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Local-field effects and excitonic polaritons in semiconductors: A new insight

R. Bonneville
Centre National d'Etudes des Télécommunications, 196 rue de Paris, 92220 Bagneux, France*

G. Fishman
Groupe de Physique des Solides de l'Ecole Normale Supérieure,† 4 place Jussieu, 75005 Paris, France

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The influence of the local-field effects on the interpretation of the excitonic polariton dispersion curves in cubic semiconductors is investigated. It is shown that in real compounds for which the background dielectric constant is appreciably greater than one and for which the electron and the hole of the exciton are weakly bound, the usual description of the polariton effect is no more valid. In particular, the longitudinal-transverse splitting no longer preserves the center of gravity of the optically allowed multiplet; as a consequence, the value of the exchange energy which can be deduced from the available experimental data can be modified up to an order of magnitude.

I. INTRODUCTION

Since the pioneering work of Thomas and Hopfield on cadmium sulfide,[1] a number of theoretical and experimental studies have been devoted to excitonic polaritons[2] in semiconductors, and recent progress in dye lasers has allowed very accurate investigations in this field.[3-7] As a starting point, the transverse excitonic frequency and the longitudinal-transverse splitting (LTS) related to the oscillator strength measuring the photon-exciton coupling look sufficient to characterize the polariton effect near the center of the Brillouin zone[8]; nevertheless, reality can be more complex. In wurtzite compounds (e.g., CdS), the separation between transverse excitonic branches and longitudinal excitonic branches is imposed by the C_{6v} crystal-field symmetry.[1] In cubic crystals, those among the excitonic modes which are coupled to the electromagnetic field in the electric dipole approximation are associated with three-time-degenerate energy levels.[8,9] When the long-range dipolar exciton-exciton interaction is taken into account, the triplet excitonic level splits into two levels: The singlet upper one is the longitudinal exciton level and the twice-degenerate lower one is the transverse exciton level. In a first-order perturbation theory, the center of gravity of these levels is preserved,[8,10] and this last point is important if one aims to calculate the exact exchange energy from the experimental data. As a matter of fact, in the cubic T_d semiconductors where the conduction and the valence band are, respectively, of Γ_6 and Γ_8 symmetry, it is now possible to obtain in the same experiment the transverse and longitudinal frequencies (Γ_5), and also the energies of the usually optically forbidden levels ($\Gamma_3+\Gamma_4$).[7,11,12]

At this stage, it is worth noting two limitations to the demonstrations of Refs. 8 and 9: First, they are performed by assuming that the background dielectric constant is equal to one; i.e., there is no higher resonance frequency in the crystal. Such a simplifying hypothesis is not realistic, since as a matter of fact this constant is of the order of 10 in the excitonic energy range ($\epsilon_\infty \simeq 9$ in ZnSe, $\epsilon_\infty \simeq 5$ in CuBr). Second, the calculation is strictly valid only for a localized, i.e., tightly bound exciton (Frenkel exciton), while in many semiconductors the hole and the electron are weakly bound (Wannier exciton).[9,13] So one can ask what modifications to these ideas occur in real semiconductors where the background dielectric constant is appreciably greater than one[14] and where the excitons may be strongly delocalized. We shall see later on that these features can greatly influence the interpretation of the available experimental data.[15] In both cases, however, the relevant question is: What is the effective field undergone by an elementary excitation in the crystal?

More explicitly, the two following relations are customarily applied to excitons in cubic semiconductors,[9,11,16] assuming that there is only one resonance in the frequency range of interest:

$$\omega_L + 2\omega_T = 3\omega_0 \;, \tag{1}$$

$$\epsilon = \epsilon_\infty + 4\pi\beta/(1-\omega^2/\omega_T^2) \;. \tag{2}$$

In these equations, ω_L and ω_T are, respectively, the longitudinal and transverse frequencies, ω_0 is the bare exciton frequency, i.e., when the long-range part of the exchange interaction is not taken into account. For Wannier excitons where the effective-mass approximation is valid, ω_T is a quadratic function of the wave vector $\vec{k}$, $\epsilon = (kc/\omega)^2$ is the dielectric function, and $4\pi\beta$ is the so-called oscillator strength. Equation (1) expresses the conservation of the center of gravity of the excitonic multiplet level, and Eq. (2) expresses the dispersion relation of the coupled exciton-photon mode. If the medium presents two resonances in the same frequency range, the dispersion curve is then currently written as[11,16]

$$\epsilon = \epsilon_\infty + \frac{4\pi\beta_1(\vec{k})}{1-\omega^2/\omega_1^2} + \frac{4\pi\beta_2(\vec{k})}{1-\omega^2/\omega_2^2}\,. \tag{3}$$

It is precisely the goal of this paper to discuss the validity of those well-known ideas, and particularly:

(i) to give evidence that Eq. (1) is never verified for excitonic polaritons in real semiconductors;

(ii) to show that Eq. (3) cannot be used without caution since the local-field effects come out through a renormalization of the oscillator strengths and frequencies;

(iii) to propose a set of more general equations to be used instead of Eqs. (1)–(3), and to make precise the conditions of applicability of (1)–(3) which appear as limiting cases (i.e., $\epsilon_\infty = 1$, no charge delocalization) of our results; and

(iv) to investigate the consequences upon the values of the parameters deduced from the experiments by using various relations.

Section II deals with the polariton effect in the presence of a polarizable background within the Lorentz local-field approximation. In Sec. III we consider the delocalization of the excitation where the above approximation is no longer valid. Section IV is devoted to the interpretation of some recent experimental results concerning the exchange energy.

Before beginning explicit calculations, we have to point out that for excitonic polaritons the full quantum-mechanical calculations, though necessary to obtain the position and dispersion of the excitonic level as well as the coupling strength between photon and exciton, essentially lead for the polariton dispersion curve to the same formal results as a classical theory.[8] So, henceforth, we shall develop a non-quantum-mechanical treatment, which is easier to carry through.

II. FRENKEL EXCITONS AND POLARIZATION OF THE BACKGROUND

In this section we intend to investigate the consequences upon the dielectric properties of the exciton of the other resonance levels in the crystal. Another point of view is to consider the exciton as a polarizable quasiparticle immersed in a continuum characterized by the background dielectric constant ϵ_∞. Let E be the macroscopic electric field inside the medium; the dielectric polarization P can be viewed as originating from a set of polarizable oscillators. The ith oscillator is characterized by a dispersion relation $\omega_i(\vec{k})$ and its polarizability at the frequency ω is given by[17]

$$\alpha_i(\vec{k},\omega) = \frac{\alpha_i^{(0)}(\vec{k})}{1-\omega^2/\omega_i^2(\vec{k})}\,. \tag{4}$$

If we now assume that the effective field undergone by such an oscillator is the Lorentz local field,[18] $F_i = E + \frac{4}{3}\pi P$ (it is worth noting here that strictly speaking this hypothesis is meaningful only when the charge densities are sharply localized and thus refers to Frenkel's description; this point will be discussed in more detail in Sec. III). Then the dielectric function $\epsilon(\vec{k},\omega)$ is given by

$$\epsilon(\vec{k},\omega) = 1 + 4\pi\frac{\sum_i n_i\alpha_i(\vec{k},\omega)}{1-\frac{4}{3}\pi\sum_i n_i\alpha_i(\vec{k},\omega)}\,, \tag{5}$$

where n_i equals the density of oscillators of the ith kind. Let us suppose that the frequency of interest ω_0 is much lower than the upper resonances $\omega_{i\neq 0}$. The background dielectric constant ϵ_∞ originating from those high levels is then given by Eq. (5) where the summation is now restricted to $i \neq 0$.

Then, a short calculation leads to the following expression of the dielectric constant in the vicinity of ω_0:

$$\epsilon(\vec{k},\omega) = \epsilon_\infty + \frac{4\pi n_0\alpha_0(\vec{k},\omega)[\frac{1}{3}(\epsilon_\infty+2)]^2}{1-\frac{4}{3}\pi n_0\alpha_0(\vec{k},\omega)[\frac{1}{3}(\epsilon_\infty+2)]}\,. \tag{6}$$

Equation (6) can be formally written as Eq. (2) if one sets

$$\beta = \frac{n_0\alpha_0^{(0)}[\frac{1}{3}(\epsilon_\infty+2)]^2}{1-\frac{4}{3}\pi n_0\alpha_0^{(0)}[\frac{1}{3}(\epsilon_\infty+2)]}\,. \tag{7}$$

The dimensionless quantity $n_0\alpha_0^{(0)}$ characterizes the photon-exciton coupling. In the linear-combination-of-atomic-orbitals approximation it is given by

$$n_0\alpha_0 = \frac{2N}{V}\frac{|eX_{12}|^2}{\hbar\omega_0}\,,$$

where N/V is the number of atoms per volume unit and eX_{12} is the atomic electric dipole matrix element between the ground state and the excited state. The transverse and longitudinal frequencies ω_T and ω_L are the solutions of $\epsilon^{-1}(\omega_T) = 0$ and $\epsilon(\omega_L) = 0$, i.e.,

$$\begin{aligned} \omega_T^2 &= \omega_0^2 - \tfrac{4}{3}\pi[\tfrac{1}{3}(\epsilon_\infty+2)]n_0\alpha_0^{(0)}\omega_0^2\,, \\ \omega_L^2 &= \omega_0^2 + (8\pi/3\epsilon_\infty)[\tfrac{1}{3}(\epsilon_\infty+2)]n_0\alpha_0^{(0)}\omega_0^2\,. \end{aligned} \tag{8}$$

From Eqs. (8) one obtains the equality

$$\epsilon_\infty\omega_L^2 + 2\omega_T^2 = (\epsilon_\infty+2)\omega_0^2\,. \tag{9}$$

Since in excitonic states the LTS $E_{LT} = \hbar\omega_L - \hbar\omega_T$

is much weaker than $\hbar\omega_0$, Eq. (9) reduces to

$$\epsilon_\infty \omega_L + 2\omega_T \simeq (\epsilon_\infty + 2)\omega_0 . \tag{10}$$

At this point we want to make some remarks. Only if $\epsilon_\infty = 1$ does Eq. (6) appear identical to Eqs. (29) or (30) of Ref. 8, and only in this case is Eq. (1) verified. However, Eq. (1) has been frequently applied until now together with Eq. (2), although they are not compatible in actual cases ($\epsilon_\infty > 1$). It must be pointed out that using Eq. (6), and therefore Eq. (10), instead of Eqs. (1) and (2) can greatly alter the value of the exchange energy (up to an order of magnitude) as will be shown in Sec. IV.

Precisely, the interest of Hopfield's calculations is to show that *in Frenkel's description* the dipolar exciton-exciton interaction is equivalent to a local-field effect which leads to Eq. (1) if no other polarizable excitation is taken into account. The important point is the following: Introducing a realistic background dielectric constant $\epsilon_\infty > 1$ requires that the center of gravity of the multiplet be no longer conserved, even at first order. The reason is the following: In a first-order perturbation theory, the conservation of the center of gravity comes from the assumption that the degenerate level is coupled to the other levels weakly enough so that those levels will not appreciably intervene in the removal of the degeneracy. Now, the background dielectric constant precisely accounts for the coupling to the higher levels, which can be neglected strictly only if $(\epsilon_\infty - 1) \ll 1$.

III. DELOCALIZATION OF THE EXCITON

Until now, we have supposed that the excitons undergo the Lorentz local field while saying it was only valid for localized excitations (Frenkel's excitons), and so we found again the commonly used formulas in the limit $\epsilon_\infty \to 1$. Now, one knows[19] that in semiconductors the elementary electronic excitations are delocalized and more suitably described by the Mott-Wannier picture. However, the complete quantum-mechanical treatment of the dipolar exciton-exciton interaction for Wannier excitons would be tremendous[20] and has never been published[21]; nevertheless, it appears that in this case the effective field differs from the Lorentz field,[22] and one can question its expression. One knows[23] that for completely delocalized charge distributions, the dielectric constant is more accurately given by the Sellmeier formula

$$\epsilon = 1 + 4\pi n\alpha , \tag{11}$$

which means that the effective field is just the macroscopic field in the medium, than by the Lorentz formula

$$\epsilon = 1 + \frac{4\pi n\alpha}{1 - \frac{4}{3}\pi n\alpha} . \tag{12}$$

On the other hand, the delocalization of the exciton (we mean how closely the electron and the hole are bound together) is indeed connected to the delocalization of the charge density. If the atomic overlap is weak, the electron and the hole stay concentrated on the same center, although the exciton can migrate from one center to another. Conversely, if the overlap is important, the electron and the hole are weakly bound and may be separated by several cells. Most generally, one can write the effective field undergone by an exciton as

$$F = E + \tfrac{4}{3}\pi\gamma P , \tag{13}$$

where the local-field factor γ, lying between 0 and 1, is an increasing function of the charge localization measured by the ratio of the cell parameter to some characteristic length of the extension of the atomic charge.[19] The upper limit $\gamma = 1$ corresponds to a strongly localized density, and thus to Frenkel's description. The effective field is then properly described by a Lorentz field, and thus Eq. (6) is fully valid. Conversely, a Wannier exciton corresponds to a completely delocalized excitation, so it is very likely that there is no local-field correction at all; the effective field is just the macroscopic field in the medium and $\gamma \to 0$. Now, most generally, one deals with an intermediate case where $0 < \gamma < 1$.

One can take up again the preceding calculation which introduced the γ factor, and one finds[24]

$$\epsilon = 1 + \frac{4\pi \sum_i n_i \alpha_i}{1 - \frac{4}{3}\pi\gamma \sum_i n_i \alpha_i} . \tag{14}$$

If the frequency ω_0 of the level of interest is very much weaker than all the others, some algebra shows that the dielectric function is given by

$$\epsilon(\omega) = \epsilon_\infty + \frac{[4\pi n_0 \alpha_0^{(0)}/(1 - \omega^2/\omega_0^2)]\{\frac{1}{3}[3 + \gamma(\epsilon_\infty - 1)]\}^2}{1 - \frac{4}{3}\pi\gamma[n_0\alpha_0^{(0)}/(1 - \omega^2/\omega_0^2)]\{\frac{1}{3}[3 + \gamma(\epsilon_\infty - 1)]\}} . \tag{15}$$

The expressions of the transverse and longitudinal frequencies are

$$\begin{aligned} \omega_T^2 &= \omega_0^2 - \tfrac{4}{3}\pi\gamma\{\tfrac{1}{3}[3 + \gamma(\epsilon_\infty - 1)]\} n_0 \alpha_0^{(0)} \omega_0^2 , \\ \omega_L^2 &= \omega_0^2 + \tfrac{4}{3}\pi[(3 - \gamma)/\epsilon_\infty]\{\tfrac{1}{3}[3 + \gamma(\epsilon_\infty - 1)]\} n_0 \alpha_0^{(0)} \omega_0^2 , \end{aligned} \tag{16}$$

from which one gets the relation

$$\gamma\epsilon_\infty \omega_L^2 + (3 - \gamma)\omega_T^2 = [3 + \gamma(\epsilon_\infty - 1)]\omega_0^2 . \tag{17}$$

Both $\hbar\omega_L$ and $\hbar\omega_T$ increase but E_{LT} decreases when γ goes from 1 to 0; clearly, the center of gravity is not conserved. For $\gamma = 1$, one finds again the results of the preceding section. In the completely delocalized case ($\gamma = 0$), the dielectric constant is given by a Sellmeier-type formula:

$$\epsilon = \epsilon_\infty + \frac{4\pi n_0 \alpha_0^{(0)}}{1 - \omega^2/\omega_0^2} ,$$

where ω_T goes to ω_0, and ω_L^2 to

$$\omega_0^2 (1 + 4\pi n_0 \alpha_0^{(0)}/\epsilon_\infty) .$$

The case $\gamma < 1$ with $\epsilon_\infty = 1$ can be obtained from the above formulas if needed.[25] We note that Eqs. (15) and (16) are different from the corresponding ones (i.e., taking into account the electronic delocalization) which have been obtained for phonons [see Eqs. (39) and (40) of Ref. 19].

If one deals with two oscillators of frequencies ω_1 and ω_2 of the same order, both much lower than the upper resonances, the local-field effects couple the two oscillators together so that it is less legitimate to treat them separately the closer the frequencies are. It is a straightforward matter to find that

$$\epsilon = \epsilon_\infty + \frac{\left(\frac{4\pi n_1 \alpha_1^{(0)}}{1 - \omega^2/\omega_1^2} + \frac{4\pi n_2 \alpha_2^{(0)}}{1 - \omega^2/\omega_2^2}\right)\left(\frac{3 + \gamma(\epsilon_\infty - 1)}{3}\right)^2}{1 - \frac{4}{3}\pi\gamma\left(\frac{n_1 \alpha_1^{(0)}}{1 - \omega^2/\omega_1^2} + \frac{n_2 \alpha_2^{(0)}}{1 - \omega^2/\omega_2^2}\right)\left(\frac{3 + \gamma(\epsilon_\infty - 1)}{3}\right)} , \tag{18}$$

with

$$\epsilon_\infty = 1 + \frac{4\pi \sum_{i \neq 1,2} n_i \alpha_i}{1 - \frac{4}{3}\pi\gamma \sum_{i \neq 1,2} n_i \alpha_i} . \tag{19}$$

Some manipulations of rational fractions show that Eq. (18) can be written as

$$\epsilon = \epsilon_\infty + \frac{4\pi\beta_1^*}{1 - \omega^2/\omega_1^{*2}} + \frac{4\pi\beta_2^*}{1 - \omega^2/\omega_2^{*2}} , \tag{20}$$

which is formally identical to (3). However, one has to keep in mind that ω_1^*, ω_2^*, β_1^*, and β_2^* are complicated expressions of γ, ϵ_∞, ω_1, ω_2, $n_1\alpha_1^{(0)}$, and $n_2\alpha_2^{(0)}$. Equation (20) expresses two kinds of couplings between the two oscillators: The first one is made explicit in Ref. 17 whereas the second one is due to the local-field corrections. It is only when $\gamma = 0$ that the equations used in Refs. 11 and 16 for two interaction resonances are found again[26]; in this case, the second type of coupling vanishes.

IV. CONSEQUENCES UPON THE VALUE OF THE EXCHANGE ENERGY

In light of the preceding, we aim to discuss in this section the interpretation of some experimental results of interest. In some semiconductors[7,11] where the conduction band belongs to the Γ_6 symmetry and the valence band to the Γ_8 symmetry, the energies $E(J,M)$ of the levels originating from the $\Gamma_6 \times \Gamma_8$ exciton have been measured. The short-range part of the exchange interaction splits the eightfold-degenerate level of the $\Gamma_6 \times \Gamma_8$ exciton into a triplet $\Gamma_5\,|J = 1, M\rangle$ of energy $\hbar\omega_0$ and a quintuplet $(\Gamma_3 + \Gamma_4)|J = 2, M\rangle$, whose separation is precisely the so-called exchange energy Δ. The long-range part of the exchange interaction splits the triplet level into a longitudinal mode $|1, 0\rangle$ of energy $\hbar\omega_L$ and a transverse mode $|1, \pm1\rangle$ of energy $\hbar\omega_T$. The $|2, M\rangle$ states are not coupled to the electromagnetic field in the center of the Brillouin zone in the electric dipole approximation, but the $|2, M\rangle$ states can mix[27] with the $|1, M\rangle$ states at a nonzero wave vector, which allows one to know their position.[7,11] From an experimental point of view, the two attainable parameters are

$$E(1,0) - E(1,\pm1) = E_{LT} \text{ and } E(1,\pm1) - E(2,M) = \delta .$$

The LTS is thus known without any further interpretation, but we see (Fig. 1) that the measurement of the exchange energy Δ demands the knowledge of the exact relation between Δ and the splittings δ and E_{LT}. From Eq. (17), one easily deduces

$$\Delta = \delta + E_{LT} \frac{\gamma\epsilon_\infty}{\gamma(\epsilon_\infty - 1) + 3} . \tag{21}$$

Table I shows the practical influence on the value of the exchange energy of various expressions using different values of ϵ_∞ and γ. It appears that this influence may be sometimes weak (ZnSe) but it can also modify the calculated exchange energy by an order of magnitude (CuBr) according to the equation involved.[28]

We can now answer the following question: What kind of experiments should be made to decide upon

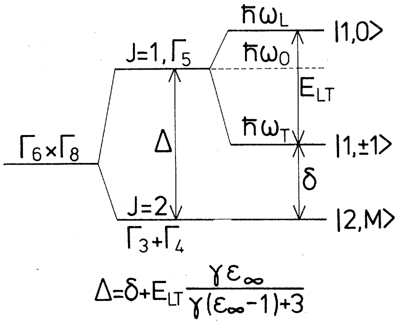

FIG. 1. Schematic diagram showing the position and symmetry of the levels originating from the $\Gamma_6 \times \Gamma_8$ exciton in cubic semiconductors. The underlying formula shows the relation between the exchange energy Δ and the actually measured energy splittings δ and E_{LT} (longitudinal-transverse splitting); ϵ_∞ is the background dielectric constant and γ the delocalization factor.

TABLE I. Influence of the effective-field expression on the calculated exchange energy. (The experimental data for CuBr and ZnSe are extracted from Refs. 11 and 7.)

Energies (meV)		CuBr ($\epsilon_\infty = 5.4$)	ZnSe ($\epsilon_\infty = 9.0$)
Experimental data	$E_{\rm LT}$	12	1
	δ	1.5	1.8
Δ	$\Delta = \delta + \frac{1}{3}E_{\rm LT}$ $(\epsilon_\infty = 1, \gamma = 1)$	5.5	2.1
	$\Delta = \delta + E_{\rm LT}(\epsilon_\infty/\epsilon_\infty + 2)$ $(\epsilon_\infty > 1, \gamma = 1)$	10.3	2.6
	$\Delta = \delta$ $(\gamma = 0)$	1.5	1.8

the local-field correction to be applied to an exciton? Indeed, it is necessary to know independently $E_{\rm LT}$, Δ, and δ. Then, Eq. (17) gives the local-field factor γ. As a matter of fact, $E_{\rm LT}$ and δ can both be obtained in the same experiment,[7,11] and there exist other independent ways to get the exchange energy Δ.[29] Nevertheless, all these measurements must be precise enough to determine the relative energy position of all the levels of interest. Such is the order of magnitude of the exchange interaction that getting such precision appears to be difficult, except in cuprous halides where it is hoped it can be performed.[30]

V. CONCLUSION

We have shown that, even in the Frenkel approximation, it is not consistent to maintain the relation of conservation of the center of gravity of the optically allowed excitons when simultaneously the long-range part of the exchange interaction is taken into account, and that a background dielectric constant larger than 1 is used. More generally, in semiconductors where the Wannier picture is known to describe more accurately the Coulomb interaction between electron and hole, a careful examination of the effective field acting on the excitons is needed to deduce the exchange energy from the experiment. In some cases this value can be altered up to an order of magnitude. Conversely, the precise knowledge of the longitudinal transverse splitting, the so-called energy δ, and the exchange energy Δ permits one to use the relations established in the text to measure the effective field acting on the exciton.

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*Laboratoire associé au CNRS (LA 250).

†Laboratoire associé au CNRS (LA 17).

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[17] As a matter of fact, if the resonance frequency ω_i of an oscillator is very far from the other ones, $\alpha_i^{(0)}(\vec{k})$ does not depend on $\vec{k}$. If the resonance frequencies of two oscillators α_1 and α_2 are close together, $[n_1\alpha_1^{(0)}(\vec{k})+n_2\alpha_2^{(0)}(\vec{k})]$ is constant.

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[25] If γ is smaller than one, this calculation does not give back exactly the results of M. Born and K. Huang, but of course, in any case the well-known relation of Lyddane-Sachs-Teller is verified.

[26] Equation (4) of the paper of J. Lagois [Phys. Rev. B 16, 1699 (1977)] is also obtained for $\gamma=0$. In this paper the local-field effects are implicitly neglected. This is valid because in CdS and ZnO the excitons are Wannier excitons.

[27] In CuBr (Ref. 11) the mixing is due to a $\vec{k}$-linear term and in ZnSe (Ref. 7) the mixing is due to the existence of heavy and light excitons.

[28] In GaAs (Ref. 3) only two branches have been explored, the highest and the lowest ones, but not the intermediate one which originates from the light excitons. In this case it has been shown, in Ref. 16, that the measured value $(E_{\mathrm{LT}})_{\mathrm{expt}}$ of the LTS is in fact equal to $E_{\mathrm{LT}}+\frac{1}{4}\delta$; δ was taken to be equal to $\Delta-\frac{1}{3}E_{\mathrm{LT}}$ which gives $(E_{\mathrm{LT}})_{\mathrm{expt}}=\frac{1}{4}\Delta+\frac{11}{12}E_{\mathrm{LT}}$. One sees that if δ is indeed equal to the exchange energy Δ, then $(E_{\mathrm{LT}})_{\mathrm{expt}}=\frac{1}{4}\Delta+E_{\mathrm{LT}}$. The other conclusions of Ref. 16 are unchanged.

[29] For instance, a value of Δ in GaAs has been obtained from a measurement of the spin-relaxation time of conduction electrons [G. Fishman and G. Lampel, Phys. Rev. B 16, 820 (1977)].

[30] Indeed, in CuBr where the Bohr radius of the exciton is of the order of 9 Å, i.e., weaker than two lattice constants, it is likely that the local-field factor differs from zero; then Eq. (22) is needed to determine γ if E_{LT}, δ, and Δ can be measured.