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## Compensation of CdTe by Doping With Gallium

Semi-insulating CdTe single crystals doped with Ga were grown from the vapour phase by the modified Markov technique MMT. The study of the resistivity map in the cross-sections cut along the growth direction has been performed. The compensation phenomenon is analysed in the framework of the three levels Fermi-statistic model. It is shown that a semi-insulation behaviour throughout the ingot is due to the compensation of shallow impurities by the deep level. From the low-temperature photoluminescence spectra it was concluded that shallow donors ( $\text{Ga}_{\text{Cd}}$ ) are partly compensated by ( $\text{Ga}_{\text{Cd}}\text{-V}_{\text{Cd}}$ ) and ( $\text{Ga}_{\text{Cd}}\text{-Cd}_{\text{Te}}$ ) complexes and by residual acceptors ( $\text{Na}_{\text{Cd}}$ ,  $\text{Cu}_{\text{Cd}}$ ). The microscopic structure of ( $\text{Ga}_{\text{Cd}}\text{-Cd}_{\text{Te}}$ ) complex is proposed based on the value of its local phonon mode and the growth conditions. A native defect like  $\text{Te}_{\text{Cd}}$  which has a deep level near the middle-band-gap is supposed to give a stable compensation and a tolerance for variation in shallow impurity concentrations.

Keywords: CdTe, Ga doping, resistivity map, compensation

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### 1. Introduction

In the last years high resistivity CdTe related compounds has been studied due to their applications such as X-ray or gamma detectors and in photorefractive devices. For these applications, semi-intrinsic resistivity, low dark current and low defect density materials are required (FIEDERLE et al.1999). The elements of the III group (In, Ga, Al) are used for residual group I acceptors compensation in CdTe and can be introduced into the crystal either in the growth process (SETO, WRUBEL) or by the post-growth diffusion (FRANCOU, SOCHINSKII). As it was reported by FRANCOU et al. the solubility of Ga in a donor state ( $\text{Ga}_{\text{Cd}}$ ) is better than the solubility of In at the same time-temperature conditions. The segregation of Ga is larger than In, because the value of its segregation coefficient  $k$  is more close to unity compared with In ( $k_{\text{In}} = 0,5$ ) (DE NOBEL, PAUTRAT), following the exponential dependence of  $k$  vs. covalent radius (1.18Å for Al, 1.26 for Ga and 1.44 for In). It was shown earlier that similar to the doping with In (MARFAING), a high level of Ga doping ( $10^{18} \text{ cm}^{-3}$ ) leads to a strong compensation and a resistivity value up to  $3 \times 10^9 \Omega \cdot \text{cm}$  (JOERGER).

A compensation of CdTe with the III group impurities is a complex phenomenon which can include: i) the compensation of residual acceptors of the I group by shallow donors (MARFAING); ii) the formation of complexes between donors (D) and native defects, like  $\text{D-V}_{\text{Cd}}$  or  $2\text{D-V}_{\text{Cd}}$  (MARFAING, KRÖGER, KRÖL); iii) the deep-level defect creation for the Fermi-level stabilisation in the middle-band-gap (FIEDERLE et al.1998); iv) the impurity segregation in tellurium enriched inclusions, metal droplets, precipitates of the second phase, like  $\text{In}_2\text{Te}_3$  or  $\text{Ga}_2\text{Te}_3$ , etc. (PAUTRAT, LAASCH, MAGEE).

In this work semi-insulating CdTe:Ga material was studied by a resistivity cross-section mapping and a low-temperature photoluminescence (LT PL) in order to include both elementary and complex defects into analysis of the compensation phenomenon and its tolerance for variation in impurity concentrations. It was shown that an impurity inhomogeneity is responsible for the resistivity variation.

Beside of the known A-centre defect like  $(\text{Ga}_{\text{Cd}}-\text{V}_{\text{Cd}})$  a new complex  $(\text{Ga}_{\text{Cd}}-\text{Cd}_{\text{Te}})$  was proposed. The microscopic model of  $(\text{Ga}_{\text{Cd}}-\text{Cd}_{\text{Te}})$  complex is based on the LT PL data and on the growth condition. Annealing was carried out in a Cd vapour atmosphere in order to clear up the origin of these defects. Both of these complexes as well as the elementary shallow acceptor  $(\text{Na}_{\text{Cd}}, \text{Cu}_{\text{Cd}})$  are partly responsible for the compensation of shallow donors  $(\text{Ga}_{\text{Cd}})$ , but for the stable compensation a native deep-level defect must be present in the crystal.

## 2. Experimental details

CdTe:Ga single crystals were grown from the vapour phase by the modified Markov method (JOERGER) under vapour control pressure of  $P_{\text{Cd}}/P_{\text{Te2}} = 2$ . The samples were cut off from the as-grown single crystal and mechanically and chemomechanically polished with a Br/Etilenglycol solution (2%Br) (BABENTSOV).

The annealing was carried out in evacuated and sealed off quartz ampoules at 600 °C for 4 hours in a Cd vapour atmosphere, followed by a fast cooling to RT. Afterwards the samples were chemomechanically polished to remove the surface layer at a depth of about 10  $\mu\text{m}$ .

After etching the (111) oriented samples in the Nakagava solution (NAKAGAVA) the dislocation density was studied with an optical microscope with a spatial resolution of 2  $\mu\text{m}$ . A SIEMENS equipment was used for the high resolution X-ray diffraction (HRXRD), in the two-crystals configuration, using  $\text{Cu K}\alpha$  radiation.

Resistivity mapping was done by the temperature dependent capacity measurements (TDCM) (STIBAL). As-grown CdTe:Ga was semi-intrinsic material with resistivity of  $(1-5) \times 10^9 \Omega \cdot \text{cm}$ , which decreases to  $10^8 \Omega \cdot \text{cm}$  after the annealing in Cd vapour.

LT PL measurements were done in a helium inflows cryostat in an interval of 4.2 - 40 K in which temperature is controlled by a calibrated sensor with an accuracy of  $\pm 0.1$  K. An Argon laser with emission at 5145 Å was used for PL excitation, the signal was detected with a photomultiplier (S1) cooled to 80 K and recorded in the regime of photon counting.

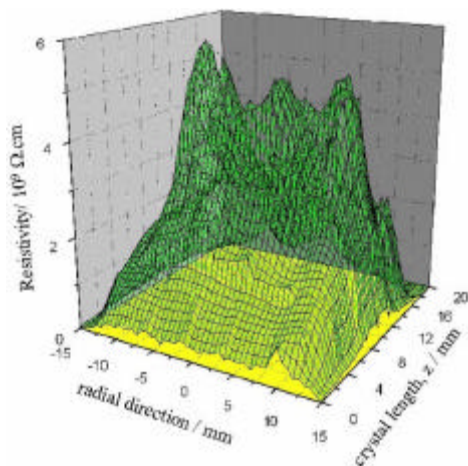


Fig. 1: Resistivity map of CdTe:Ga cross-section.

### 3. Results and discussion

The as-grown CdTe:Ga samples were twin free, with an average dislocation density less than  $10^4 \text{ cm}^{-2}$ , with good crystallinity. The FWHM of the HRXRD curve amounted to 63 arc-sec.

#### A. Resistivity

In Fig. 1 the resistivity map of a CdTe:Ga sample cut from the central part of the ingot along the growth direction is shown. The main result is that the ingot is semi-insulating throughout the whole sample. From this figure it can be seen that high resistivity of  $10^9 \Omega \cdot \text{cm}$  is established from the beginning of the growth and its value is increasing gradually toward the end of the boule up to  $5 \cdot 10^9 \Omega \cdot \text{cm}$  with fluctuations in the radial direction.

#### B. Photoluminescence

##### a) Excitonic spectrum

In Fig. 2 the LT PL spectra at 4.2 K of an as-grown CdTe:Ga sample is shown. The main line in the excitonic region of this spectrum is a neutral acceptor-bound-exciton ( $A^0 \cdot X$ ) at the position of 1.5895 eV due to the Cu residual impurity (MOLVA et al. 1984). A donor-bound-exciton line ( $D^0 \cdot X$ ) due to the  $\text{Ga}_{\text{Cd}}$  shallow donor (FRANCOU) is not clearly pronounced in this spectra. A new prominent V- line at 1.5850 eV in the spectrum has a phonon replica ( $V-L0^*$ ) at 1.5611 eV which gives the value of 23.9 meV, while the LO-phonon energy in CdTe is 21.3 meV (ZANIO). Both lines have the same temperature behaviour (Fig. 3) and have disappeared after the annealing procedure. The second feature of Fig. 2 is the W-line at 1.5810 eV. These V and W lines have been already seen in the LT PL spectra of CdTe films doped with Ga (FRANCOU).

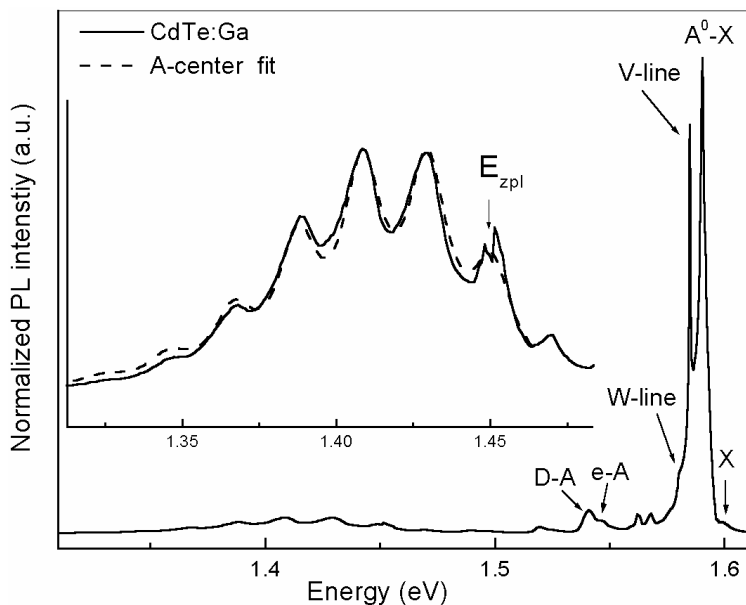


Fig. 2: Low-temperature photoluminescence spectra of CdTe:Ga at 4K

## b) Donor-acceptor recombination spectrum

In the near band-gap D-A region the (e-A) band at 1.5455 eV and (D-A) band at 1.5398 eV can be identified as due to  $\text{Na}_{\text{Cd}}$  acceptor (MOLVA et al. 1982). The broad D-A emission band in the energy interval of 1.35-1.45 eV with LO-phonon structure is often attributed to A-centre or to  $\text{Cu}_{\text{Cd}}$  emission (JOERGER, HOFFMAN, SOLTANI). A computer fitting procedure similar to that used by SOLTANI et al. gives the position of the zero phonon band at 1.4543 eV and the Huang-Rhys parameter of 1.92. This value differs from the value of 1.3 for Cu impurity, because of the A-centre and Cu bands overlapping (SOLTANI). The acceptor ionisation energy ( $E_{\text{A}} = 0.138$  eV) was obtained from the zero-phonon band position (hv) using the expression for the D-A emission:  $h\nu = E_{\text{g}} - (E_{\text{A}} + E_{\text{D}})$  at  $E_{\text{g}} = 1.606$  eV and  $E_{\text{D}} = 0.0138$  eV (FRANCOU, SOLTANI). A summary of defect levels identification in CdTe:Ga is presented in Table 1.

Table 1: Summary of the levels identification in semi-insulated CdTe:Ga. a- FRANCOU; b- present work; c- JOERGER; d- MOLVA et al. 1984; f- HOFFMAN; g- FIEDERLE et al. 1998; h- FIEDERLE et al. 1995;

Levels (eV)	$E_{\text{C}} - 0,0138^{\text{a,b}}$	$E_{\text{C}} - 0,068^{\text{c}}$	$E_{\text{V}} + 0,059^{\text{b,d}}$	$E_{\text{V}} + 0,146^{\text{b,f}}$	$E_{\text{V}} + 0,138^{\text{b,d}}$	$E_{\text{C}} - 0,75^{\text{c,g,h}}$
Origin	$\text{Ga}_{\text{Cd}}$	Donor complex	$\text{Na}_{\text{Cd}}$	$\text{Cu}_{\text{Cd}}$	A-centre	$\text{Te}_{\text{Cd}}$
Method	LT PL	PICTS	LT PL	LT PL	LT PL, photo-EPR	PICTS, Hall

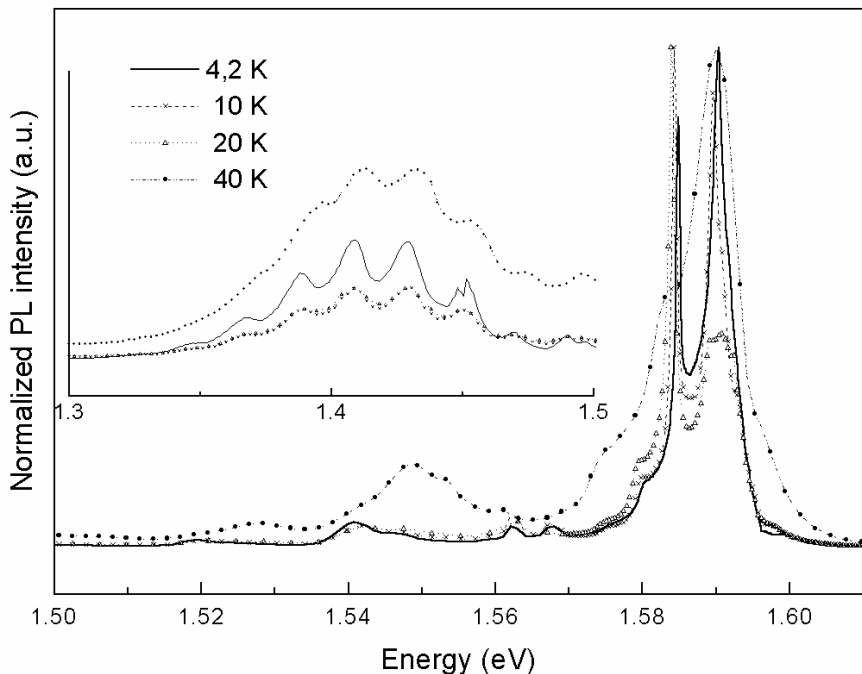


Fig. 3: Temperature dependence of the excitonic PL spectra

In Fig. 3 a temperature dependence of PL spectra is shown. From this figure it is seen that at 4.2 - 20K the V-line position is shifted to the low energy side due to the band-gap shrinkage with temperature (ZANIO, LOPEZ-CRUZ) and its intensity is increased in comparison with intensity of ( $D^0 \cdot X$ ) and ( $A^0 \cdot X$ ) lines. When the temperature is increased up to 40K the V-line intensity is falling down and a broad line appears at 1.59 eV, which is a superposition of free exciton and free-to-bound D-h transitions. Such behaviour is typical for the bound-exciton emission (ZIMMERMAN).

### C. Defect model

Due to its sharpness, temperature behaviour and intensity we can suggest that V-line has a bound-exciton origin. The exciton binding energy to the defect ( $E_{loc}$ ) can be calculated from the LT PL spectra using the expression:  $E_{loc} = h\nu - E_x = 0.0115$  eV, where:  $h\nu = 1.5850$  eV - is the V-line energy position;  $E_x = 1.5965$  eV - is the free exciton energy (FRANCOU). Because the  $E_{loc}$  is linearly linked with the shallow donor ionisation energy ( $E_i$ ) by the Haynes's rule:  $E_{loc} = 0.246 \cdot E_i$  (FRANCOU) we can estimate this energy as  $E_i = 0.047$  eV if the V-line defect is a donor-like complex. A shallow donor with  $E_i = 0.068$  eV which is close to the value estimated from the LT PL was really registered in CdTe:Ga (JOERGER).

The chemical model of this defect can be based on the unusually high value of the V-line's phonon replica (23.9 meV). Similar to the analysis of the phonon mode in the PL emission of CdTe:I (LEE) we can use a linear diatomic chain model (ASHCROFT) where the following expression for the frequency of the local phonon mode can be written:

$$(h \nu^*)^2 = K/m \quad (1)$$

where  $h \nu^*$  is the local phonon energy,  $K$  the force constant and  $m$  the reduced mass of a new chain.

In order to estimate this energy we can do some assumptions, following the previous discussion given by LEE et al.: i) the reduced mass of the only one atom in a Cd-Te diatomic chain is responsible of the total increasing of the mode energy. This atom is  $Ga_{Cd}$  with the atomic weight of 69.72; ii) the force constant of the chain is the same as in the Cd-Te lattice, that is the new chain, which should be at least neutral; iii) to fulfil the last condition the second component of the new chain can be a native acceptor -  $Cd_{Te}$  or  $V_{Cd}$ ; iv) the atomic weight of a this acceptor should be close to the atomic weight of Cd and Te (112.4 and 127.6). The most widely discussed in CdTe native acceptor  $V_{Cd}$  (BERDING) cannot be used in this analysis because of its mass uncertainty.

At all these conditions the  $(Ga_{Cd} - Cd_{Te})^0$  close pair seems to be the defect, which can give the new phonon mode in the LT PL spectra. Its mode energy calculation gives an energy of 24.5 meV using expression (1) with reduced mass. This energy is close to the experimentally observed value of 23.9 meV. The analysis of the growth conditions can support the proposed  $(Ga_{Cd} - Cd_{Te})$  defect:

i) The crystals were grown under not too high Cd over-pressure  $P_{Cd}/P_{Te2} = 2$ , which corresponds to the stoichiometric vapour composition (GREENBERG), that is at conditions at which Ga atoms can occupy the Cd site (BERDING). At these condition it is difficult to expect a high  $V_{Cd}$  concentration, which is known as a constituent part of the A-center ( $Ga_{Cd} - V_{Cd}$ ) (DE NOBEL, HOFFMAN);

ii) Non-equilibrated  $V_{Te}$  in CdTe can be created due to a known Te precipitation (YADAVA), so  $Cd_{Te}$  defect can be a dominant defect in the vicinity of Te inclusions and at a high Ga concentration the  $(Ga_{Cd} - Cd_{Te})^0$  pairs can exist in these areas.

### D. Analysis of the Fermi statistic model

It was shown previously (FIEDERLE et al.1998) that in CdTe with resistivity up to  $10^{10} \Omega \cdot \text{cm}$  the compensation model that is tolerant to the shallow impurity non-homogeneity ( $N_A - N_D$ ) is based on the Fermi statistic, which includes the deep level ( $N_{DD}$ ). Such level really is detected in a high resistivity CdTe with the ionisation energy  $E_{DD}$  near the  $E_g/2$  (FIEDERLE et al.1999, FIEDERLE et al.1998, FIEDERLE et al.1995). At the conditions  $p + N_D + N_{DD} = n + N_A$  and  $N_{DD} > N_A - N_D$  the following expression is valid for the electron concentration ( $n$ ) (FIEDERLE et al.1995);

$$n = [(N_{DD}/N_A - N_D) - 1] \cdot N_C \cdot g \cdot \exp(-E_{DD}/kT) \quad (2)$$

where  $N_C$  is the density of states in the conduction band,  $g$  the degeneracy factor,  $E_{DD}$  the deep level ionisation energy.

The specific resistivity value can be calculated using the formula:

$$\rho = 1/e \cdot n \cdot \mu_n \quad (3)$$

From this expression it can be concluded that the constant value of resistivity can be achieved if the relation  $N_{DD}/N_A - N_D$  and the mobility value  $\mu_n$  are constant throughout the crystal.

To simulate the experimental cross-sectional resistivity distribution shown in Fig. 1 one should know the distribution of all parameters, which are in the formula (2). Usually the mobility value differs not more than in 1.2 – 1.5 times in a high resistivity doped or undoped CdTe (FIEDERLE et al.1998, FIEDERLE et al.1995). Because of that we can neglect the variation of mobility value throughout the CdTe:Ga ingot.

The defect identification made by LT PL has shown that the residual acceptors  $\text{Na}_{\text{cd}}$ ,  $\text{Cu}_{\text{cd}}$  or complexes like  $(\text{Ga}_{\text{cd}} - \text{V}_{\text{cd}})^{\cdot}$  and  $(\text{Ga}_{\text{cd}} - \text{Cd}_{\text{te}})^{\cdot}$  have shallow energy levels (Table 1). They can partly compensate shallow donors ( $\text{Ga}_{\text{cd}}$ ) and a rough estimation of the ( $N_A - N_D$ ) difference can be done from the LT PL spectrum like a deviation of the D-A band to the  $A^{\cdot} \cdot X$  line intensities ratio (FENG, BECKER). In a given sample this ratio can reflect non-homogeneity in the compensation ratio and according to our measurement this ratio differs by the order of magnitude in the axial cross section.

On the other hand it can be shown from Eq. (2) that a semi-intrinsic value of resistivity up to  $5 \cdot 10^9 \Omega \cdot \text{cm}$  can be reached only at the condition  $N_{DD} > N_A - N_D$ , that is all throughout the axial cross section, where the concentration of the deep donor has to be high enough (in fact of the order of  $10^{16} \text{cm}^{-3}$ ). During the CdTe growth from the vapour phase, one should suggest the non-homogeneous distribution of the deep donor which is believed to be  $\text{Te}_{\text{cd}}$  (FIEDERLE et al.1998) and due to the different evaporation rate of impurities from the source material a non-homogeneous axial distribution of the shallow donors and acceptors can be also expected.

## 5. Conclusions

Characterisation of high resistivity ( $5 \cdot 10^9 \Omega \cdot \text{cm}$ ) CdTe:Ga, grown from the vapour phase by the modified Markov method, has been made by resistivity mapping and LT PL. A high crystal quality was confirmed by low dislocation density (less than  $10^4 \text{cm}^{-2}$ ), by low FWHM of the HRXRD curve which was not more than 63 arc-sec and by sharp lines of the bound excitons. Point defects, namely shallow donors  $\text{Ga}_{\text{cd}}$ , shallow acceptors due to residual impurities ( $\text{Na}_{\text{cd}}$ ,  $\text{Cu}_{\text{cd}}$ ) and complexes like A-centre  $(\text{Ga}_{\text{cd}} - \text{V}_{\text{cd}})^{\cdot}$  and  $(\text{Ga}_{\text{cd}} - \text{Cd}_{\text{te}})^{\cdot}$  were

identified by LT PL. A native antisite defect  $\text{Te}_{\text{Cd}}$  is supposed to be responsible for the high level of the compensation giving semi-intrinsic resistivity. The nature of the deep level has to be still studied.

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