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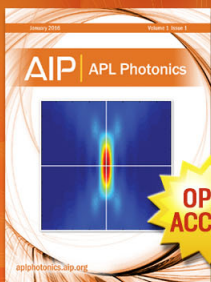
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Cd_{0.88}Zn_{0.12}Te group index measurements near the exciton energy at low temperature

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The group index of Cd_{0.88}Zn_{0.12}Te has been measured by a time-of-flight technique near the exciton energy at 10 K, in the energy range between 1.61 and 1.65 eV. Even at low temperatures, the standard model relying on a single excitonic contribution to the dielectric constant leads to erroneous optical constants for CdZnTe. By contrast, the experimental results are well fitted with a model taking all excitonic states into account. The parameters obtained by this fit are in excellent agreement with other experimental results and give the refractive index of Cd_{0.88}Zn_{0.12}Te at 10 K in the transparency region and close to the absorption threshold. © 1998 American Institute of Physics. [S0021-8979(98)01312-7]

INTRODUCTION

Ternary compounds of II–VI semiconductors are materials of considerable technological importance since they open the possibility for novel optoelectronic devices in the visible. The determination of the optical properties of Cd_{1-x}Zn_xTe, whose energy gap is direct, regardless of alloy composition, is therefore an asset for the design and operation of devices based on this compound.^{1–11} Recent work^{2,3} has been devoted to the determination by ellipsometry of the pseudodielectric function of CdZnTe in an extended energy range. A detailed description of the refractive index n and of the group index n_g of Cd_{1-x}Zn_xTe, in the direct vicinity of the fundamental exciton, would be a useful complement to these results. It is also well known that Coulomb effects are greater for II–VI materials than for III–V compounds.¹² Since n and n_g exhibit an enhanced dispersion near the absorption threshold, a strong variation of both indices for energies just below the fundamental exciton and large values of n_g are expected.

In this work, we have used a time-resolved photoluminescence setup to measure the group index of Cd_{0.88}Zn_{0.12}Te at 10 K, just below the exciton energy. We have used two models of the dielectric constant to fit the experimental data. The first—standard—model corresponds to the addition of a background term to a single excitonic line contribution. The second model uses a recently developed expression for the complex dielectric constant of Wannier excitons, which ensures that all excitonic effects are taken into account, and a background contribution as well. From the values of the different parameters obtained in the fits, we have calculated the refractive index n in the transparency region and close to the band gap.

The article is organized as follows. First, we describe the experimental setup and our measurements of the group index. Second, we present the two models used to fit the experimental data, and explicit the constraints imposed on the parameters used therein. We then detail the fitting procedures. We show that the widely used approximation for the dielectric function, consisting of a single excitonic contribution and a background, fails to deliver acceptable values for the exciton oscillator strength and the longitudinal-transverse splitting, and can lead to seriously incorrect values for the refractive index. On the contrary, the fit parameters provided by the Wannier model are consistent with other experimental data and provide the refractive index of Cd_{0.88}Zn_{0.12}Te at 10 K.

DESCRIPTION OF THE EXPERIMENTAL SETUP

The measurements of the group index have been performed on a Fabry–Perot microcavity sample, epitaxied on a high-quality Cd_{0.88}Zn_{0.12}Te substrate [see Figs. 1(a) and 1(b)]. This sample was initially designed for the study of the Rabi splitting in a II–VI microcavity. The Bragg reflectors consist in alternate quarter-wavelength layers of Cd_{0.4}Mg_{0.6}Te and Cd_{0.75}Mn_{0.25}Te: 17 and 15 periods are used for the back and front mirrors, respectively. Three 8.8-nm-thick CdTe quantum wells separated by 8-nm-thick barriers of Cd_{0.8}Mn_{0.2}Te are located at the center of the λ cavity, which is made of Cd_{0.8}Mn_{0.2}Te [see Fig. 1(b)]. Because of the thickness gradient of the cavity, the Fabry–Perot resonance, whose variation is about 4.6 meV for a 1 mm displacement of the excitation spot across the sample surface, exhibits an anticrossing with the excitonic energy of the CdTe quantum wells at 1.623 eV. The semiconductor structure, maintained at 10 K, is resonantly excited by the 1.5 ps pulses of a mode-locked titanium–sapphire laser tuned close to the anticrossing wavelength (typical spot size: 50 μ m).

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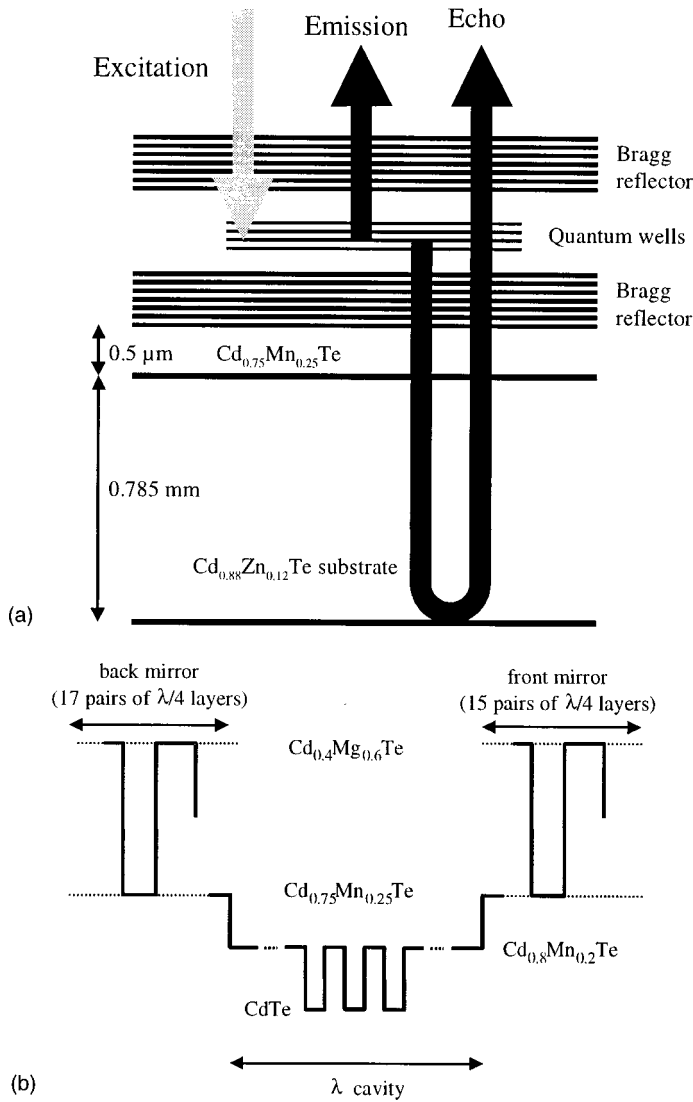


FIG. 1. (a) Schematic representation of the sample. After excitation, the emitted photoluminescence escapes the microcavity at once or undergoes a reflection on the back face of the $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ substrate. (b) Schematic representation of the microcavity structure.

The emitted photoluminescence signal is then collected and directed to a spectrometer coupled to a streak camera. When the excitation spot is moved along the direction of the thickness gradient of the microcavity, we observe an anticrossing of the luminescence energies indeed, as already seen in III–V microcavities.^{13,14} Contrary to what happens for most III–V samples grown on GaAs substrates, the $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ substrate used here is transparent for the emitted luminescence. Therefore, we not only observe a “direct” photoluminescence signal, but also the photoluminescence reflected by the back end of the substrate and transmitted by the microcavity [see Fig. 1(a)], which has a window at this wavelength. Photoluminescence decay curves are displayed in Fig. 2, which shows echoes appearing a few tens of picoseconds after the main peak. The delay Δt between the first peak and the echo is approximately given by

$$\Delta t = \frac{2W}{v_g} + \tau_c, \quad (1)$$

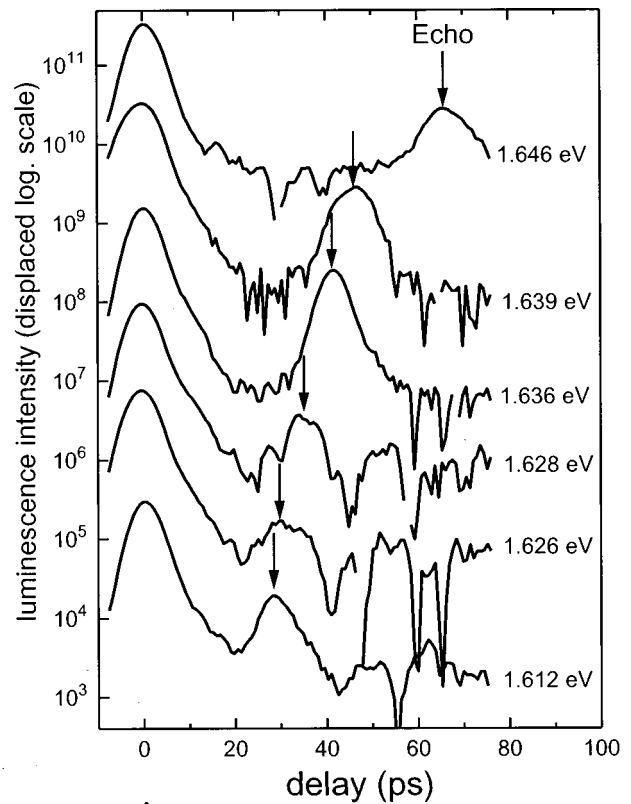


FIG. 2. Luminescence decay curves under resonant excitation at different positions on the sample. The corresponding energies are given at the right of each curve.

where W is the substrate thickness, $v_g = c/n_g$ the group velocity in the substrate and τ_c the photon lifetime in the cavity. Measurements of the substrate thickness using a digital length gauge gave $W = 0.785 \pm 0.006$ mm. From the width of the dip in the reflectivity spectrum of the Fabry–Pérot microcavity, we deduce $\tau_c \approx 0.6$ ps. One can safely neglect in Eq. (1) the contribution of τ_c to Δt , which is essentially the round-trip duration of light propagation in the substrate. Consequently, we have access to the group index value of $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ in an energy range very close to the fundamental exciton, whose energy $E_0 = 1.662$ eV, as determined by photoluminescence spectra. Such values would be extremely difficult to obtain from standard refractive index determinations, because of the onset of absorption in that spectral range. Let us now turn to the models used in this work to fit the experimental data.

THE DIELECTRIC CONSTANT MODELS

Let us recall that the group index n_g is given by

$$n_g(E) = n(E) + E \frac{dn}{dE} = \frac{d}{dE} (E \text{Re}\{\epsilon(E)^{1/2}\}). \quad (2)$$

We can thus obtain a description of the group index from a model of the dielectric constant. At low temperatures and in the vicinity of the fundamental exciton, the dielectric constant is usually taken as the contribution of the $n=1$ line, plus a constant background term. In the following, we will

also distinguish two contributions to the dielectric constant. The first one is caused by absorption processes near the absorption threshold, and the second one is a background term. Since we are interested in the group index in a region of strong dispersion, we cannot neglect the dispersion of the background term $\epsilon_{\text{background}}(E)$ anymore. We choose a single effective oscillator model, namely

$$\epsilon_{\text{background}}(E) = 1 + \frac{a}{b^2 - E^2}. \quad (3)$$

b is the energy of the effective oscillator, a rough average of the transition energies corresponding to the $E_1/E_1 + \Delta_1$ and E_2 peaks in the absorption spectra of the semiconductor. In the present study, we expect $b \approx 4$ eV from various experimental spectra²⁻⁷ or from simple fits of the refractive index in the transparency region.^{11,15-18} Note that $\epsilon_{\text{background}}(E)$ is purely real and that this simplified model works quite well as long as E is well below the strong absorption regions.

The usual description of the contribution to the dielectric constant of the $n=1$ exciton is given by an oscillator at the exciton energy E_0 , with an oscillator strength $4\pi\beta_0$ and a broadening γ . Consequently, the first of the two models is

$$\epsilon_{\text{model 1}}(E) = 1 + \frac{a}{b^2 - E^2} + 4\pi\beta_0 \frac{E_0^2}{E_0^2 - E^2 - iE\gamma}. \quad (4)$$

The calculation of the complex dielectric constant $\epsilon_{\text{Wannier}}(E)$ of Wannier excitons¹⁹ has shown that the excitonic continuum states are chiefly responsible for the dispersion of the optical properties near the absorption threshold. Coulomb effects are greater for II-VI materials than for III-V compounds.¹² Because the high frequency dielectric constants ϵ_∞ of CdTe or ZnTe are much smaller than that of GaAs, we expect the relative weight of $\epsilon_{\text{Wannier}}(E)$ and $\epsilon_{\text{background}}(E)$ near $E = E_g$ to be accordingly higher. Finally, the spin-orbit energy Δ_0 is about 0.93 eV for both CdTe and ZnTe, so that we may include the contribution of the $E_0 + \Delta_0$ threshold in $\epsilon_{\text{background}}(E)$, and write

$$\epsilon_{\text{model 2}}(E) = 1 + \frac{a}{b^2 - E^2} + \epsilon_{\text{Wannier}}(E), \quad (5)$$

$$\epsilon_{\text{Wannier}}(E) = \frac{A\sqrt{R}}{(E+i\Gamma)^2} [g[\xi(E+i\Gamma)] + g[\xi(-E-i\Gamma)] - 2g[\xi(0)]], \quad (6)$$

$$\xi(z) = \left(\frac{R}{E_g - z} \right)^{1/2}, \quad (7)$$

$$g(\xi) = 2 \ln(\xi) - 2\Psi(1 - \xi) - 1/\xi, \quad (8)$$

where A is a constant characteristic of the interband transition, R the exciton binding energy, E_g the band gap energy, Γ a broadening energy, and $\Psi(z) \equiv d \ln \Gamma(z)/dz$ the digamma function. Note that for the sake of simplicity, we have kept identical notations (a and b) for the background contribution in models 1 and 2. However, the values obtained from the two models are likely to differ since $\epsilon_{\text{Wannier}}(E=0)$ is not vanishingly small.

We then use the two preceding models to calculate the group index and evaluate the different parameters appearing in Eqs. (4)–(8). Before turning to the numerical results of the fits, we think it worthwhile to detail the procedures we have followed, and to precise the physical constraints placed on the fit parameters. In order to avoid unrealistic values for the parameters introduced in Eqs. (4)–(8), and because our group index measurements are limited to the 1.61–1.65 eV energy range, we will add a further constraint on these parameters: $\epsilon_{\text{model 1}}(E=0)$ and $\epsilon_{\text{model 2}}(E=0)$ must give ϵ_∞ , the value of ϵ for frequencies just above the reststrahlen region. We give in the next section an estimate of this high frequency dielectric constant at 10 K.

EVALUATION OF THE HIGH FREQUENCY DIELECTRIC CONSTANT AT LOW TEMPERATURES

To the best of our knowledge, there is no determination of ϵ_∞ for the ternary $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ at very low temperatures. Furthermore, there is a very large dispersion in the published data at room temperature, even for CdTe and ZnTe.²⁰⁻²³ We will therefore give in the following a detailed description of our estimation of the low-temperature value of ϵ_∞ . As noted by Bertolotti *et al.*,²⁴ its temperature dependence is determined mainly by contributions of transitions occurring at high energies (X and Λ transitions). $d\epsilon_\infty/dT$ is approximately given by²⁵

$$\frac{d\epsilon_\infty}{dT} \approx \frac{\epsilon_\infty - 1}{\bar{E}} \left(-\frac{d\bar{E}}{dT} \right), \quad (9)$$

where \bar{E} is an average energy—not very different from b —and $d\bar{E}/dT$ the corresponding temperature variation, assuming a rigid band shift of the absorption spectrum. Several authors consider that all critical points vary identically with temperature.^{10,24}

Since dn_∞/dT is equal to 1.0×10^{-4} and $0.85 \times 10^{-4} \text{ K}^{-1}$ for ZnTe and CdTe at room temperature, respectively,^{17,24} Eq. (9) leads to $d\bar{E}/dT \approx -3.5 \times 10^{-4} \text{ eV K}^{-1}$ for $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ at room temperature, a value slightly larger than Kim and Sivananthan's ($-3.0 \times 10^{-4} \text{ eV K}^{-1}$), but twice as large as Bertolotti *et al.*'s ($-1.7 \times 10^{-4} \text{ eV K}^{-1}$).^{10,24}

Basically, the variation of \bar{E} with temperature should follow that of the energy gaps. The latter are known to be well described by Varshni's law $E(T) = E(0) - \alpha T^2/(\beta + T)$. We can thus consider that

$$\Delta\epsilon_\infty(293 \text{ K} \rightarrow 10 \text{ K}) \approx -2n_\infty \left. \frac{dn_\infty}{dT} \right|_{300 \text{ K}} \Delta T_{\text{eff}}, \quad (10)$$

where ΔT_{eff} will be less than the actual temperature difference of 283 K. The room temperature ϵ_∞ is known with a relatively good accuracy for both CdTe (7.21 in Ref. 11, 7.18 in Ref. 17) and ZnTe (7.27 in Ref. 11, 7.28 in Ref. 17; see also Ref. 8). Taking $\Delta T_{\text{eff}} = 240 \text{ K}$ and $dn_\infty/dT \approx 0.98 \times 10^{-4} \text{ K}^{-1}$, we conclude that $\epsilon_\infty(\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}, 10 \text{ K}) = 7.08$. We will use this value in the following.²⁶

Let us briefly mention here our evaluation of dE_g/dT for $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$. From the binding energies^{27,28} of the ex-

citon in CdTe (10.5 meV) and ZnTe (13.2 meV) we deduce $R=10.8$ meV for the alloy. Adding the exciton energy $E_0 = 1.662$ eV leads to $E_g(10\text{ K}) = 1.6728$ eV. Castaing *et al.* give³ $E_g(293\text{ K}) = 1.5802$ eV for an alloy parameter $x = 0.12$. This 92.6 meV difference between room and low temperatures compares with the estimate of Ref. 16 for ZnTe. Keeping the same ΔT_{eff} , we find $dE_g/dT|_{300\text{ K}} \approx -4.0 \times 10^{-4} \text{ eV K}^{-1}$, in agreement with some determinations of this constant.

Let us now detail the constraints that must be satisfied by the fit parameters.

CONSTRAINTS ON THE FITTING PARAMETERS

Our fitting procedures are as follows. In model 1, we leave b , $4\pi\beta_0$, and γ as free parameters, E_0 being fixed at its experimental value of 1.662 eV, and a such that $\epsilon_{\text{model 1}}(E=0) = \epsilon_\infty$. Since the excitonic oscillator strength is very small, $a \approx (\epsilon_\infty - 1)b^2$. In the case of model 2, b is fixed at a reasonable value slightly above 4 eV corresponding to an average of critical point energies where the absorption spectra exhibits strong peaks, and a is such that $\epsilon_{\text{model 2}}(E=0) = \epsilon_\infty$. A and Γ are the only free parameters.

One must keep in mind that all the above parameters cannot be arbitrary: they must give realistic values for other optical constants which, while not measured in this work, are tabulated in the literature. Therefore,

- (1) γ and Γ must be compared to experimental values of the broadening of a few meV.^{1,29}
- (2) $4\pi\beta_0$ is related to the longitudinal-transverse splitting Δ_{LT} by

$$\Delta_{\text{LT}} = 4\pi\beta_0 \frac{E_0}{2\bar{\epsilon}}, \quad (11)$$

where $\bar{\epsilon}$ is the value for $E=E_0$ of the dielectric constant in which the contribution of the $n=1$ exciton has been removed. With the parameters of model 2, one gets

$$\Delta_{\text{LT}} = \frac{4AR^{3/2}}{\bar{\epsilon}E_0^2}. \quad (12)$$

Since Δ_{LT} has been determined experimentally by resonant Brillouin scattering—0.65 and 0.7 meV for CdTe and ZnTe, respectively^{30,31}—Eqs. (11) and (12) are a strong test for the values of A and $4\pi\beta_0$ obtained in our fits.

- (3) The numerical value of A can be further checked since¹⁹

$$A \propto \mu^{3/2} P^2, \quad (13)$$

where μ is the reduced exciton mass and P^2 the Kane factor. P^2 is known to be smaller for II–VI than for III–V semiconductors¹²—roughly 17 eV instead of 22.7 eV for GaAs. Since $A = 2.6 (\text{eV})^{3/2}$ for GaAs,³² Eq. (13) provides a straightforward estimate of its $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ value: $P^2 = 17 \text{ eV}$ ¹² and a linearization of the effective masses in the alloy parameter lead to $A_{\text{CdZnTe}} = 3.55 (\text{eV})^{3/2}$. Alternatively, the imaginary part of the dielec-

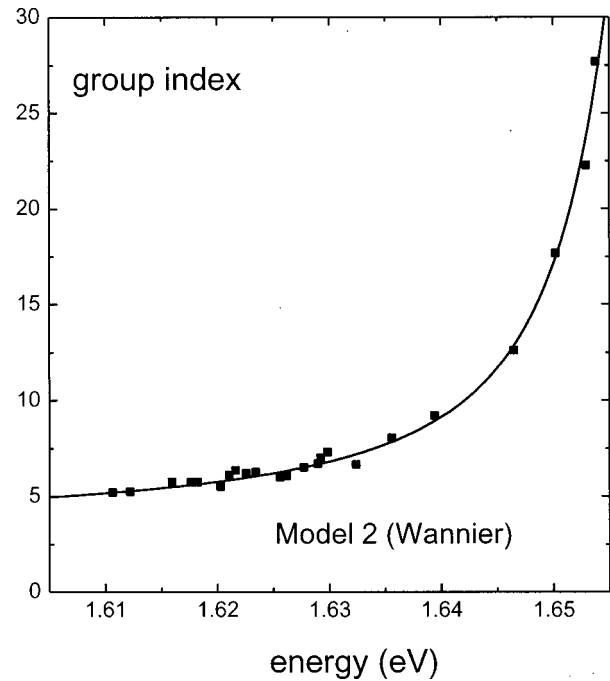


FIG. 3. Fit of the experimental values of the group index using model 2, for $\epsilon_\infty = 7.08$. The parameters deduced from the fit are: $a = 92.42 (\text{eV})^2$, $b = 4.1 \text{ eV}$, $\Gamma = 2.3 \text{ meV}$, and $A = 3.63 (\text{eV})^{3/2}$.

tric constant exhibits at the absorption threshold^{2,3} a quasi step function—caused by the Sommerfeld enhancement factor—whose magnitude is

$$\text{Im } \epsilon(E=E_g) \approx \frac{2\pi A \sqrt{R}}{E_g^2}. \quad (14)$$

- (4) Even though the fit parameters are determined from our values of the group index, one should obtain acceptable values of the refractive index near the gap. In the transparency region, these values should be lower at 10 K than at room temperature.

RESULTS OF THE FITS AND DISCUSSION

Our measurements of n_g have been performed in a narrow energy range. It is therefore not too surprising that both models fit the experimental data very well. However, by following the guidelines given in a preceding section, we will show that model 1 leads to unrealistic values of the optical parameters, whereas model 2 agrees quite well with independent measurements.

In the case of model 1, a very good agreement with our values of n_g can be reached with $4\pi\beta_0 = 9.61 \times 10^{-3}$, $b = 2.73 \text{ eV}$, $a = 45.1 (\text{eV})^2$, and $\gamma = 5.2 \text{ meV}$. However, these values are not satisfying since b is too small by at least one electron volt: No effective oscillator can be found at this energy.^{2–6} Likewise, the fitted oscillator strength is too large by approximately 50%. From the longitudinal-transverse splitting values of CdTe and ZnTe already mentioned, and for $\bar{\epsilon} \approx 9$, one would expect $4\pi\beta_0 = 6.7 \times 10^{-3}$.

Figure 3 shows the fit of the experimental data using model 2, for $b = 4.1 \text{ eV}$, $A = 3.63 (\text{eV})^{3/2}$, $a = 92.42 (\text{eV})^2$, and $\Gamma = 2.3 \text{ meV}$. This value of A is in excellent agreement

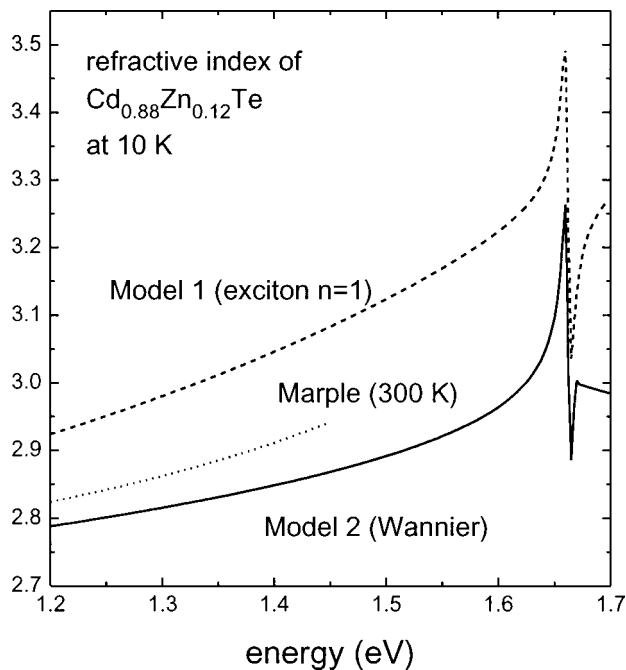


FIG. 4. Values of the refractive index of $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ deduced from the fit parameters of model 1 (dashed line) and model 2 (solid line). A room-temperature estimate based on a linear regression with respect to the alloy parameter of Marple's data for CdTe and ZnTe is also displayed (dotted line). Model 1 must therefore be rejected.

with our estimate of $3.55 \text{ (eV)}^{3/2}$ using Eq. (13).³³ From these values and Eq. (12), we get $\Delta_{\text{LT}} = 0.656 \text{ meV}$, close to the values for CdTe and ZnTe given in the preceding section. From Eq. (14) we also find $\epsilon_2(E = E_g) = 0.847$, in agreement with experimental spectra of the imaginary part of the dielectric constant.²⁻⁶ This is all the more satisfying as absorption experiments are quite different from group index measurements. The value obtained for Γ compares quite well with results given in the literature for CdZnTe alloys.¹

A final comparison between the two models can be made by plotting the value of the refractive index for $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ at 10 K, as deduced from the fit parameters. Figure 4 shows the two corresponding curves, along with a curve representing $0.88n_{\text{CdTe}} + 0.12n_{\text{ZnTe}}$ deduced from Marple's data at room temperature.¹¹ Obviously, the refractive index should be smaller at 10 K than at room temperature. However, this is not the case for the curve given by model 1, which should be definitely discarded. This shows unambiguously that although the fit of the group index with model 1 may be quite good in a very narrow spectral range, the associated fit parameters cannot reproduce the refractive index correctly.

CONCLUSION

We have performed group index measurements for $\text{Cd}_{0.88}\text{Zn}_{0.12}\text{Te}$ at 10 K, very close to the exciton energy. The experimental data have been fitted by two models. The simple model consisting of a single exciton line and a background dielectric constant presenting dispersion can fit the group index values in a limited spectral range, but it leads to erroneous values of optical constants such as the refractive

index or the excitonic oscillator strength. We think that further use of such a model in the vicinity of the excitonic line may be quite hazardous, even in the case of semiconductors other than II-VI compounds, for which the Coulomb effects on the optical properties are enhanced near the absorption threshold.

The important result presented in this work is that a model of the dielectric constant based on the complex dielectric constant of Wannier excitons describes both group index and refractive index quite satisfactorily, and agrees quite well with independent determinations of the absorption at threshold, of the longitudinal-transverse splitting, etc. We have thus been able to give the low-temperature value of the refractive index from group index measurements performed in a limited spectral range near the band gap.

These conclusions should hold all the more at room temperature, since the $n = 1$ exciton effects are then smeared out. Such a consideration must be kept in mind when one evaluates the group index of a material near the absorption threshold, an important parameter in the design and operation of optoelectronic devices such as lasers or optical waveguides.

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