Deep center luminescence in *p*-type CdTe

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The deep-level photoluminescence (PL) emission in the 1.1 eV spectral region in *p*-type CdTe:Cl polycrystalline samples was studied as a function of the excitation laser power and temperature. The relatively broad 1.1 eV PL band has a nonsymmetrical shape which can be easily changed by varying the excitation laser power. Detailed analysis of the line shape shows that the 1.1 eV emission contains two distinct separate bands. These two bands have their zero-phonon peaks located at 1.08 and 1.17 eV, respectively, and they have quite different half-widths. A donor–acceptor (DA) pair model with a deep donor and a deep acceptor with E_D and E_A both >0.5 eV is proposed to explain the observed experimental findings. In this model the 1.08 and 1.17 eV bands are formed as a DA recombination between pairs of the nearest neighbors, and between pairs of the next-nearest neighbors, respectively. It is concluded that the acceptor in these pairs must be an interstitial atom. One possible realization for this kind of a DA pair is the $V_{\rm Te}$ -Te_i complex, where $V_{\rm Te}$ is acting as a donor and Te_i as an acceptor. © 1996 American Institute of Physics. [S0021-8979(96)11215-9]

I. INTRODUCTION

The increasing interest in the study of the *p*-type CdTe is mainly related to its technological applications in solar cells. A large variety of methods has been used to make CdTe/ CdS-based photovoltaic devices with surprisingly good results. Despite these results, the defect structure and the physical processes giving rise to a high *p*-type conductivity in CdTe are still obscure.

Photoluminescence (PL) spectroscopy is a widely used method to analyze the defect structure of semiconductors. In CdTe the majority of published PL results is focused to the exciton and edge-emission spectral regions. It is obvious that in order to get a complete survey about the defect structure, a deep level PL study is inescapable. The deep levels seem to have an essential and interesting role in the electrical properties of CdTe. There is a serious suspicion that deep levels in CdTe have also an effect on the compensation mechanism.

There are four groups of deep PL bands generally found in CdTe and located at about 0.5, 0.8, 1.1, and 1.4 eV, respectively, each group containing at least two bands.^{1–7} According to Hofmann *et al.*⁸ there is clear evidence that the 1.4 eV group of PL bands may contain more than three independent emission bands. In many cases these bands may all be present causing considerable confusion. Therefore, in any study, it is extremely important first to distinguish between separate bands under study. In this article we investigate experimentally the 1.1 eV group of PL bands in CdTe.

The nature of the 1.1 eV emission in CdTe has been interpreted in various ways. Although the role of residual impurities has not been completely ruled out (see, for example, Lischka, Brunnthaler, and Jantsch,⁹ where the emission in the 1.1 eV spectral region is associated with an iron dopant), the prevailing general opinion is that only intrinsic defects are involved in this emission of undoped or chlorine doped CdTe. In the earliest article only one band was presumed to be present in the 1.1 eV region and therefore some results and conclusions were apparently erroneous.

Bryant and Webster² measured the temperature quenching of the 1.1 eV band and the activation energy value E_T =0.17 eV was found. Radiation damage experiments in CdTe verified the connection between the 1.1 eV PL band and the displacement of tellurium.^{1,2} Proposed defects were V_{Te} or Te_i and the recombination was assumed to be *c*-*a* type. The same origin for this band was also assumed by Sobiesierski, Dharmadasa, and Williams.¹⁰

Davis *et al.*¹¹ employed the photothermal deflection spectroscopy (PTDS) and a defect absorption near 1.02 eV at room temperature was detected. This absorption was assumed to be related to the 1.1 eV PL band. The well-defined absorption peak found indicated that the transition must be between two localized states.

Valdna, Buchmann, and Mellikov¹² discussed also the connection between PL emission in the 1.1 eV region and good p-type conductivity in CdTe:Cl.

As pointed out by Panossian,⁴ there exist two PL bands in the 1.1 eV region at 77 K with peak positions of 1.0 and 1.11 eV, respectively. The intensities of these bands had a different dependence upon the excitation intensity.

In the latest article by Stadler *et al.*¹³ PL bands in $Cd_{1-x}Zn_xTe$ were studied. In doped CdTe two PL bands with peak positions of 1.135 and 1.145 eV were detected at 2 K. The peak position of the 1.145 eV band shifts with *x* and follows the band-gap dependence in $Cd_{1-x}Zn_xTe$, while the 1.135 eV band peak position remains constant over the complete alloy range. According to Stadler *et al.* this behavior is well explained with an internal recombination model.

Despite intensive research, the nature of the 1.1 eV PL band in CdTe is still not resolved and therefore new experimental work is needed.

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II. EXPERIMENT

The samples were prepared by the isothermal sintering near 500 °C of stoichiometric CdTe powder on Al_2O_3 substrates together with 2 wt % CdCl₂ flux inside evacuated quartz tubes. Although the starting material was not doped with any acceptor dopant, its PL spectrum exhibits several peaks in the edge-emission region and the so-called 1.4 eV PL band with the zero-phonon line at 1.450 eV. According to Hofmann *et al.*⁸ this band originates from the donor–acceptor recombination with a Cl donor and a Cu acceptor. It is well known that so-called undoped CdTe can contain impurities in a concentration range up to 10^{17} cm⁻³ and the most probable impurity is copper.¹³

After being first prepared in the aforementioned way, samples passed through three additional technological stages. The first stage was a washing with distilled water in order to remove $CdCl_2$ from the surface of the samples. PL spectra of these materials showed peaks at 1.589 and 1.585 eV in the excitonic spectral region and a chlorine related 1.4 eV band with a zero-phonon line at 1.475 eV. The 1.585 eV peak is known to be typical of highly chlorine-doped CdTe.¹⁴

The second stage was a long-term vacuum annealing to remove excess CdCl₂ and possibly also some chlorine. Finally, a thermal treatment under Te vapor pressure, followed by a slow cooling, was undertaken. Prepared by this way the samples had a good *p*-type conductivity with $p=1.7\times10^{16}$ cm⁻³ and mobility $\mu=81$ cm²/(V s), as proved by Hall measurements.

For the PL measurements, a 5 mW He–Ne laser beam with a wavelength of 632.8 nm was used for excitation. The samples were mounted in a closed-cycle He cryostat capable of cooling down to 12 K. A computer-controlled SPEX 1870 grating monochromator (0.5 m) with a spectral slit of 0.5 nm was used. The chopped signal was detected with a liquid-nitrogen (LN₂)-cooled Ge detector using the conventional lock-in technique. For the purpose of analysis, the emission spectra were corrected for grating efficiency variations and for the spectral response of the detector, which were calibrated separately.

III. RESULTS AND DISCUSSION

A typical PL spectrum of p-type CdTe:Cl at 13 K is shown in Fig. 1. All measured *p*-type samples exhibited very weak emission in the excitonic spectral region. A wellknown (A^0, X) peak at 1.589 eV is clearly discernible. An additional peak at 1.573 eV is also present. The energetic distance between these peaks is 16 meV. The 1.573 eV peak is seen only in materials having a strong emission in the 1.1 eV spectral region. Bryant, Totterdell, and Hagston¹⁵ assign this peak to a transition of an electron in the conduction band to the ground state of a neutral double acceptor center, probably the cadmium vacancy. The same peak was observed also by Giles-Taylor et al.¹⁶ The emission, peaked at 1.576 eV at 4.2 K in high-purity CdTe, was studied by Espinosa et al.¹⁷ This peak had a shape typical of free-to-bound recombination, and it was supposed that an acceptor level at 32 meV above the valence band is associated with the 1.576 eV PL peak. It is known that the effective-mass acceptor limit in



FIG. 1. Photoluminescence spectrum of p-type CdTe measured at 13 K.

CdTe is 56.8 meV.¹⁸ These shallow acceptor states below the effective mass limit in CdTe:Cl are believed to be due to a V_{Cd} (Cl_{Te})₂ defect.^{19,20}

In the PL-intensity spectrum of Fig. 1 there is also an emission band around the 1.4 eV spectral region with a wellresolved phonon structure. The detailed shape of this band depends on the sample preparation conditions. As was pointed out by Hofmann et al.⁸ and by Kuhn et al.²⁰ there may exist at least three different bands in the 1.4 eV spectral region. Two of them are detected regularly. The first band has a zero-phonon line near 1.450 eV and it is believed that this band is caused by donor-acceptor emission between a Cu_{Cd} acceptor and a Cl_{Te} donor. This band was clearly detected in CdTe, doped with Cu and Cl.²¹ The second band is believed to be due to a donor-acceptor emission between a Cl_{Te} donor and the so-called A center as an acceptor. In CdTe:Cl this A center has a structure $(V_{Cd}Cl_{Te})$. This band has a zero-phonon line near 1.478 eV. There is also a third band in this region of unknown origin and with quite a strange shape, but this band has been detected only in few samples.^{8,20} In our *p*-type samples the 1.450 eV band prevailed, but in some of the samples the 1.478 eV band was also visible and thus the shape of the 1.4 eV band in these samples was confused.

In all of our samples the broad band near 1.1 eV was present. In good *p*-type samples the intensity ratio I(1.1 eV)/I(1.4 eV) was greater than 1. As the relative intensity of the 1.4 eV PL band decreases in good *p*-type materials and these materials were slowly cooled after thermal treatments, it is natural to believe that during cooling process more tight complexes were formed in CdTe. Therefore, the concentration of $V_{\text{Cd}}\text{Cl}_{\text{Te}}$ defects decrease in favor of forming new complexes $V_{\text{Cd}}(\text{Cl}_{\text{Te}})_2$. Apparently, this process results in a decrease of the 1.4 eV PL band intensity and a new PL band at 1.573 eV appears.

The 1.1 eV band has a nonsymmetrical shape. This shape can be easily changed by varying the excitation laser power; see Fig. 2. Thus, it is obvious that there are two separate bands in this spectral region. If the minimum laser power is used, only one single band with peak position at 1.12 eV can be detected. The shape of each of these bands is also nonsymmetric and well represented as a sum of several



FIG. 2. Normalized PL spectra of CdTe in the 1.1 eV energy region, measured with varying excitation laser power. At low excitation only the 1.17 eV band is visible.

individual Gaussians, separated by the LO-phonon energy 21.2 meV.¹ The intensity distribution of these individual Gaussians can be extremely well described by a Poisson distribution,

$$I(n) = I_0 e^{-S} \frac{S^n}{n!},$$
(1)

with the Huang–Rhys coupling parameter *S* and a constant factor I_0 . For the best fit of Eq. (1) to the experimental points the width of the individual Gaussian components of each band must be assumed to be quite large, of the order of 130 meV for the lower-energy band and of the order of 40 meV for the higher-energy band. Due to this the magnitude of *S* can be deduced only approximately, being $S \approx 1.5$ for the lower-energy band and $S \approx 2.2$ for the higher-energy band. Figure 3 shows a typical result of this fitting. As can be seen also from Fig. 3, the two bands have quite different halfwidths and their zero-phonon peaks are located at 1.08 and 1.17 eV, respectively.

Figure 4 shows the temperature dependence of the zerophonon peak position $E_0(T)$ for both bands. It is obvious



FIG. 4. Temperature dependence of the zero-phonon lines positions $E_0(T)$ for the two deep PL bands, obtained from a fit to Eq. (1), and of $E_g(T)$ taken from Ref. 21.

that at low temperatures the zero-phonon peak positions remain constant and only at higher temperatures (T>110 K) a small decline can be detected.

The dependence of the emission intensity on the laser power is depicted in Fig. 5. It is clearly visible that the intensity of these two bands behaves quite differently as a function of the PL-excitation power. The dependence of the integrated intensity Φ of the PL bands on laser power I_{laser} can be represented as $\Phi \sim I_{\text{laser}}^{\alpha}$. The 1.08 eV PL band has a nearly linear dependence (α =1.09) while the 1.17 eV PL band has a much slower dependence (α =0.66) on I_{laser} . This rather big difference indicates that these two bands must have somehow different origins.

The temperature quenching of both of these PL bands is illustrated in Fig. 6. The experimental dependence of the intensity, as a function of temperature, for each band was fitted to the theoretical expression

$$\Phi = \frac{\Phi_0}{1 + \alpha_1 \exp(-E_1/kT) + \alpha_2 \exp(-E_2/kT)}.$$
 (2)

This corresponds to the presence of two thermally activated nonradiative recombination mechanisms, and is often found



FIG. 3. A typical result of fitting the PL emission intensity with Eq. (1).



FIG. 5. The dependence of the luminescence intensity on laser power for the two deep PL bands in CdTe.



FIG. 6. Temperature dependence of the PL intensity for the 1.08 and 1.17 eV bands. The fits using Eq. (2) are shown as continuous curves.

to excellently reproduce the luminescence intensity over a wide temperature range.²² Here Φ is the integral intensity of the band, $\alpha_{1,2}$ are constants, and $E_{1,2}$ are the thermal quenching activation energies of the low- and high-temperature quenching processes, respectively. The best fitting parameters are presented in Table I, where the error intervals correspond to the 95% statistical confidence limits.

The thermal activation energy $E_{T_2}=174$ meV for the 1.08 eV band is the same as measured by Bryant and Webster.² The low-temperature quenching with an extremely low value of activation energy, $E_{T_1}=5.7$ meV or $E_{T_1}=4.0$ meV, and of the pre-exponential factor, $\alpha_1=3.6$ or 1.5, is a typical feature for both bands. In Ref. 13 this low-temperature quenching was assumed to be due to the ionization of shallow donor levels and two quenching stages probably point toward a donor-acceptor recombination; but, the fact that two bands of different origin have practically the same low-temperature quenching activation energy probably indicates that there must be an additional mechanism for the low-temperature quenching of these bands. It is interesting that nearly the same values α_1 and E_{T_1} were obtained also for the 1.4 eV PL band low-temperature quenching.¹³

A traditional method of electron-phonon interaction analysis in solids is to compare the experimentally measured dependence of luminescence band half-width W on temperature T with the theoretical one, calculated from²³

$$W(T) = W(0) \left(\coth \frac{\hbar \omega}{2kT} \right)^{1/2} + \Delta W.$$
(3)

Here the $\hbar\omega$ is an effective phonon energy for the excited state of the defect luminescence center. In the case of low

TABLE I. Thermal quenching parameters for the 1.08 and the 1.17 eV PL bands, obtained by fitting to Eq. (2).

Parameter	1.08 eV band	1.17 eV band
Φ_0 (arb. units)	71	122
$E_1 \text{ (meV)}$	4.0 ± 2.5	5.7 ± 2.5
E_2 (meV)	174±7	113±5
α_1	1.54	3.56
α_2	2.0×10^{8}	2.2×10^{6}



FIG. 7. Temperature dependence of the half-widths of the two PL bands, found experimentally. Theoretical fits using Eq. (3) are shown as continuous curves.

defect concentration the additional term $\Delta W \cong 0$, but in "heavily doped" materials this term may have a greater value.

The broadening of each LO-phonon component line into a Gaussian-shaped band and the observed temperature dependence of W (see Fig. 7) is most naturally interpreted by an additional interaction with acoustic phonons. The fitting parameters obtained using Eq. (3) are presented in Table II.

From the results of the fit (see Table II) it should be obvious that the broadening of individual subbands is caused by interaction with acoustic phonons. The additional broadening ΔW of the 1.08 eV band is not temperature dependent and is probably due to potential fluctuations of charged defects near the recombination center. The role of these fluctuations on the zero-phonon line width in CdS:Ag:Cl has been shown in Ref. 24. It is a curious experimental finding of the present work that, according to Fig. 7, only the 1.08 eV PL band appears to be affected by these fluctuations.

As it was mentioned before, deep PL bands in CdTe commonly seem to exist as pairs. Therefore it is reasonable to assume that these pairs are caused by a recombination via a luminescence center having the same chemical nature but a slightly different physical structure. One possible reason causing such kind of pairs is a center involving donor–acceptor (DA) pairs with a deep donor and a deep acceptor. For these DA pairs, luminescence emissions can be observed at closest separation only, because of localization of the electron and the hole wave functions in these deep levels. In CdTe there are several possibilities:

- (a) an acceptor at Cd site and a donor at Te site;
- (b) an acceptor at Cd site and a donor at Cd site;

TABLE II. Parameters of electron-phonon interaction for the 1.08 and the 1.17 eV PL bands obtained from fitting to Eq. (3).

Band	1.08 eV	1.17 eV
<i>W</i> (0) (meV)	69	45
$\hbar\omega$ (meV)	8.3	6.5
$\Delta W \;({\rm meV})$	84	0

- (c) an acceptor at Cd site and a donor as an interstitial;
- (d) a donor at Te site and an acceptor as an interstitial.

Taking the lattice parameter in CdTe as a = 6.482 Å,¹ and the dielectric constant $\epsilon(\infty) = 7.1$,¹ it is possible to calculate the energy separations between pairs of the nearest or the nextnearest neighbors: $\Delta E = e^2/(\epsilon r_1) - e^2/(\epsilon r_2)$. This is simply the difference in Coulomb interaction, for different pair separations r_1 and r_2 , with a medium of dielectric constant ϵ .²⁵ For case (a) we get $\Delta E = 346$ meV, for (b) we get $\Delta E = 130$ meV, and for (c) and (d) we get $\Delta E = 96$ meV. It is obvious that case (a) does not coincide with the experimentally observed energy separation between the 1.08 and 1.17 eV PL bands, $\Delta E = 90$ meV. Second observation of importance is the fact that the half-width of the 1.08 eV band is much bigger than that of the 1.17 eV band.

A possible reason for this difference in the half-width may be a different surrounding of DA pairs in the first and second coordination spheres. In the case (b) both DA pairs have the same surrounding but in the case (c) and (d) there are differences. These differences are due to a different symmetry of interstitial positions in CdTe. In the case of tetrahedral symmetry each interstitial site is surrounded by four Te atoms and in the case of octahedral symmetry by six Te atoms. The closest interstitial for an acceptor at Cd site has an octahedral symmetry and the second closest interstitial has a tetrahedral symmetry. The closest interstitial for a donor at Te site has a tetrahedral symmetry and the second closest interstitial has an octahedral symmetry. We believe that these differences cause also the difference in the halfwidth of the two PL bands and the different dependence of the integrated intensity on the laser excitation power. It is interesting that no other deep PL bands were observed in our materials, which means that the critical distance between a deep donor and a deep acceptor to form an optically emitting DA pair must be $r \cong 3-4$ Å.

A possible energy level diagram for the deep-level PL bands investigated in the present work is presented in Fig. 8. In this model the PL bands in the 1.1 eV spectral region are associated with DA pairs with $E_D(\infty)$ and $E_A(\infty)$ both >0.5 eV.

Deep levels with E > 0.5 eV in CdTe were found by several groups,^{26–29} but the chemical nature of the corresponding defects remains, so far, undetermined.

Our materials were prepared under vapor pressure of tellurium. Therefore, the majority of defects are probably formed in the Cd sublattice. The dominating defect in this region is believed to be V_{Cd} . We may also assume that, in the first approximation, an interstitial atom is affected only by nearest neighbors, so the fluctuations of the defect concentration near the interstitial atom are the reason of the significant widening (see Table II) of the 1.08 eV PL band. According to the symmetry of the interstitial positions only octahedral interstitial positions have Cd atoms as nearest neighbors. This is why we must assume that the closest DA pair is formed with the interstitial atom in a surrounding of tetrahedral symmetry and the second closest DA pair with the interstitial atom having an octahedrally symmetric surroundings, i.e., case d) above. One possible model for this



FIG. 8. The suggested energy-level model for the deep PL bands in *p*-type CdTe. Here *m* is the configuration index, i.e., m=1 for the closest lying DA pair neighbors, m=2 for the next-closest neighbors, etc.

kind of DA pair is the V_{Te} -Te_{*i*} complex, where V_{Te} is acting as a donor and Te_{*i*} as an acceptor.

IV. CONCLUSIONS

We have measured the deep-level luminescence in the 1.1 eV spectral region in p-type CdTe:Cl polycrystalline samples. The detailed line-shape analysis of the measured PL spectra reveals that the 1.1 eV emission is composed of two PL bands with their zero-phonon positions at 1.08 and 1.17 eV, respectively. The intensity and the half-width of these two bands behave quite differently as a function of the PL-excitation power and temperature.

We conclude that these two bands are caused by a recombination within a luminescence center having the same chemical nature but a slightly different physical structure. One possible reason causing such kind of bands is a center involving a donor-acceptor pair with a deep donor and a deep acceptor. Because of the localization of the electron and the hole wave functions in these deep levels, the photoluminescence due to the recombination between the DA pairs of the closest separation only is detected. In order to explain the differences in the experimental properties of both bands, we assume that the corresponding DA pairs have different surroundings in the first and second coordination spheres. This is possible, if a donor or an acceptor defect is situated at an interstitial position, because of the different symmetry of interstitials in the CdTe lattice. Taking into the consideration the chemical conditions of the sample preparations and all the experimental data now obtained, we concluded that the 1.08 and the 1.17 eV PL bands are caused by DA pairs, where the donor defect is at a Te site and the acceptor defect is at an interstitial position having surroundings of either octahedral symmetry or of tetrahedral symmetry, respectively. One possibility to construct this kind of a DA pair is the V_{Te} -Te_i complex.

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