Nonadiabatic phonon coupling for bound excitons in ZnTe

P. O. Holtz and B. Monemar
Department of Physics and Measurement Technology, Materials Science Division, Linköping University, S-581 83 Linköping, Sweden
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The phonon coupling for some bound-exciton systems in ZnTe has been studied optically in both absorption and emission. The one-phonon—assisted spectra in optical absorption (photoluminescence excitation spectra) have been compared with emission spectra (photoluminescence) for several shallow-bound-exciton systems in ZnTe, with the aid of tunable-dye-laser spectroscopy. The phonon-coupling strength appears to be typically more than an order of magnitude larger in absorption than in emission. These drastic differences between absorption and emission are interpreted as a breakdown of the adiabatic approximation in the bound-exciton—phonon coupling formalism. Strong interaction between phonon-assisted transitions and electronic transitions of comparable energies for the bound excitons may explain the observed nonadiabatic behavior.

I. INTRODUCTION

Bound excitons (BE's) are localized excitations in solids, which can be studied in great detail in optical spectroscopy by virtue of the corresponding very sharp electronic transitions observed in both absorption and emission.[1] The binding energy of the BE depends on the strength of the central-cell potential for one or both bound electronic particles. The phonon-coupling strength is usually increased with the degree of localization of the most tightly bound particle. In the case of a large binding energy for the BE, it is generally observed that the strength of the phonon coupling is similar in both kinds of optical transitions (i.e., absorption and emission, corresponding to creation and annihilation, respectively, of the localized excitation). This is nicely demonstrated as a mirror symmetry in optical spectra, observed, e.g., for ZnTe:O (Ref. 2) and GaP:Cd,O (Ref. 3). Here the phonon wing in the absorption spectrum has its counterpart in the emission spectrum (luminescence) both concerning shape and coupling strength.

Our knowledge about the phonon coupling to the more shallow BE states is often less detailed, mainly because it is usually much weaker. Further, when the binding energy of the BE is small, the absorption spectra related to the BE overlap with much other stronger optical transitions close to the band gap. It has generally been assumed that a simple picture with linear phonon coupling is adequate even in this case, in the absence of relevant data proving the opposite. In this paper we demonstrate that this appears not to be correct, at least not for some systems in the direct band-gap semiconductor ZnTe, which we have studied in detail in this respect. Comparing absorption and emission spectra for shallow BE's with moderately weak phonon coupling, we find consistently that the overall coupling strength to the phonon spectrum differs remarkably from absorption to emission, the former is more than an order of magnitude larger. We propose that the rather drastic differences observed in both shape and coupling strength of the phonon spectrum between BE absorption and emission is due to a breakdown in the adiabatic approximation usually applied in treatments of phonon coupling to electronic transitions in solids. The physical reason for this breakdown in absorption will be discussed in terms of a strong interaction between the phonon-assisted excitations and the electronic excited states of the BE.

In Sec. II experimental details about samples and measurements will be summarized. Section III contains the main experimental results on optical spectra for BE's, related to different Cu-related defects in ZnTe referred to in this paper. Both photoluminescence spectra (partly with tunable-dye-laser excitation) and absorption spectra (mainly selective absorption data derived from dye-laser-excited excitation spectra) are described in detail, with emphasis on the phonon-coupling problem.

In Sec. IV the general formalism for phonon coupling in BE spectra is briefly discussed. The Toyozawa model[4,5] for exciton-phonon interaction is mentioned, and its relevance to the observed optical-phonon coupling in this work is pointed out. A discussion on the electronic structure of the studied BE system in ZnTe is provided, with reference to extensive investigations published separately.[6,7] Finally, a tentative picture is sketched on general ideas of phonon coupling for bound excitons, along which a theory for the observed enhancement of the active one-phonon spectrum in BE absorption may be developed in the future.

II. SAMPLES AND EXPERIMENTAL PROCEDURE

Bulk single-crystalline ZnTe, doped with Cu by a single-step diffusion procedure was employed in the experiments reported here. Details of the sample preparation and doping have been reported separately, and are not repeated here.[6,7] Highly Cu-doped samples used in this work contain a total Cu concentration in the $(10^{17}-10^{18})$-cm^{-3} range, distributed among a large number of dif-

ferent Cu-related defects.[7]

Optical spectra were recorded in transmission at 2 K with a conventional tungsten lamp as the light source. The light transmitted through the sample was recorded via a 0.75-m double monochromator. For the photoluminescence measurements the same monochromator was used, but the excitation source was either an Ar^+ laser or a tunable cw dye laser, which could be continuously scanned with a stepping motor arrangement.

III. EXPERIMENTAL RESULTS FOR PHONON COUPLING IN BOUND-EXCITON SPECTRA

The physical system studied in this work, i.e., Cu-doped ZnTe, offers unique possibilities for a detailed study of phonon interaction in optical transitions involving bound excitons. The main advantage is that a large number of bound-exciton spectra occur at photon energies well separated from the band gap, i.e., they are dispersed over a range >0.1 eV towards lower energies.[6,7] This is illustrated in Fig. 1 for a typical case of rather strongly Cu-doped ZnTe, exhibiting several sharp lines in photoluminescence (PL), due to excitons bound to different Cu-related complex defects.[7] Obviously it is possible in this case to study phonon coupling of bound excitons in both emission and absorption, over at least the entire one-phonon range of phonon-assisted transitions, a rather unusual situation for semiconductors. For shallow substitutional donor or acceptor-related BE states in ZnTe, e.g., only phonon-assisted spectra from the optical emission process is experimentally available. In addition, the application of tunable-dye-laser excitation permits the recording of selectively excited spectra for each defect-related exciton. This can be applied both to defect selective absorption via photoluminescence excitation (PLE) spectra, and in addition to selectively excited photoluminescence (SPL) spectra.

The effect of such selective excitation for a particular BE in ZnTe is shown in Fig. 2. With nonselective Ar^+ laser excitation, few details are observed in the phonon-assisted part of the BE recombination spectrum, which in addition is disturbed by overlapping electronic lines associated with other defect BE's (Fig. 2). With selective-dye-laser excitation in the electronic line of the particular defect BE (2.3452 eV), the overlapping structure associated with other defects decreases, and the one-phonon wing in PL is sufficiently enhanced in intensity to reveal novel spectral features (Fig. 2), which were hidden in the background with nonselective excitation.

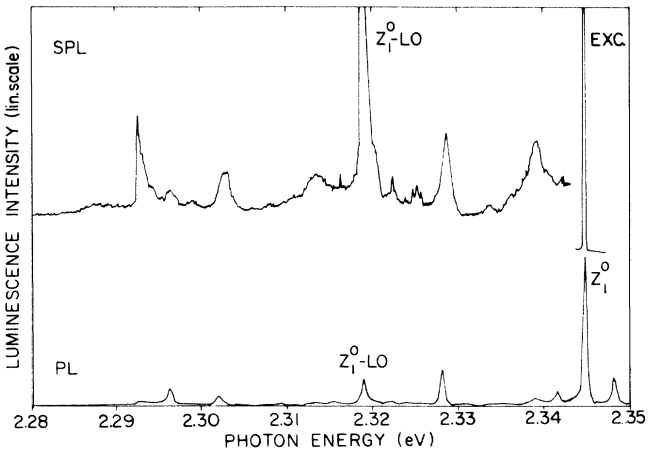


FIG. 2. Portion of the photoluminescence spectrum in the vicinity of the Z_1 emission, with excitation above the band gap (5145 Å) in the lower curve. With selective-dye-laser excitation in the electronic Z_1^0 line, the details in the phonon spectrum are better resolved, as shown in the upper curve.

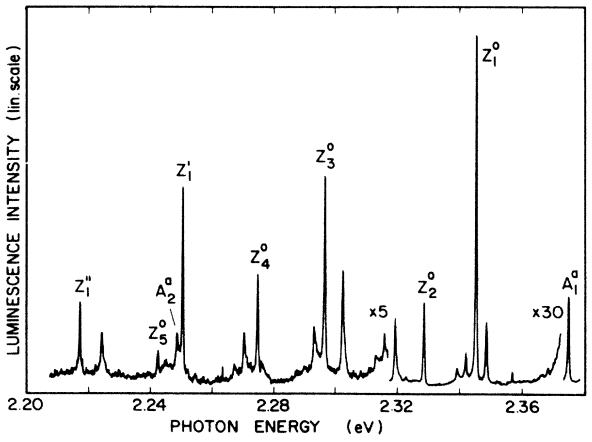


FIG. 1. Photoluminescence spectrum for strongly Cu-doped ZnTe. The sample has been doped at a diffusion temperature of 550°C. The spectrum is measured at 2 K with above band-gap excitation (5145 Å) from an Ar^+ laser.

The spectral details revealed in the one-phonon wing of such SPL spectra include an appreciable coupling to the continuum low-energy acoustic phonons, upon which quasilocalized defect-related modes are superimposed. Such quasilocalized modes are common for BE spectra associated with complex defects.[8] In addition, sharp so-called gap modes (also defect-related) appear further away from the electronic line. In the optical-phonon range a rather strong structure is usually observed, probably due to a combination of lattice modes and quasilocalized modes (Fig. 2). (This is obvious since the energies and shape of this structure differ slightly for BE spectra related to different defects.) Finally, the LO^Γ phonon is rather strong, as is normally the case in such BE emission spectra.

A typical transmission spectrum for a similarly Cu-diffused ZnTe crystal is shown in Fig. 3. Obviously the same electronic bound-exciton lines as in Fig. 1 are seen in transmission, with good resolution. Also, phonon-assisted transitions are observed, here at photon energies higher than the electronic BE line. These features are difficult to observe in detail in transmission spectra, since all bound excitons overlap spectrally. This means that the strength of various phonon replicas are rather difficult to estimate. In addition, for the more shallow bound excitons, e.g., Z_1, the associated one-phonon wing is too close to the band gap to be detected with good resolution (Fig. 3).

A much more detailed picture of the one-phonon part

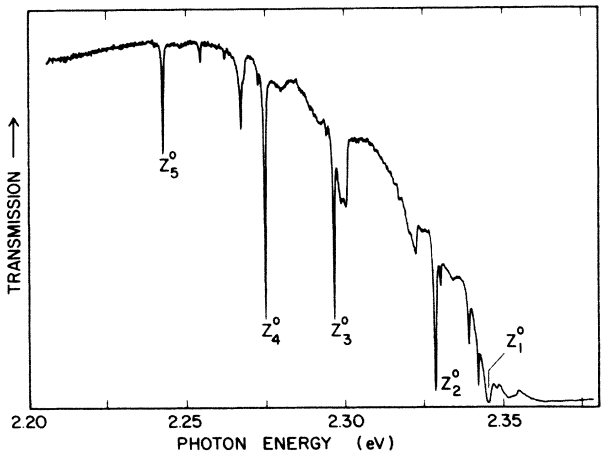
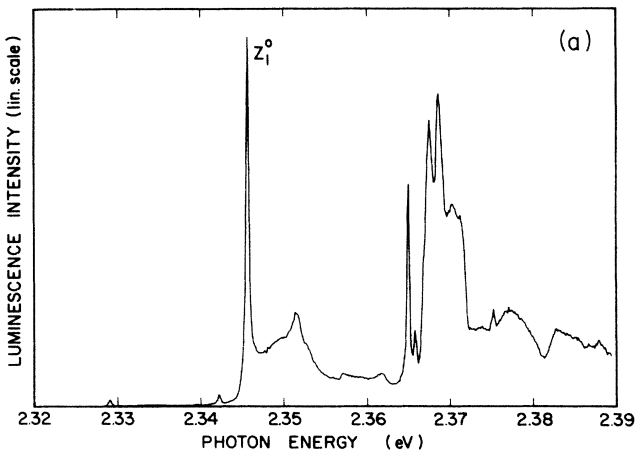


FIG. 3. Transmission spectrum for the same Cu-doped ZnTe sample as in Fig. 1.



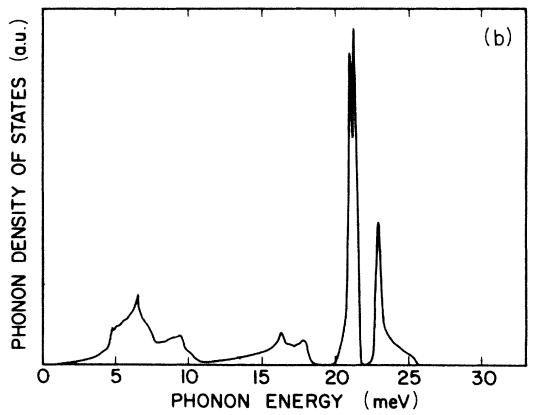


FIG. 4. (a) Photoluminescence excitation (PLE) spectrum for the Z_1 emission obtained with tunable-dye-laser excitation with the detection in the LO replica of the electronic Z_1^0 line. (b) Calculated phonon density of states for ZnTe (from Ref. 21).

of the BE absorption spectrum is obtained with tunable-dye-laser-excited PLE spectra. A typical such spectrum for the Z_1 BE is shown in Fig. 4(a). Obviously this selective spectrum shows only the absorption spectrum associated with the detected BE, and therefore provides remarkably detailed picture of the one-phonon-assisted spectrum of the Z_1 BE. This spectrum contains the details shown in the PL spectrum of Fig. 2, but with a strongly-enhanced coupling strength for the one-phonon part compared to emission. In addition, the LO replica is completely missing in absorption, but a very strong coupling is instead observed to several bands in the optical range just below the LO phonon energy [Fig. 4(a)].

The integrated enhancement of the one-phonon part between emission (PL) and absorption (PLE) can be more than an order of magnitude, e.g., for the Z_1 BE system this factor is about 30. Comparison with transmission data reveals that the sample employed for Fig. 4(a) was sufficiently thin to allow a homogeneous excitation profile across the sample, also for the electronic BE line. From a comparison with the computed phonon density of states [Fig. 4(b)], it seems clear that the coupling is strongly enhanced in BE absorption for the entire one-phonon spectrum (including the defect-related quasilocalized modes and gap modes). This remarkable effect was not previously discussed in the literature, to our knowledge. Further, the observed spectrum in the energy range of optical phonons [Fig. 4(a)] suggests strong renormalization effects of the LO-phonon energy in BE absorption. Such effects were observed previously,[9–12] but were less obvious than in this case, and were assigned to "bound phonons."[4,5]

In the above presentation the effects of phonon coupling in BE spectra were illustrated for one particular defect, the Z_1 Cu complex with a BE at 2.3452 eV. To emphasize that the observed phenomena are of a general nature for BE states in ZnTe, additional examples of Cu-related BE spectra in both emission (PL) and absorption (PLE) are shown in Fig. 5. A comparison between PL and PLE spectra for the same BE shows that the observed asymmetry for the strength of phonon coupling between BE emission and absorption is observed in all cases. Similarly, the strong renormalization effects in the optical-phonon range occur as a general phenomenon. These differences between emission and absorption are strongest for more shallow-BE states, which have a relatively much stronger phonon coupling in absorption, while the phonon coupling in PL is of comparable strength for both shallow and slightly deeper Cu-related BE states in ZnTe.

IV. DISCUSSION

A. General model for phonon coupling in defect-related optical transitions

Usually, optical transitions related to localized electronic excitations at defects in semiconductors involve some simultaneous interaction with phonons.[13] This leads to the well-known phonon sidebands in optical spectra, which have a strength relative the electronic line(s) varying over many orders of magnitude between different materials and different defects.[13] The simple model usually discussed in treatments of such phonon coupling involves

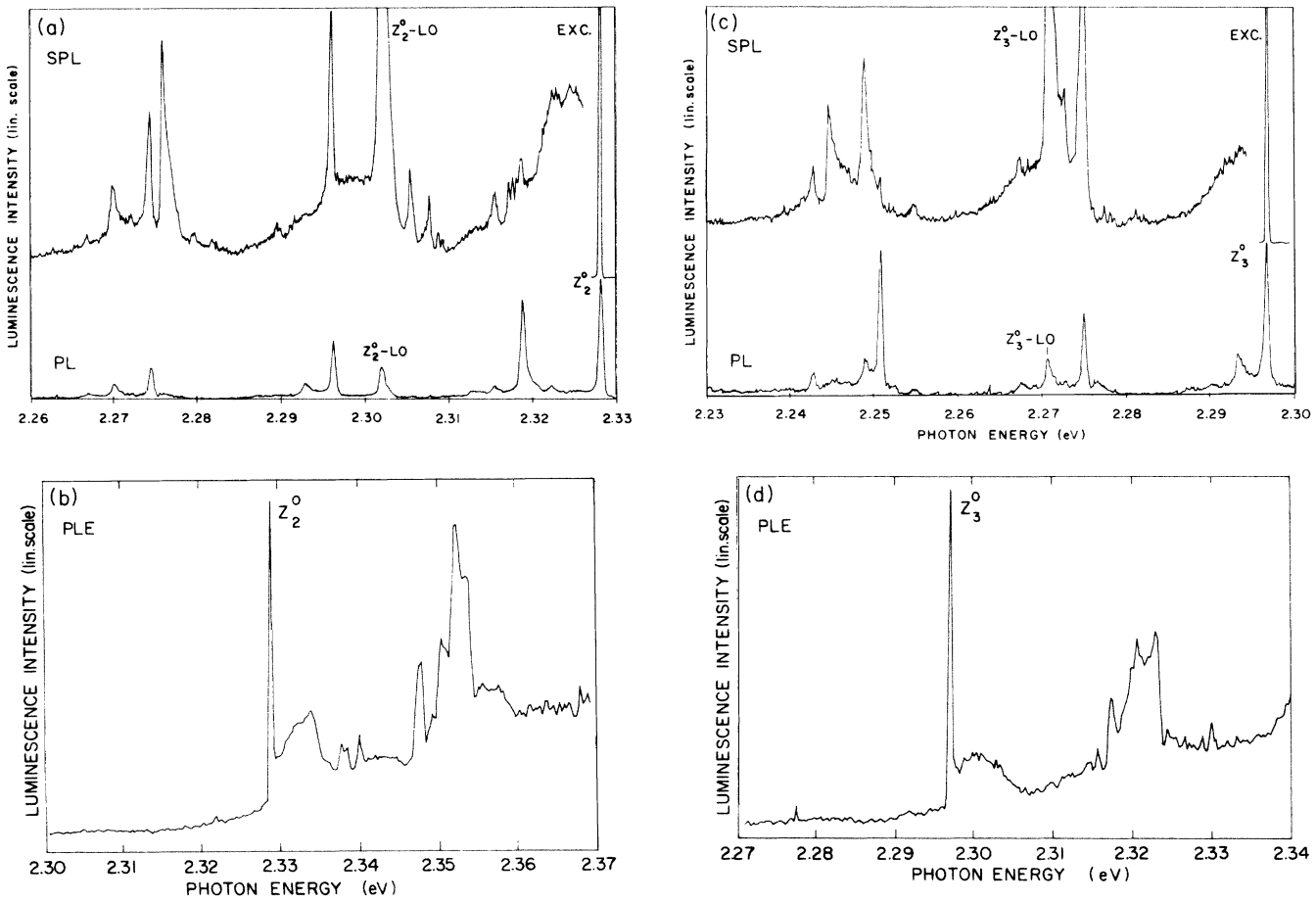


FIG. 5. PL and PLE spectra for two different BE emissions. These spectra show that the effects observed for the Z_1 emission concerning the enhancement of the phonon coupling in PL compared with absorption (PLE), as well as the renormalization effects in the optical-phonon range, appear as a general phenomenon. The BE emissions shown in this figure have no-phonon lines at (a) and (b) 2.3296 eV and (c) and (d) 2.2980 eV, respectively.

the so-called adiabatic approximation. In this model the electronic and vibrational wave functions $\varphi_e(\mathbf{r},\mathbf{q})$ and $X(\mathbf{q})$, respectively, for a localized transition (like a BE excitation) are treated as independent, separate quantities. The total wave function $\psi(\mathbf{r},\mathbf{q})$ for a BE may then be written as a simple product $\psi(\mathbf{r},\mathbf{q})=\varphi_e(\mathbf{r},\mathbf{q})X(\mathbf{q})$.[13] This treatment appears adequate when it can be assumed that the defect-related vibrations interacting with the electronic transition studied are independent of the dynamics of the electronic excitation. The simplest case within this adiabatic approximation involves linear coupling to phonons, which is usually sufficient to explain the general features observed in defect-related optical spectra. Similar phonon coupling is often observed in emission and absorption.[2,3] Asymmetries between emission and absorption for a defect-related optical transition may simply be explained in terms of different adiabatic potentials for the two vibronic states studied.[13]

The explanation previously offered for the so-called bound-phonon problem for excitons in semiconductors and insulators obviously require a treatment beyond the adiabatic approximation. A strong interaction between purely electronic and phonon-assisted transitions has been suggested, which is by definition a nonadiabatic phenomenon.[4,5] The previous treatments of this renormalization problem for optical phonons will be briefly discussed below, and its relevance for the phonon coupling observed in the BE spectra discussed in this paper will be critically evaluated. Complementary models to treat all aspects of the strongly asymmetric phonon coupling in this case will also be mentioned in a tentative fashion.

B. Interaction between electronic and phonon-assisted transitions for bound excitons

The model suggested by Toyozawa[4,5] is based on an interaction between electronic excited states of the bound exciton $\varphi_e^{\lambda}(\mathbf{r},\mathbf{q})$ (λ is an index for the different excited electronic states) and the states involving the lowest electronic bound-exciton state $\varphi_e^0(\mathbf{r},\mathbf{q})$ plus a phonon state, $X_k(\mathbf{q})$. In this case the total wave function $\psi(\mathbf{r},\mathbf{q})$ for the exciton-phonon bound state can be schematically written as

$$\psi(\mathbf{r},\mathbf{q})=\sum_{\lambda}\alpha_{\lambda}\varphi_e^{\lambda}(\mathbf{r},\mathbf{q})X_0(\mathbf{q})+\sum_{k}\beta_k\varphi_e^0(\mathbf{r},\mathbf{q})X_k(\mathbf{q})\ .$$

Here λ denotes the different electronic states of the bound exciton, $\lambda=0,1,2,\ldots$. Consequently $\varphi_e^0(\mathbf{r},\mathbf{q})$ is the lowest fundamental electronic BE state. $X_0(\mathbf{q})$ denotes the zero-phonon vibrational state, while $X_k(q)$ are the one-phonon states for all phonon modes k interacting with the BE under study.

It has been shown that realistic values for parameters involved in the exciton-phonon coupling leads to a lowering of the energy for the LO-phonon replica in absorption of typically 10% compared with the normal value for LO_Γ observed in optical emission spectra for bound excitons in, e.g., AgBr,[12] CdS,[14] and ZnTe.[15] This has been interpreted as evidence for the relevance of the creation of an exciton-phonon bound state induced by an attractive force between the exciton and the LO phonon.[4,5] Experimental evidence for the existence of such bound states have also been suggested, e.g., for excitons bound to donors in GaP (where the effects are weak),[10] and for NN pairs in GaP.[16]

Oscillator strengths $|\langle 0|P|\psi^*\rangle|^2$ for optical transitions from the ground state $\langle 0|$ to the exciton-phonon bound state $|\psi^*\rangle$ via the dipole operation P has also been estimated by Toyozawa and Hermanson for the case of LO-phonon interaction.[5] The result is that the ratio between the transition to $|\psi^*\rangle$ and the zero-phonon transition $|\psi^0\rangle$ for a bound exciton state can easily exceed 1 if the exciton binding energy exceeds the LO phonon energy.[4] No calculation for the ratio between the strength of the ordinary LO-phonon replica in emission and absorption, respectively, was presented, however.

The Toyozawa treatment of exciton-phonon interaction in absorption was restricted to the LO phonon via the simple Fröhlich coupling term.[4] Similar effects for coupling to other optical phonons, as well as to the entire acoustical-phonon branch, was not included in this treatment.[4,5] In fact, the discrete model of one bound-phonon mode is not very natural when coupling to the entire phonon spectrum of the crystal occurs. This problem will be commented on in more detail below.

C. Discussion on the electronic structure of bound excitons in ZnTe

To evaluate the relevance of models for interaction between electronic and phonon-assisted transitions in BE absorption it is necessary to have a detailed picture of the electronic states involved. The picture is here slightly complicated by the fact that the defects binding the excitons under study here are complex defects, for which the electronic structure of the associated bound excitons has not previously been studied in detail. The phenomena discussed in this paper are common to excitons bound at both acceptors and neutral "isoelectronic" associates. In the acceptor case the situation is different for complex acceptors and single substitutional acceptors in tetrahedral symmetry, both with respect to the splitting of the bound-exciton states due to exchange interaction and the effect of the local strain field at the defect. In tetrahedral symmetry these effects cause a threefold, rather small, splitting of the lowest bound-exciton states, usually within about 1 meV.[17,18] For complex defects, on the other hand, the axial strain field has a very strong effect on the bound-hole states, so that for acceptors generally only one BE state is observed in the band gap.[7] A similar situation is found for the case of neutral "isoelectronic" defects, where only one hole and one electron are bound to the defect. The electron-hole exchange interaction for such BE states is usually very small in cases where the bound-electron wave function is delocalized, as is the case for the particular Cu-related defects with hole-attractive central-cell potentials referred to in this work.[7]

Assuming therefore that the bound exciton has a single state at lowest energy, with no many-particle excited states in the band gap, the spectrum of excited electronic states to the bound exciton is reduced to possible single-particle excitations. Zeeman data at 10 T for the BE lines discussed in this work reveal a strong diamagnetic shift rate for both acceptors and neutral "isoelectronic" defects.[7] This is indicative of a loosely-bound electron, consistent with the well-known "pseudo-donor model"[18] for acceptor bound excitons, or the Hopfield-Thomas-Lynch (HTL) model[19] for the case of neutral isoelectronic defects with a hole-attractive central-cell potential. Thus we may expect single-particle excitations of bound electrons into excited states, and even into continuum states. In this case one might expect the continuum edge to be close to the binding energy of a shallow donor electron, or slightly smaller as a result of the repulsive effect of the defect central-cell potential on the electron. Since the binding energy of shallow donors in ZnTe is about 19 meV,[20] the continuum edge for excitations of the electron in the bound exciton is assumed to be about 15–19 meV. This is a lower energy than the optical-phonon branch in ZnTe.[21] Therefore the excitations of optical phonons simultaneous with the bound excitons is a process resonant with a continuum of electronic excitations of the bound exciton. This situation is schematically visualized in Fig. 6.

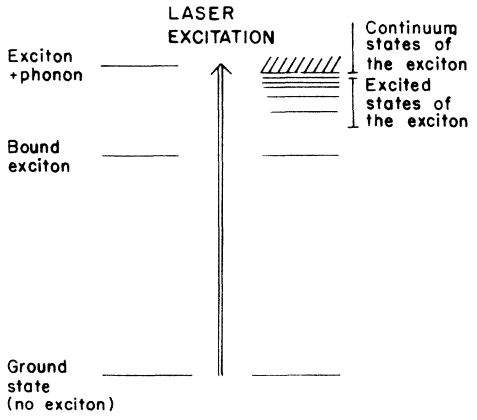


FIG. 6 Illustration of the model for nonadiabatic behavior in the phonon coupling for BE's. The phonon-assisted transitions are resonant with the continuum for electronic excitations of the BE, giving rise to a strong interaction between these transitions. Therefore the usually applied adiabatic approximation will not be valid, but instead a strong asymmetry between the phonon-coupling strength in PL and absorption (PLE) will result.

D. Enhancement of phonon coupling involving a broad spectrum of phonons

The Toyozawa model for bound-phonon states may adequately explain the observed downshift in energy of the LO phonon in absorption in the BE spectra reported in this paper. A considerable enhancement of the coupling strength of this perturbed "LO" mode in absorption compared to the strength of the LO replica in PL emission is also in agreement with simple estimates of oscillator strengths for the absorption case.[4] It should be noted that in the case of the Cu-related BE's in ZnTe discussed here, the renormalization effects are indeed dominating, since no trace is observed of the normal LO_Γ replica in absorption. Factors favoring a strong effect of exciton-phonon interaction are a comparable magnitude of the exciton binding energy E_x and the optical-phonon energy ω_{LO}, preferably $E_x > \omega_{LO}$, and a large difference between effective masses for electrons and holes ($m_e \approx 0.12 m_0$; $m_h \approx m_0$ in ZnTe). Further, the factor $(\epsilon_0^{-1} - \epsilon_\infty^{-1})$ appearing in the Fröhlich coupling Hamiltonian for LO phonons is also quite large in ZnTe. Therefore conditions for strong renormalization effects of the LO phonon in absorption are favorable according to the Toyozawa theory.[4,5] The strength of the interaction between electronic exciton-related transitions and phonon-assisted processes is favored in this case by the comparable energies for the continuum of one-particle (electron) excitations and the LO-phonon energy (see Sec. IV C). In this sense the observed broad features in the optical-phonon range of the one-phonon sideband in absorption of Fig. 4 should rather be described as "quasibound" phonon states, consistent with their strength and width.[5]

However, the general spectral shape of the enhanced phonon coupling in absorption (Fig. 4) looks like an enhancement of the entire one-phonon spectrum (including quasilocalized defect-related modes and gap modes). The Toyozawa theory suitable for treatment of the phonon interaction for a discrete LO phonon is inadequate in describing this continuum spectrum in absorption. Clearly any model based on a renormalization of a discrete exciton-phonon state is too simplified when the state is part of a broad continuum. What is needed in this case is a theory that provides a more general description of the enhancement of exciton-phonon coupling.

The exciton-phonon coupling to a continuum of acoustic and optical phonons is, in general, described by a quite different Hamiltonian than the simple Fröhlich term in the Toyozawa treatment (appropriate for interaction between free excitons and LO phonons). The interaction between an electronic particle and phonons in the one-phonon continuum can be described by the deformation potential and the piezoelectric interaction processes.[22] These coupling mechanisms for continuum modes are generally considered to be less efficient than the Fröhlich term active for LO phonons.[23,24] The drastic enhancement in absorption of the entire one-phonon continuum is therefore not easily attributed to the strength of these Hamiltonians in the framework of ordinary adiabatic phonon coupling. A contribution from exciton-phonon interaction will have to be involved to enhance the oscillator strength in the same way as for the discrete LO phonons. A more complete formulation of the whole theoretical problem of exciton-phonon interaction is required, but will not be attempted in this paper.

For bound-exciton spectra it appears natural to separate the phonon-coupling Hamiltonians for the individual particles, in contrast to the free-exciton case.[4,5] For bound excitons the individual particles usually experience quite different potentials, and their mutual interaction is often quite weak. For the systems studied in this work the defect potentials are, in general, hole-attractive, due to the Cu_{Zn} core. The electron is usually bound as a secondary particle in a "shallow-donor-like" orbit once the hole(s) is (are) bound as primary particle(s). This means that the hole wave function is rather localized [typical Bohr radius ($r_B \approx 5$ Å)] while the electron is delocalized ($r_B \approx 50$ Å). The enhancement of exciton-phonon coupling reported here is similar irrespective of whether the defect is an acceptor or a neutral isoelectronic complex. The effect is not strongly dependent on whether there are one or two holes in addition to the electron in the BE excitation.

A tentative interpretation of these experimental observations would be that the conventional adiabatic phonon coupling might be a reasonable model for the most localized electronic particle, in this case the hole. The electron is quite delocalized for all defect BE's studied here, but the continuum of excited electron states below the band gap appears to be the important factor triggering the nonadiabatic exciton-phonon interaction causing the enhancement effects in the phonon coupling in absorption. This is indeed a remarkable situation, since for a delocalized particle the usual effects of particle-phonon coupling in the adiabatic scheme are quite small, strongly decreasing with a decreasing particle localization at the defect.[23]

The explanation discussed above in terms of mixing of excited electronic BE states and the one-phonon sideband requires that the oscillator strength of the single-particle excitation (in this case excitations of the electron in the BE to an excited state or continuum) is considerably stronger than the no-phonon BE line. This is because the oscillator strength in the enhanced BE-phonon sideband in absorption must, in principle, be taken from the oscillator strength of the interacting excited electronic states.[4] Since the experimental observation is concerned with the mixed BE-phonon states, it is not possible to experimentally test this assumption of a quite high oscillator strength for the excited single-particle states of the BE electron in the cases studied here, unfortunately.

ACKNOWLEDGMENTS

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