeta+i\eta)^+$	$\Delta_0+\Delta_1^a-h_z-e_z$

longitudinal and transverse states can only be separated after the exciton wave vector $\mathbf{Q}$ has been specified.

The exchange interaction now separates into

$$\Delta_1 = \Delta_1^a + \Delta_{\mathrm{LT}} f(\mathbf{Q},\Theta), \tag{10}$$

where Θ denotes the angle between the dipole moment and the wave vector $\mathbf{Q}$. Equations (4) and (7) give rise to interaction terms, which separately involve spin flips of electron or holes. They originate from a spin-orbit coupling that leads to coupling of exciton states. Let us denote

$$e_x = g_e B_x \text{ or } K_E[(Q_y^2 - Q_z^2)Q_x],$$

$$e_y = g_e B_y \text{ or } K_E[(Q_z^2 - Q_x^2)Q_y],$$

$$e_z = g_e B_z \text{ or } K_E[(Q_x^2 - Q_y^2)Q_z], \tag{11a}$$

and similarly

$$h_x = g_h B_x \text{ or } K_H[(Q_y^2 - Q_z^2)Q_x],$$

$$h_y = g_h B_y \text{ or } K_H[(Q_z^2 - Q_x^2)Q_y],$$

$$h_z = g_h B_z \text{ or } K_H[(Q_x^2 - Q_y^2)Q_z], \tag{11b}$$

depending on whether Eq. (4) or (7) is considered. The interaction matrix then takes the form given in Table I.

Moreover, $\Delta_0-3\Delta_1^a$ accounts for the energy of the triplet exciton, $\Delta_0+\Delta_1^a$ gives the energy of the transverse excitons, and $\Delta_0+\Delta_1^a+\Delta_{\mathrm{LT}}$ that of the longitudinal exciton state at $\mathbf{Q}=0$. Here, $(\zeta+i\eta)^{\pm}=[(e_x+ie_y)\pm(h_x+ih_y)]/\sqrt{2}$ and $(\zeta-i\eta)^{\pm}=[(e_x-ie_y)\pm(h_x-ih_y)]/\sqrt{2}$. It is important to note that the terms in Eqs. (4) and (7) directly couple states with the $J=0$ state ($J_z=0$) to the three states with the $J=1$ states [$J_z=(+1,0,-1)$].

Concerning Eq. (5), (6), and (8), or (9), they describe interaction terms, which involve simultaneous spin flips of electrons and holes. They originate from the exchange interaction that may also depend on intrinsic or extrinsic perturbations. These terms are symmetric under time reversal and may therefore also depend on electrical or strain fields. We obtain the following coupling scheme from these terms:

$$M = \beta_0 B^2,\ G_0 Q^2,\ \gamma_0(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}),\ \text{or}\ \alpha_0 E^2,$$

$$L = \beta_1 B^2,\ \delta_1 Q^2,\ \gamma_1(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}),\ \text{or}\ \alpha_1 E^2,$$

$$x = \beta_3 B_y B_z,\ \delta_3 Q_y Q_z,\ \gamma_3 \varepsilon_{yz},\ \text{or}\ \alpha_3 E_y E_z \text{ or } \kappa_3 E_x,$$

$$y = \beta_3 B_z B_x,\ \delta_3 Q_z Q_x,\ \gamma_3 \varepsilon_{zx},\ \text{or}\ \alpha_3 E_z E_x \text{ or } \kappa_3 E_y,$$

$$z = \beta_3 B_x B_y,\ \delta_3 Q_x Q_y,\ \gamma_3 \varepsilon_{xy},\ \text{or}\ \alpha_3 E_x E_y \text{ or } \kappa_3 E_z,$$

$$u = 3\beta_2(B_x^2 - B_y^2),\ 3\delta_2(Q_x^2 - Q_y^2),$$

$$3\gamma_2(\varepsilon_{xx}-\varepsilon_{yy}),\ \text{or}\ 3\alpha_2(E_x^2 - E_y^2),$$

$$v = \beta_2(2B_z^2 - B_x^2 - B_y^2),\ \delta_2(2Q_z^2 - Q_x^2 - Q_y^2),$$

$$\gamma_2(2\varepsilon_{zz}-\varepsilon_{xx}-\varepsilon_{yy}),\ \text{or}\ \alpha_2(2E_z^2 - E_x^2 - E_y^2). \tag{12}$$

The symmetry breaking effects considered here do not couple the $J=0$ state with the $J=1$ states.

IV. EXCITON-SPIN DYNAMICS AND RELAXATION

Equations (11a) and (11b) show the well-known result that the terms, which are linear in the magnetic field or cubic in wave vector, lead to the same coupling scheme between the different exciton states. Therefore, the terms cubic in wave vector can be considered as an effective magnetic field. The coupling of the exciton states results in spin dephasing if an exciton is prepared in a well-defined spin state. Therefore, field fluctuations lead to spin relaxation.

We see in Table I that dipole-active states ($J^X=1$, $J_z^X=\pm 1$) are only coupled to dark states ($J^X=0$ or $J^X=1$, but $J_z=0$). In order to simplify the system, let us consider the energies of the states: In the absence of perturbations, only ($J^X=1$, $J_z^X=\pm 1$) states are degenerate, all other states are at different energies due to the exchange interaction. Therefore, at small but finite perturbations, one can neglect the dark states and consider the nearly degenerate transverse states only. These dipole-active states can be selectively excited by an electromagnetic field leading to optical injection of a spin. Now, considering the two states subspace of transverse excitons ($J^X=1$, $J_z^X=\pm 1$) in the matrix form of Eq. (4) (Table I), one sees that the diagonal elements of the interaction are different due to the effective magnetic field, but the nondiagonal elements are equal to zero. When the two circularly polarized states are interpreted as spin states in a two level model, this behavior is the signature of a purely longitudinal effective magnetic field. The well-known behavior of a two-level system when subjected to fluctuations in time or space of such a field can be applied here. It leads to fluctuations of the energy splitting between these states, which affect the Rabi frequency and give rise to pure dephasing of the states, which are characterized by a pure transverse relaxation time constant $T_\perp^*$. Pure dephasing keeps the population of the states constant but changes the coherence relation between the states. It is interesting to note that only fluctuations of e_z and h_z (i.e., of the field components B_Z and $[(Q_x^2-Q_y^2)Q_z]$) have an influence on this dephasing process. The other components are responsible for a coupling to the dark states, which have been neglected.

The interaction terms given in Table II, which is the matrix form of Eq. (5) and equivalent, involve the electron-hole exchange interaction, which corresponds to a simultaneous

TABLE II. Interaction terms between excitons, as given by Eq. (5), (6), and (8), or (9), which are due to even powers of magnetic fields or equivalent quantities. They are due to the electron-hole exchange interaction in excitons.

	$\|0,0\rangle$	$\|1,+1\rangle$	$\|1,0\rangle$	$\|1,-1\rangle$
$\|0,0\rangle$	$\Delta_0-3\Delta_1^a+M-3L$	0	0	0
$\|1,+1\rangle$	0	$\Delta_0+\Delta_1^a+M+L+2v$	$\sqrt{2}(y-ix)$	$6u-2iz$
$\|1,0\rangle$	0	$\sqrt{2}(y+ix)$	$\Delta_0+\Delta_1^a+\Delta_{\mathrm{LT}}+M+L-4v$	$\sqrt{2}(-y+ix)$
$\|1,-1\rangle$	0	$6u+2iz$	$\sqrt{2}(-y-ix)$	$\Delta_0+\Delta_1^a+M+L+2v$

spin reversal of electron and hole. The interaction matrix shows that the states with $J^X=0$ and $J^X=1$ are not coupled. If we consider again that the longitudinal-transverse splitting is large compared to the coupling between the states, we can restrict our discussion to the two states subspace of transverse excitons ($J^X=1$, $J_z^X=\pm 1$). We see that the diagonal elements are equal, which correspond to a common energy shift of both states, but a Rabi frequency equal to zero. The nondiagonal elements are different from zero, however, indicating that the states are coupled. This configuration corresponds to the effect of a pure transverse effective magnetic field in a two level model. This gives rise to spin beating between the two states. If the field fluctuates, additional transitions are induced between the states, which lead to spin relaxation. It is characterized by the longitudinal $T_\parallel$ relaxation time, which governs the spin population dynamics, and by a corresponding transverse dephasing time $T_\perp=2T_\parallel$.

Let us consider the DP mechanism in the case of free carriers wherein the quantization direction is fixed and the magnetic field randomly changes its direction. In the motional narrowing regime, the system will be subject to an effective, time-dependent, randomly oriented magnetic field **B**. It very rapidly changes its direction with a characteristic time τ_{col} that is much shorter than the precession time. In this case, the precession cannot follow the randomly orientated field and the spin memory is conserved over long times.

As discussed in detail in Ref. 10, motional narrowing cannot occur in bulk dipole-active excitons due to changes in the wave vector since the direction of quantization cannot be kept constant in scattering processes in which the direction of propagation changes. In contrast, when considering electric fields or strain fields as described in Eq. (8) or (9), we see that they can give rise to such effects: If again the circularly polarized exciton states are interpreted as spin states in a two level model, these strain or electric fields correspond to pure transverse effective magnetic fields. The spin precesses around this transverse field, leading to spin beating. If the effective magnetic field changes in direction or module, the spin precession changes accordingly, leading to spin relaxation. As discussed above, such fluctuations do not affect the pure dephasing rate of the excitons described by the interactions enumerated in Table I. The interactions discussed in Table II, however, can lead to an increase in the spin coherence time: if the characteristic fluctuation time is much shorter than the precession time, the precession cannot follow the randomly orientated field and the spin memory is conserved over longer times.

It is interesting to note that the different components of the electric field are not equally important: Concerning the term that is linear in the electric field, only rapid fluctuations of E_z in which the direction changes can slow down exciton-spin relaxation. Then, the precession direction is reversed and the spin state is stabilized. The same effect occurs if higher order components of the magnetic or electric field [as B_xB_y and $(B_x^2-B_y^2)$ or E_xE_y and $(E_x^2-E_y^2)$] or fluctuations of the strain field components ε_{xy} and $(\varepsilon_{xx}-\varepsilon_{yy})$ become important. In such a case, rapid fluctuations of the field direction may be at the origin of an increase in the exciton-spin-relaxation times.[15]

V. CONCLUSION

We have shown that the spin dynamics of excitons is very different from that of free carriers in bulk semiconductors since exciton- and free-carrier states have different symmetries. Therefore, their spin dynamics are differently affected by symmetry breaking perturbations.

On the contrary to the D'yakonov–Perel mechanism for free carriers, which results from spin-orbit coupling, scattering of dipole-active excitons leads to an important decrease in their spin coherence time and not to motional narrowing. On the other hand, electrons and holes are coupled through exchange interaction. As shown in Eq. (12), similar to the motional narrowing case for free carriers, rapid fluctuations of some components of magnetic, electric, or strain fields can lead to an increase in the spin coherence time for excitons. Such fields lead to transitions between the spin states but do not affect the pure dephasing.

If the electron-hole exchange interaction is sufficiently strong, our results are also valid for dipole-active excitons in systems with a more complicated band structure or in quantum wells, where important electric field or strain fluctuations occur. In such situations, coupling to all dark exciton states can be neglected and the exciton system can be reduced to the subspace of the two nearly degenerate dipole-active exciton states, which obey the relaxation scenario discussed above.

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*Present address: Groupe d'étude des Semiconducteurs-GES, UMR 5650 CNRS-Universitée Montpellier 2, Place Eugène Bataillon, 34095 Montpellier Cedex, France.
†pierre.gilliot@ipcms.u-strasbg.fr

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