Fundamental absorption edge in mixed single crystals of II-VI compounds

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The energy and temperature dependence of the absorption coefficient near the fundamental absorption edge has been measured in mixed single crystals of II-VI compounds. In all the crystals the Urbach rule is valid. Prominent differences have been found in the temperature dependence of the σ parameter of these crystals, according to which class the crystal belongs: amalgamated or persistent type. Most of the results are explained by the Dow-Redfield theory of the internal Franz-Keldysh effect. In order to explain the differences in the optical properties of the crystals of the two classes (Raman, infrared reflection, and the optical absorption near the fundamental absorption edge), a model of coupled oscillators was developed. The application of that model, together with Toyozawa's approach of the self-trapped excitons, helps to explain the above differences. The latter theory was found to be applicable especially in the case where localization of phonons occurs.

I. INTRODUCTION

In a wide variety of materials the absorption coefficient (α) near the fundamental absorption edge is energy (E)dependent according to the Urbach rule:¹

$$\alpha = \alpha_0 \exp[S(E - E_0)], \qquad (1)$$

where α_0 and E_0 are material-dependent constants, and S is the measure of the slope. This dependence refers to the low-energy side of the energy gap, i.e., for $E < E_g$. In addition a σ parameter is defined as

$$\sigma = SkT \quad . \tag{2}$$

In amorphous or polycrystalline materials σ is linearly temperature dependent,^{2,3} but in crystalline materials σ behaves as4,5

$$\sigma = \sigma_0 \frac{2kT}{\hbar\omega_0} \tanh\left[\frac{\hbar\omega_0}{2kT}\right]$$
(3)

where σ_0 and $\hbar\omega_0$ are constants. In alkali halides,^{4,6} $\hbar\omega_0$ fits the energy of the LO phonons over the whole temperature range, while in pure II-VI compound single crystals,⁷ which are also piezoelectric, $\hbar\omega_0$ fits the energy of the LA phonons in the lower temperature range (T < 100 K), and the energy of the LO phonons at higher temperatures.

In this paper we summarize the results of the measurements of the absorption coefficient near the fundamental absorption edge in two series of single crystals of two mixed II-VI compounds: $Zn_xCd_{1-x}Se$ and CdS_xSe_{1-x} crystals; and in two additional samples: ZnS_{0.7}Se_{0.3} and CdSe_{0.65}Te_{0.35}.

Intensive use is made of a previous paper⁸ reporting the preliminary results for single crystals of $Zn_r Cd_{1-r} Se_r$, where the Urbach rule and the mechanism of the thermal dependence of the absorption edge are discussed in detail.

II. THEORY

Two physical processes have been proposed as the source of the Urbach rule in pure single crystals. The older and more commonly used theory, developed by Toyozawa *et al.*, 9,10 ascribes the exponential shape of the absorption coefficient to the coexistence of free excitons and momentarily localized self-trapped excitons. In this self-trapped excitons (STE) model, the excitons are trapped by deformation-potential fluctuations due to short-range acting phonons. The degree of their localization by trapping is inversely proportional to the steepness index σ_0 .

The second theory, developed by Dow and Redfield (DR),¹¹ ascribes the exponential shape of the absorption coefficient to the influence of internal electric microfields on the electron transition during the absorption process, thus causing an internal Franz-Keldysh (FK) effect. The electric fields can be generated in several ways, such as by disorder (structural or compositional), impurities, or phonons. The main feature of the DR theory is the use of those internal fields in electroabsorption calculations, explaining the shift of the absorption edge as due to the LO-phonon-induced electrical fields.

Theoretical considerations have led us to prefer the DR theory when discussing the experimental results of the measurements in the $Zn_x Cd_{1-x}$ Se crystals. However, more recent measurements in single crystals of CdS_xSe_{1-x} have again raised the possibility of the usefulness of the STE model in the case of some mixed II-VI compounds as well.

It is proposed here that in crystalline materials one can use the σ_0 value of Eq. (3) as a sensitive indicator of internal disorder in the crystal, vibrational, structural, or compositional. To confirm this approach, one has to review some properties of the mixed single crystals of II-VI compounds. These can be classified into two broad categories according to the compositional dependence of their Raman and ir reflection spectra.

In the first class, known as the amalgamated type (one-mode), the Raman and ir reflection spectra are similar to that of a pure crystal, and the phonons energies vary continuously and linearly with composition between the two limiting values of the pure constituents. Some additional peaks appear in the Raman spectrum due to anharmonic couplings between phonons. Crystals such as $Zn_xCd_{1-x}Se$ (Ref. 12) and $Zn_xCd_{1-x}S$ (Ref. 13) belong to this class.

In the second class, known as the persistent type (twomode), a double spectrum appears, each spectrum corresponding to one of the pure constituents of the mixed crystal, with a slight dependence on composition. This double spectrum is observed in the Raman spectra as well as in the ir reflection spectra. Although the number of peaks due to anharmonic couplings between phonons is greater in this class than in the one-mode class, the main peaks are definitely related to each of the pure original crystals. Crystals such as CdS_xSe_{1-x} ,¹⁴ ZnS_xSe_{1-x} ,¹⁵ and CdS_xTe_{1-x} ,¹⁶ belong to this latter class.

III. EXPERIMENT

The experimental apparatus, the measurement procedures and the preparation of the $Zn_x Cd_{1-x}$ Se crystals are described in a recent paper.⁸ The CdS_xSe_{1-x} crystals were prepared from the melt. All of them had a wurtzite structure. The $ZnS_{0.7}Se_{0.3}$ crystal was prepared by the iodine vapor transport method. It had a zinc-blende structure. The $CdSe_{0.65}Te_{0.35}$ crystal was prepared by the modified Bridgeman technique and had a wurtzite structure. All the measured crystals were a few mm² in area, and their thicknesses varied between 80 and 250 μ m. At thicknesses lower than 200 μ m the CdS_xSe_{1-x} crystals were very fragile and tended to cleave in a direction perpendicular to their c axes.

The crystals of the series of CdS_xSe_{1-x} were of compositions x = 0, 0.01, 0.27, 0.35, 0.73, 0.9, 0.95, and 1.0.

The crystals were also investigated by x-ray luminescence in a scanning electron microscope (SEM) and by ir reflection in an ir photospectrometer.

IV. RESULTS

A. General

Crystals of both classes were measured: the $Zn_x Cd_{1-x}$ Se crystals of the amalgamated type, and the $CdS_x Se_{1-x}$, $ZnS_{0.7}Se_{0.3}$, and $CdSe_{0.65}Te_{0.35}$ crystals of the persistent type.

It was found out that the results of the measurements of the fundamental absorption edge also exhibit clear-cut differences, according to the crystal class. It should be emphasized that all the crystals, of both classes, had a definite and well-defined crystalline structure, as observed in x-ray measurements. Only at compositions of 0.4 < x < 0.6 does structural disorder appear in the $Zn_x Cd_{1-x}$ Se crystals due to lattice parameters misfits, and also because of the structural transition from wurtzite at the lower values of x to zinc-blende at higher



FIG. 1. $\ln(\alpha d)$ vs photon energy in $Zn_{0.3}Cd_{0.7}Se$ (amalgamated).

values of x. The disorder in these crystals appeared in the coexistence of domains of different structures in the same bulk, like in $Zn_{0.5}Cd_{0.5}Se$. In CdS_xSe_{1-x} crystals the compositional range of structural disorder is somewhat wider. As observed by x-ray luminescence in the SEM, the substitutional atoms are randomly distributed, also in the samples which exhibited a high degree of structural disorder. In samples of CdS_xSe_{1-x} with values of x = 0.27, 0.35, and 0.73, the structural disorder was expressed through the appearance of powderlike domains with no definite crystalline structure. These domains not only influenced the x-ray oscillation photographs, but were also clearly visible under an optical microscope.

The ir reflection measurements in $Zn_x Cd_{1-x}$ Se crystals also proved the existence of a continuous, almost linear variation of the optical phonon energies with composition. Results of similar measurements of CdS_xSe_{1-x} crystals are taken from literature.

The behavior of the mixed single crystals will now be discussed in detail. In all the crystals, of both classes, the absorption coefficient near the fundamental absorption edge depends on the photon energy (E) according to the Urbach rule. That is proven by the linear dependence of $\ln(\alpha d)$ versus E, as can be seen in Figs. 1 and 2. d is the crystal's thickness.



FIG. 2. $\ln(\alpha d)$ vs photon energy in CdS_{0.95}Se_{0.05} (persistent).



FIG. 3. σ vs temperature in Zn_{0.30}Cd_{0.70}Se (amalgamated).

B. Amalgamated-type crystals

In $Zn_x Cd_{1-x}$ Se crystals, the variation of σ with temperature is undoubtedly according to Eq. (3). An example is illustrated in Fig. 3, and all the other crystals of that series exhibited a similar temperature dependence of σ . At lower temperatures (T < 90 K) the value of $\hbar \omega_0$, which is calculated as a fitting parameter, fits the energy of the LA phonons, and in the higher temperature range, $\hbar\omega_0$ fits the energy of the LO phonons. Both values of $\hbar\omega_0$ (which correspond to the low and high temperature ranges) follow the compositional dependence of the LA and LO phonons energies as have been observed in the Raman spectra of those crystals. The σ_0 values in the LO phonon range varied between 2.3 and 2.7, which are similar to the values of σ_0 in the pure crystals.⁴ But when structural disorder appears as x gets closer to 0.5, for example in $Zn_{0.4}Cd_{0.6}Se$, the σ_0 value (for E1c polarization), decreases to 1.4. The influence of the increased disorder is apparent also through growing discrepancy between the experimental temperature dependence of σ and the theoretical values according to Eq. (3), as can be seen in Fig. 4.

On the other hand, the value of σ_0 for the LA phonon range, in all the $Zn_x Cd_{1-x}$ Se crystals, varies between 0.6 and 1.6, with no clear correlation with the crystal composition.



FIG. 4. σ vs temperature in Zn_{0.4}Cd_{0.6}Se (amalgamated).



FIG. 5. σ vs temperature in CdS_{0.90}Se_{0.10} (persistent).

C. Persistent-type crystals

In CdS_xSe_{1-x} crystals the temperature dependence in general also fitted Eq. (3), as can be seen in Figs. 5 and 6. The discrepancy between the experimental values of σ and Eq. (3) is more noticeable as x approaches the value of 0.5, and it increases even more strongly when structural disorder appears. On the whole, the fit of the experimental values of σ to Eq. (3) is better in the amalgamated-type crystals than in the persistent-type crystals.

The measured values of σ_0 , $\hbar\omega_0$, and E_0 of the persistent-type crystals are shown in Table I. (For the pertinent values for the Zn_xCd_{1-x} Se crystals, see Ref. 8.) In the CdS_xSe_{1-x} crystals, as well as in the $Zn_xCd_{1-x}Se$ crystals, at temperatures lower than approximately 90 K, the σ values behave according to Eq. (3), with $\hbar\omega_0$ which fits the energy of the LA phonons rather than the energy of the LO phonons. The value of

 σ_0 is about 1.0, and is not influenced by the crystal composition. However, when x approaches the value of 0.5, the temperature dependence of σ tends to become linear (see Fig. 7).

The value of $\hbar\omega_0$ in the higher temperature range fits the energy of an "effective" LO phonon, whose energy varies continuously, but not linearly, with composition,



FIG. 6. σ vs temperature in CdS_{0.73}Se_{0.27} (persistent).



FIG. 7. σ vs temperature in CdS_{0.27}Se_{0.73} (persistent) in the low-temperature range (LA phonons).



As can be seen from Table I, when x approaches 0.5, the value of σ_0 decreases for both polarizations. When structural disorder is observed, as in x = 0.27, 0.35, and 0.73, the decrease in the σ_0 values becomes even more noticeable.

Measurements of $ZnS_{0.7}Se_{0.3}$ and $CdSe_{0.65}Te_{0.35}$ also exhibit low values of σ_0 of 1.4–1.6 for the higher temperature range and significant deviations from Eq. (3), but definitely not towards a linear dependence on temperature.

D. The absorption edge

The energy of the absorption edge was measured at a high and constant value of $\alpha_g = 800 \text{ cm}^{-1}$. Its shift with



FIG. 8. E_0 and gap energy (at room temperature) vs composition in $Zn_x Cd_{1-x} Se$ (amalgamated).

temperature (in the higher temperature range) behaves as

$$E_{g}(T) = E_{g}(0) \coth\left[\frac{\hbar\omega_{0}}{2kT}\right] + E_{0}\left[1 - \coth\left[\frac{\hbar\omega_{0}}{2kT}\right]\right],$$
(4)

as was previously calculated in Ref. 8. $\hbar\omega_0$ is the energy of the LO phonons, which also appears in Eq. (3). $E_g(T)$ is the gap energy at $\alpha = \alpha_g$ at temperature *T*, and the constant energy E_0 is a constant of the Urbach rule [Eq. (1)].

The compositional dependence of the room-temperature $E_g(RT)$ absorption-edge energy is different in the two crystals classes: In the $Zn_x Cd_{1-x}$ Se crystals this dependence is linear for all the crystals with the wurtzite structure (x < 0.4) (Fig. 8), while in CdS_xSe_{1-x} this dependence is parabolic (Fig. 9) rather than linear.

TABLE I. The values of σ_0 and $\hbar\omega_0$ in the two temperature ranges, and E_0 vs composition in the persistent-type crystals. Str. denotes structure; Pol., polarization; W, wurzite; and Z, zinc-blende.

| | | | | Low temperatures | | | High temperatures | | |
|------------------------------------|------|------|-------------|-----------------------------|------------|-----------------------------|-------------------|------------|-------------------------|
| Crystal | x | Str. | Pol. | $\hbar\omega_0 ({\rm meV})$ | σ_0 | $\hbar\omega_0 ({\rm meV})$ | σ_0 | E_0 (eV) | $E_g(\mathbf{RT})$ (eV) |
| CdS _x Se _{1-x} | 0.00 | W | Eic | 7.0 | 1.05 | 27.0 | 2.30 | 1 895 | 1 727 |
| | 0.01 | W | Ellc | 10.0 | 1.20 | 27.0 | 2.20 | 1.976 | 1 804 |
| | 0.01 | W | Elc | | | 27.0 | 2.50 | 1 954 | 1.004 |
| | 0.27 | W | E c | 12.0 | 1.00 | 30.0 | 1.80 | 2 040 | 1.785 |
| | 0.27 | W | Elc | 12.0 | 0.95 | 30.0 | 1.80 | 2.051 | 1.870 |
| | 0.35 | W | E∥c | | | 31.0 | 1.30 | 2.097 | 1 999 |
| | 0.35 | W | Elc | | | 31.0 | 1.50 | 2.230 | 1.966 |
| | 0.73 | W | E∥c | | | 35.0 | 1.36 | 2.200 | 1.900 |
| | 0.73 | W | Elc | | | 35.0 | 1.51 | | |
| | 0.90 | W | Ellc | | | 37.0 | 1.74 | 2 534 | 2 345 |
| | 0.90 | W | E 1c | | | 37.0 | 1.80 | 2.537 | 2.345 |
| | 0.95 | W | Ellc | | | 37.5 | 1.74 | 2.613 | 2.52 |
| | 0.95 | W | Elc | | | 37.5 | 1.90 | 2.625 | 2.402 |
| | 1.00 | W | E c | | | 38.0 | 2.60 | 2.662 | 2.375 |
| | 1.00 | W | Eic | | | 38.0 | 2.80 | 2.650 | 2.402 |
| ZnS_xSe_{1-x} | 0.70 | Z | | | | | 1.4-1.5 | | |
| $CdSe_xTe_{1-x}$ | 0.65 | W | E∥c | | | 23.5 | 1.60 | 1.629 | 1.469 |
| | 0.65 | W | Elc | | | 23.5 | 2.00 | 1.593 | 1.454 |



FIG. 9. E_0 and gap energy (at room temperature) vs composition in CdS_xSe_{1-x} (persistent).

V. DISCUSSION

These results have to be discussed in terms of the influence of spatial disorder, as expressed through its influence on the electric microfields which determine the slope parameter S in the Urbach rule equation. As is known, the slope parameter is equal to¹¹

$$S = C/F_{\rm rms} \tag{5}$$

where C is a constant, and $F_{\rm rms}$ is the root mean square of the average square internal electric field, which dominates the absorption process. Using Eqs. (2) and (5), the temperature dependence of the σ parameter serves to indicate the source of those electric fields.

The results of the measurements in the amalgamatedtype crystals, as well as in the persistent-type crystals, indicate that the dominant electric fields in these crystals are generated by the piezoelectric LA phonons at lower temperatures, and by LO phonons at higher temperatures. This dependence of σ on the phonon energies in both crystal types leads to the conclusion that in both cases the dominant electric internal fields are *not* determined by structural or compositional potential fluctuations. If it were so, the dependence of σ on temperature would be linear, as in amorphous or polycrystalline materials.^{2,3} Those very same electric fields, induced by the phonons, also determine the thermal shift of the absorption-edge energy, as indicated by the experimental results, which fit Eq. (4).

However, the decrease of the value of σ_0 in the LO phonon range with increasing disorder suggests that the σ_0 value is a sensitive indicator for the localization of excitons due to electric fields, whatever their source might be. It should be emphasized that only the value of σ_0 which corresponds to the LO phonon range decreases with increasing disorder, while in the LA phonon range the influence is only on the form of the temperature dependence of σ and not on the σ_0 value.

In explaining the fundamental difference in the compositional dependence of the Raman and ir reflection spectra of the crystals of those two classes, it has been suggested previously¹⁷ that in the amalgamated-type crystals the two vibrational systems of the pure constituents of the mixed crystal merge into one system of strongly coupled oscillators, with one single averaged frequency for each phonon type. In the persistent-type crystals, on the other hand, each pure constituent preserves to a large extent its own vibrational spectrum, as the system acts as an ensemble of weakly, almost uncoupled oscillators.

This model of coupled oscillators can account also for the difference in the influence of composition on the temperature dependence of the σ parameter between the two classes.

In the amalgamated-type crystals all the LO phonons belong to a single vibrational frequency system, and most of them have similar frequencies. Therefore the electric fields at each crystal site are determined by phonons of similar frequencies, as in a pure single crystal. Hence, the exciton's degree of localization is like that in a pure crystal, i.e., relatively low, and thus its σ_0 value is in the same range of values as in the pure crystals.

On the other hand, in the persistent-type crystals, the electric fields at each site are determined by phonons of different frequencies, depending on the individual ions present in the close vicinity of that site. The uncoupling between their vibrations contributes also to the localization of the LO phonons in the persistent-type crystals, causing the dominant interactions to be with shortwavelength phonons. In the case of the amalgamatedtype crystals, the strong coupling enables the propagation of long-wavelength phonons through the crystal, as well as short-wavelength phonons. The contribution of two different localized phonon systems in the persistent-type crystals introduces an effect of spatial disorder in the electric field distribution. Consequently, in the persistent-type crystals the exciton is localized in a confined volume, whose dimensions decrease as the pure constituents equalize in concentration, i.e., as x approaches 0.5. This localization results in the decrease of the σ_0 value. When structural disorder is introduced, the dimensions of the confining volume decrease, and that results in a further decrease of the σ_0 value. In the amalgamated-type crystals, only structural disorder causes localization of the exciton, and therefore only at x = 0.4, where our samples are close to the region of structural transition, does a decrease in the σ_0 value occur.

It should be emphasized that the potential wells which localize the excitons in the mixed crystals differ in their causes and dimensions from those in the STE model. In the STE model these wells are created by the deformation potential of short-wavelength phonons, and so their dimensions do not much exceed the unit cell. Such potential wells are far too small and too weak to localize Wannier-type excitons, with large radii, which exist in the II-VI compound single crystals, pure or mixed. In the mixed crystals the potential wells are much wider, since they are correlated with the spatial distribution of the substitutional atoms, and they are created by the uncoupling between their vibrations (in the persistent-type crystals). In the temperature range which is influenced by the LA phonons, the influence of the changing composition is quite different.

As also observed in quantum-well structures^{18,19} of $GaAs/Al_xGa_{1-x}As$, the localization of the LO phonons does not influence their spectrum, while the LA phonons are almost destroyed by increased localization into a more confined volume. This explains the difference in the influence of the composition on the temperature dependence of σ in the two temperature ranges in the persistent-type crystals: In the higher temperature range the phonons are not destroyed by their localization, and therefore the average electric fields induced by them continue to increase with temperature, causing the fit of the experimental values of Eq. (3). The value of $\hbar\omega_0$ is that of an "effective" phonon because the exciton radius includes several unit cells, and so averages its vibrational frequency over a few crystal sites. However, the uncoupling between the oscillators causes the increasing discrepancy of the experimental results from Eq. (3). In the LA phonon range, the localization of the phonons almost destroys them, and the electric fields induced by structural disorder overcome the electric fields induced by the phonons. Consequently, the experimental values of σ tend towards a linear dependence on temperature, since the electric fields induced by structural or compositional disorder are constant and independent of temperature. An additional reason for observing the influence of the structural disorder preferentially over the electric fields of the LA phonons is that the electric fields of those phonons are far weaker than the fields induced by the LO phonons, since they are induced by the piezoelectric effect and not "directly" like the LO phonons. The influence of the structural disorder is of the same nature as the LA phonons, i.e., characterized by changes in density and the introduction of strains, accompanied by piezoelectric fields. In the amalgamated-type crystals no localization of phonons occurs, and so the experimental results fit Eq. (3) in all the compositions, at both temperature ranges.

The influence of the localization of the exciton on the value of σ_0 has been observed in the quantum-well structures²⁰ of GaAs/Ga_xAl_{1-x}As. Every decrease in the dimensions of those wells was followed by the widening of their excitons. This widening, which is temperature independent and is correlated with structural factors, is equivalent to the decrease in the value of σ_0 on the low-energy side of the exciton ($E < E_g$). The widening of the exciton causes the decrease of the slopes of the absorption coefficient over the whole temperature range, and consequently the decrease of the σ_0 value.

The use of the σ_0 value as an indicator for the degree of localization of the exciton can be viewed also in a wid-

er context: In alkali-halides,⁴ which are highly ionic, the values of σ_0 range between 0.75 and 1.0; in II-VI compounds (in the LO-phonon range) between 2.2 and 2.8; and in III-V compounds,²¹ σ_0 is about 4.0–4.5. (These values refer to pure single crystals.) As the crystal becomes more covalent and less ionic, its excitons have larger radia and they are less localized. As a result, their σ_0 values are higher.

VI. CONCLUSIONS

The absorption coefficient near the fundamental absorption edge is energy dependent according to the Urbach rule in mixed single crystals of II-VI compounds as well as in pure single crystals. The internal electric microfields which determine the slope parameter are induced by the LA phonons at lower temperatures, and by LO phonons at higher temperatures.

Essential differences in the absorption spectra of the crystals have been observed according to which class the crystal belongs, as in the case of the Raman and ir reflection spectra. These differences are explained by a model which describes the amalgamated-type crystals as an ensemble of coupled oscillators, while the persistenttype crystals are described as an ensemble of uncoupled oscillators. One of the results of this model is the introduction of localized-phonon-induced disorder even in crystals with well-defined crystalline structures and compositional homogeneity.

The experimental results are explained by the Dow-Redfield theory, with partial application of Toyozawa's theory of the self-trapped excitons, as being due to the influence of both spatial and vibrational disorder on the localization of excitons. As far as is known, this is the first time those two theories are applied in the same material. Until now, the classification of the excitons in the material as Frenkel- or Wannier-type excitons determined the preferred model: Toyozawa's self-trapped excitons model for the Frenkel-type excitons, and the Dow and Redfield model for Wannier-type excitons, in accordance with the basic assumptions of each model. We succeed in integrating both theories, thus illuminating the different aspects of our experiments.

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