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Influence of γ-irradiation on photoluminescence spectra of CdTe:Cl

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Abstract. Photoluminescence properties of high-resistivity CdTe:Cl crystals irradiated with γ-rays have been studied. An enhancement of near-edge luminescence intensity is observed after a low dose γ-irradiation ($D \leq 10$ kGy). For larger doses of γ-irradiation both a decrease of the total luminescence intensity and the intensity redistribution between the lines of excitons bound to different acceptors occur. For the donor-acceptor recombination with A-centers participation, an increase of the Huang-Rhys factor S is found with D increase. This fact can be explained by decrease of the A-centers concentration. The experimentally determined S values are compared with a calculated $S(R)$ dependence for different distances R between donors and acceptors.

Keywords: cadmium telluride, photoluminescence, γ-irradiation, Huang-Rhys factor, low dose effect, self-compensation.

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1. Introduction

Due to a high average atomic number and a large energy gap, cadmium telluride is a very promising material for production of non-cooled X- and γ-radiation detectors. During the last decade, great advancements were achieved in growing high-quality CdTe material and in development of effective methods for detector signal processing. This promoted a wider application of CdTe detectors in medicine, radiology, space studies, etc. (for a recent review, see, e.g., [1-2] and references therein). For effective detection of ionizing radiation, CdTe crystals should meet the following requirements: *i)* high resistivity to reduce the leakage current and, thus, to increase the signal/noise ratio and *ii)* good transport properties, namely high value of mobility-lifetime product (μτ) for electrons and holes, in order to realize the full charge collection at electrodes and, hence, to increase the sensitivity and to improve the energy resolution.

Among different dopants used to obtain high-resistivity ($\rho > 10^8$ Ω·cm) CdTe crystals the best results were obtained by doping with chlorine. Despite the behavior of Cl impurity in CdTe is not completely understood yet, chlorine was established to contribute to the self-compensation phenomena which is typical for II-VI compounds (see, e.g. [3]). According to the most studied model of self-compensation, it is thought that the main intrinsic defects in CdTe is Cd vacancies (V_{Cd}). In the process of CdTe:Cl material growth, chlorine atoms occupy Te sites (Cl_{Te}) and act as shallow donors (E_C-0.014 eV). A part of Cd_{Te} donors interacts with V_{Cd} creating complexes (V_{Cd}-Cl_{Te}) (the so-called A-centers, which act as acceptors with energy level at E_v+0.126 eV) and complexes (V_{Cd}-2Cl_{Te}). A-centers, in turn, compensate influence of Cl_{Te} donors on CdTe:Cl electrical properties [4, 5]. It should be noted however that in some publications [6-8] this mechanism is considered as insufficient to explain alone such high resistivity of the Cl-doped crystals.

A lot of CdTe:Cl detector material studies have been performed earlier. Nevertheless, up to now no complete picture exists on changes in the impurity and defect system of CdTe crystals induced by different kinds of crystal irradiation. Recent studies by Cavallini et al. revealed that high doses of gamma, neutron and electron irradiation cause a significant deterioration of CdTe and CdZnTe detector performance up to complete loss of the detecting ability [9, 10]. Since CdTe detectors may be exposed to very high fluxes of γ-radiation, it is extremely important to have detailed knowledge on radiation-induced changes which can occur in the crystal volume. Therefore, it is necessary to study influence of γ-irradiation on fundamental characteristics of CdTe:Cl material, which determine directly or indirectly its detector properties.

In this paper, to study influence of γ-radiation on the impurity and defect system of high-resistivity CdTe:Cl crystals we have used a method of low-temperature photoluminescence (PL) spectroscopy.

2. Experimental

CdTe:Cl single crystals with Cl concentration of about $2 \cdot 10^{19}$ cm^{-3} were grown by the Bridgman method; the growth technique details were described elsewhere [11]. The electrical resistivity of the samples was $\rho \sim 10^{10}$ Ω·cm and the electron mobility was $\mu_e \sim 700$ $cm^2V^{-1}s^{-1}$. For a comparison we studied γ-irradiation influence on undoped CdTe samples, too, but here we will focus on the properties of CdTe:Cl.

The PL experiments were performed at 5 K using for excitation a cw Ar-laser operating in all-lines mode. The excitation power was about 10 mW/cm^2 and kept constant in all experiments. PL signal was dispersed with a 0.6 m monochromator and detected by a cooled photomultiplier and a conventional lock-in technique.

The samples were exposed to γ-ray using a ^{60}Co γ-cell at a MRX-γ-25 M setup with a dose rate of 95.28 R/s. A predetermined irradiation dose was delivered to the sample by setting the necessary exposure time. The irradiation doses used were in the range D = 1 – 1000 kGy. Measurements of the PL spectra of the samples were performed after each irradiation session and were followed by further irradiation to reach the next dose. The time period between the irradiation sessions and the PL experiments were as short as possible to avoid any relaxation of lattice defects created by γ-rays.

3. Experimental results

In Fig. 1, PL spectra taken at 5 K are plotted for the unirradiated CdTe:Cl sample and g-irradiated with doses 1 kGy and 10 kGy. In the exciton region of the spectra four PL lines denoted in the figure as (D^0, X), (A^0, X), G and W are resolved. A weak luminescence from the free excitons is also observed at 1.598 eV. The origin of the (D^0, X) and (A^0, X) lines is well established. The (D^0, X) line at 1.593 eV is due to excitons bound to neutral donors Cl_{Te} and the (A^0, X) line at 1.589 eV results from annihilation of excitons bound to neutral Cu acceptors.

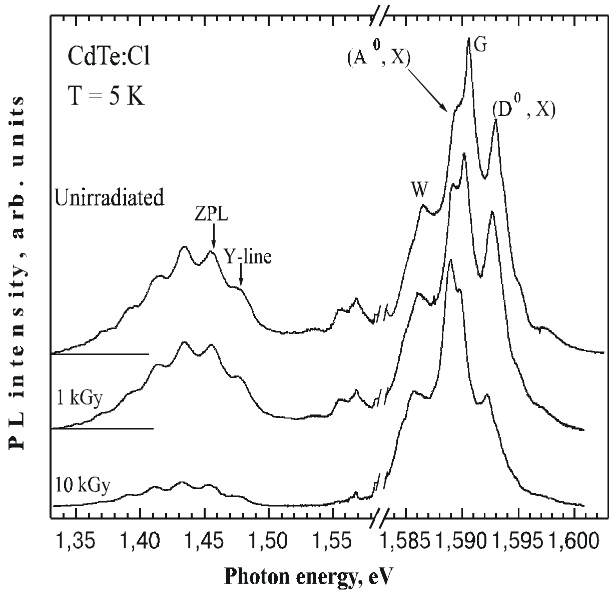

Fig. 1. PL spectra of a CdTe:Cl sample before g-irradiation and irradiated with doses of 1 kGy and 10 kGy. The spectra are normalized to the respective intensity of the (A^0, X) line.

At the same time, some discrepancy exists in the literature in identification of the lines G (1.590 eV) and W (1.586 eV). In Ref. [12] the G line was ascribed to excitons bound to A-centers, and in Refs. [13, 14] it was attributed to excitons bound to acceptors (V_{Cd}-$2Cl_{Te}$). The W line at first was attributed to donor-acceptor (D-A) recombination [15]. Later the excitonic nature of this line was established and it was identified as recombination of excitons bound to acceptors (V_{Cd} - Cl_{Te}). In any case, both G and W lines appear due to introduction of the Cl impurity into the CdTe matrix, namely, due to the creation of complexes involving Cl_{Te} centers.

Irradiation of CdTe:Cl with γ-rays results in a decrease of intensity of all exciton lines connected with the presence of the Cl_{Te} centers ((D^0, X), G and W lines) as compared to the (A^0, X) line intensity. The (A^0, X) becomes the most intensive line in the spectrum after sample irradiation with a dose 10 kGy. At the same time, the total exciton intensity for the both samples increases for low exposure doses reaching the maximum at about 10 kGy for CdTe:Cl and 3 kGy for undoped CdTe. A higher irradiation dose causes a reduction of the intensities down to the initial value or even lower. A dose dependence of integral intensity of all exciton line is shown in Fig. 2 for Cl-doped and undoped CdTe.

Except these lines we also observed: (a) a weak emission band assigned in the literature to the conduction band – acceptor transitions and the D-A recombination

P > 1MW/cm^2 [21]. At the same time, intensities of the other PL lines at lower energy were almost constant or even decreased. Thus, we can conclude that the 1.35-1.50 eV PL band is a superposition of two PL lines: the Y-line and the A-center DAP recombination with zero-phonon lines at ~1.478 eV and ~1.455 eV, respectively, and corresponding LO-phonon replicas.

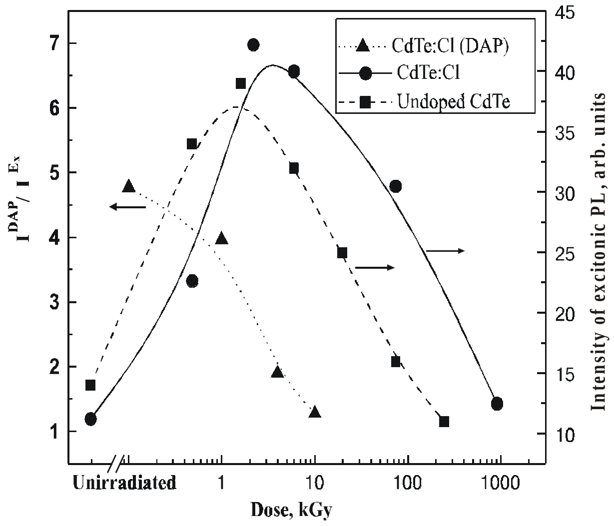

Fig. 2. Dose dependencies of intensities of the excitonic PL (right y-axis) for CdTe:Cl (dots) and undoped CdTe (squares) and intensity of the DAP recombination line normalized to I^{ex} (left y-axis, triangles). The lines are drawn as a guide to for the eye.

as well as their phonon satellites at 1.53 – 1.58 eV and (b) a broad band that includes well resolved LO-phonon replicas in the region 1.35-1.50 eV. Analysis of the last PL band at 1.35-1.50 eV is the scope of the paper. This band was assigned by de Nobel to the recombination of the donor-acceptor pairs (DAP) consisting of A-centers and Cl_{Te} [16]. However, in this spectral range the DAP recombination with participation of background impurities Cu, Ag, Au can be also detected [8]. Analysis of the 1.35-1.50 eV emission is complicated for the possible coexistence and superposition in this spectral region of PL bands having different nature [8, 17-19]. Therefore, a great inaccuracy may arise when determining the parameters of this band, for example the Huang-Rhys factor value which characterizes electron-phonon coupling. In particular, very high values of the Huang-Rhys factor (S≈2.2 or even higher) are reported for the DAP recombination [19, 20] which exceed substantially the calculated one (S≈1.4). As a rule, the most short-wavelength line (1.478 eV) of the band is considered as zero-phonon line (ZPL) of the DAP luminescence. However, the 1.478 eV line is not always observed in the PL spectra of the CdTe:Cl samples and, on the contrary, it may appear in undoped crystals in which no V_{Cd}-Cl_{Te} complexes are created at all. In several papers, it was reported however that the 1.478 eV line (the so-called Y-line) is connected with the carrier recombination at extended defects, and a weak electron-phonon coupling for this line was found [17, 18]. In our recent studies on CdTe:Cl crystals covered by a thin SiO_2 film, a very strong enhancement of the 1.478 eV line intensity was observed after the sample irradiation with the second harmonic of an YAG:Nd laser at a

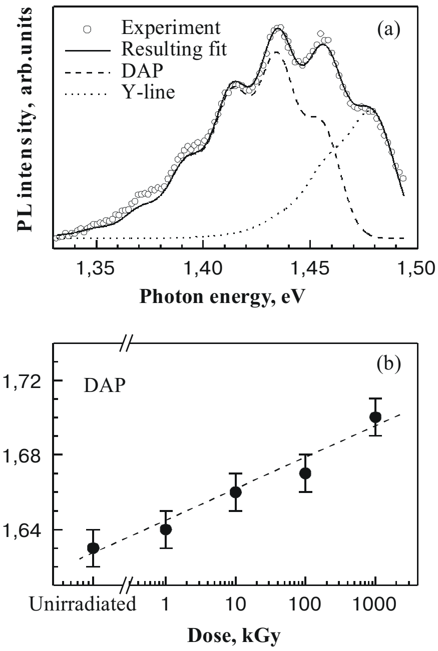

Fig. 3. (a) An example of the 1.35-1.50 eV band fitting with two lines (see text for details). (b) Dose dependence of the Huang-Rhys factor for the DAP recombination with A-centers participation.

pulse power P>1 MW/cm^2 [21]. At the same time, intensities of the other PL lines at lower energy were almost constant or even decreased. Thus, we can conclude that the 1.35-1.50 eV PL band is a superposition of two PL lines: the Y-line and the A-center DAP recombination with zero-phonon lines at ~1.478 eV and ~1.455 eV, respectively, and corresponding LO-phonon replicas.

The above speculations were taken into account when analyzing the DAP emission spectra and determining the Huang-Rhys factor in the dependence of the γ-irradiation dose. An example of fitting the experimental PL band with two PL lines (Y-line and DAP) having different S values is shown in Fig. 3(a). The best fits were obtained by changing S and the full width at half maximum for the two lines, whereas corresponding energies of the ZPLs and the LO-phonon energy (21.2 meV) were kept constant for all doses of γ-irradiation. The Huang-Rhys factor for the Y-line (S^Y≈0.6) was found to be much lower than that for the DAP PL, and it remained almost constant at increasing dose of γ-irradiation. At the same time, the Huang-Rhys factor for the DAP recombination exhibits an increase from S^{DAP}≈1.63 for unirradiated sample to S^{DAP}≈1.70 for irradiated sample with a dose 1000 kGy (Fig. 3(b)). This increase is accompanied with a decrease of the DAP recombination intensity as compared to the intensity of exciton lines, see Fig. 2.

4. Discussion

The near-edge luminescence in semiconductors is known to be very sensitive to non-radiative losses at deep recombination centers created by mechanical stresses, intrinsic defects, impurities, etc. Increasing PL intensity at low doses of γ-irradiation implies a reduction of the concentration of non-radiative centers. This phenomena is known in literature as the "low-dose effect" or the radiation-stimulated ordering of the crystalline structure. It results in relaxation of thermodynamically nonequilibrium metastable phases in crystal volume induced by a low dose of penetrative radiation. The low dose effect is well studied for Si, Ge and III-V compounds. It was found, in particular, that radiation-stimulated processes are enhanced in sub-surface region of a semiconductor and in heterostructures [22, 23]. The low-temperature PL spectroscopy was shown to be a very effective tool for studying this phenomena [24, 25]. Radiation-stimulated ordering of sub-surface region was recently observed in CdTe as well [26].

As it was shown above, exciton PL intensity depends non-monotonically on the γ-irradiation dose. An increase of the exciton PL (by ~ 4 times) indicates on "low-dose effect" manifestation. For the undoped sample, the maximal increase of the PL intensity occurs at the lower dose as compared to CdTe:Cl. Therefore, it is reasonable to suggest that the "low-dose effect" will not be observed in high purity crystals with a very low concentration of lattice imperfections. On the contrary, the greater concentration of intrinsic defects and impurities the higher irradiation dose is required to reach some improvement in the lattice perfection.

Let us analyze the shape of the PL spectrum at 1.35-1.50 eV represented by the Y-line and the DAP recombination band arising due to the A-centers participation. The effect of electron (hole) interaction with LO-phonons on radiative recombination processes at impurity centers in semiconductors was considered theoretically in a number of works (see, e.g., [27-29]). Theoretical values of the Huang-Rhys factor for exciton, free electron–acceptor and DAP recombination processes were calculated in these works mainly within the framework of the effective mass approximation and the hydrogen-like model. In many cases, however, the obtained results do not coincide quantitatively with experimental data, so a further development of the theory is necessary.

The most general, i.e. valid not only for the hydrogen-like shallow centers, but for relatively deep recombination centers too, is the quantum defect approach developed in [29]. In this approach, highly localized ground states of carriers at impurity centers are described by the envelope wave functions of the type

$$\Psi_{\nu_i}(r) = N_{\nu_i} r^{\nu_i} \exp[-r/(\nu_i a_i^*)]$$

where $i=e$ for the bound electron state at a donor and $i=h$ for the bound hole state at an acceptor, r is a radius-vector, $a_i^* = \hbar^2\varepsilon_0/(e^2 m_i^*)$, is the effective Bohr radius for the electron (hole), m_i^* - the carrier effective mass, ε_0 - the static dielectric constant of material, $N_{\nu_i} = [2/(\nu_i a_i^*)]^{\nu_i}/[(\nu_i a_i^*)^{1/2}\Gamma(\nu_i+1)]$ the normalization factor, ν_i - the so-called quantum defect parameter defined from the energy relationship $\nu_i^2 = E_i^R/E_{D(A)}$. In the last expression $E_{D(A)}$ is the donor (acceptor) ground state ionization energy determined experimentally and $E_i^R = e^4 m_i^*/(2\varepsilon_0^2\hbar^2)$ is the binding energy of the carrier at the corresponding impurity center within the framework of the hydrogen-like model.

The Huang-Rhys factor determines the probability $W_p \sim e^{-S}S^p/p!$ of radiative transitions at impurity center with emission of p LO-phonons. It can be calculated using well-known formula of the Frцhlich continuum theory

$$S = [2\pi e^2/(V\hbar\omega_{LO})](1/\varepsilon_\infty - 1/\varepsilon_0)\sum_{\mathbf{q}}|\rho_{\mathbf{q}}|^2/q^2,$$

where $\mathbf{q}$ is the wave vector, V - the crystal volume, $\hbar\omega_{LO}$ the energy of LO-phonons, ε_∞ - the high-frequency dielectric constant, r_q the Fourier-component of charge density distribution. For the recombination at an isolated center (donor or acceptor) the carrier charge distribution is determined by the formula

$$\rho_{\mathbf{q}i} = N_\rho \int \exp(i\mathbf{qr})\ r^{2(\nu_i-1)} \exp[-2r/(\nu_i a_i^*)]\ d^3\mathbf{r},$$

where N_p is the normalization constant defined from the condition $\rho_{0i} = 1$. If a D-A pair is involved in the process of radiative recombination, then Fourier-component of the common (electron+hole) charge density distribution is expressed as

$$\rho_{eh}(\mathbf{q}) = \langle\Psi_e(\mathbf{r}_e)\Psi_h(\mathbf{r}_h)|\exp(i\mathbf{qr}_h) - \exp(i\mathbf{qr}_e)|\Psi_h(\mathbf{r}_h)\Psi_e(\mathbf{r}_e)\rangle \quad (1)$$

With such charge distribution the value of the Huang-Rhys factor becomes dependent on the distance R between the donor and acceptor:

$$S(R) = e^2/(\pi\hbar\omega_{LO})\ (1/\varepsilon_\infty - 1/\varepsilon_0)\ \times \int_0^\infty \left[\ |\rho_{qe}|^2 + |\rho_{qh}|^2 - 2\rho_{qe}\rho_{qh}\sin(qR)/(qR)\right] dq. \quad (2)$$

For the case of n=1 the final result takes the analytical form of the hydrogen-like model presented in [28]. The hydrogen-like model has to be completely valid for the donor centers due to small experimental values of Cl_{Te} ionization energy (~14 meV) and practically exact observance of the equality $E_e^R/E_D = 1$. For a hole at an acceptor (A-center) situation is not so clear despite the equality $E_A \approx E_h^R \approx 120$ meV, because ***i***) Bohr radius of the ground state in the hydrogen-like model obtained with handbook values of hole effective mass and static dielectric constant is too small (~ 6 Å) for ignoring possible changes in both effective mass and screening and, as a result, in theoretical value of binding energy E_h^R itself, and ***ii***) A-center in fact is not a point defect. It has a definite spatial structure, which for such small radius of localization, can influence to some extent on the hole movement. For these reasons the use of hydrogen-like model is an open question for the case of A-centers despite formal validity of $\nu_h \approx 1$

The calculated dependence $S(R)$ for the considered D-A recombination is shown in Fig. 4. In the upper edge of this figure, several values of DAP concentration are given, which correspond to the mean distance R on the x-axis. The decrease of the Huang-Rhys factor with the decrease of inter-defect distance in the D-A pair is caused by growing mutual compensation of charge distributions of these defects and corresponding decrease in the adiabatic potential shifts for excited states in the configurational Frank-Condon diagram. With the increase of distance R between A-centers and donors the dependence $S(R)$ exhibits a saturation at $S\approx1.4$.

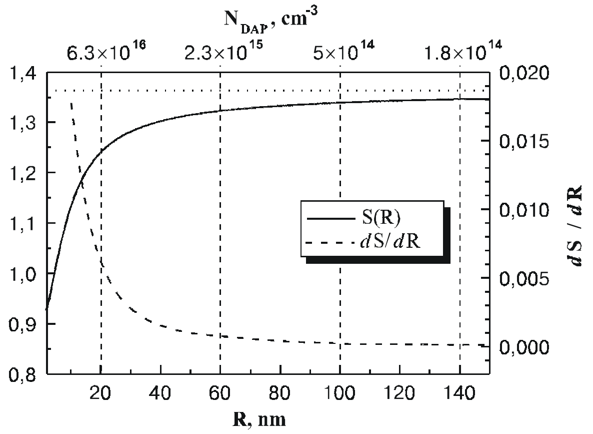

Fig. 4. Calculated dependencies of the Huang-Rhys factor S (right y-axis) and dS/dR on the mean distance R between donor and acceptor. For several values of R, respective DAP concentrations N^{DAP} are given on the top of the figure.

Spectral shape of the DAP band with account for LO-phonon replicas can be calculated using the following expression [28]:

$$I(\omega) \sim \omega^2 \sum_{p=0} \frac{S^p}{p!} e^{-S} \exp\left[-\frac{1}{2}\left(\frac{\omega_0 - p\omega_{LO} - \omega}{\Gamma}\right)^2\right] \qquad (3)$$

where frequency ω_0 determines position of the ZPL, Γ is the line width, summation is taken over all phonon replicas (p=0, 1, 2 ...).

Fitting the experimental spectra obtained at different doping levels and doses of γ-irradiation we gain information on variations in the Huang-Rhys factor, mean distance between recombination centers and DAP concentration due to the corresponding treatment. As it was mentioned above, for a better fitting between theoretical and experimental spectra in the studied spectral region, it is necessary to use more than one series described by Eq. (3) with different position of ZPLs and values of S. For example, S_A fitting in Fig. 3(a) have been obtained with two series of lines (3). As a result, the value of the Huang-Rhys factorobtained from the fitting of the DAP recombination series with A-centers participation differs from the calcu-lated one only by ~20 %. In our case, this difference is much lower as compared to other works where the influence of the Y-line with ZPL at 1.478 eV was ignored. This 20 % difference may be a consequence of above outlined shortcomings of the hydrogen-like model for A-centers.

An increase in the Huang-Rhys factor values at higher doses of γ-irradiation can be explained by a decrease of the DAP concentration. This conjecture is supported by the dependence of the relationship I^{DAP}/I^{ex} on the γ-irradiation dose obtained in experiment (see Fig. 2). About three times decrease in I^{DAP}/I^{ex} is observed with the dose rise from 1 to 10 kGy which corresponds to analogous decrease in the DAP concentration N^{DAP}.

From the experimentally obtained dependencies $I^{DAP}/I^{ex}(D)$and $S^{DAP}(D)$ it is possible to evaluate the concentration of the A-centers in the studied material using the dS^{DAP}/dR dependence (dashed line in Fig.4). It can be done supposing that the real $S^{DAP}(R)$ dependence has the same curvature as the calculated one. In practice, the following method have been used for such evaluation. For the sample irradiated with two doses D_1 and D_2 ($D_2 > D_1$) we obtain the difference in the Huang-Rhys factors $\Delta S^{DAP} = S^{DAP}_{D2} - S^{DAP}_{D1}$ from the $S_A(D)$ dependence and the DAP intensities ratio $k = I^{DAP}_{D1} / I^{DAP}_{D2}$ from the $I^{DAP}/I^{ex}(D)$ dependence. The latter corresponds to the concentration ratio $k = N^{DAP}_{D1} / N^{DAP}_{D2}$. Then, for each value of N^{DAP} that relates to a D-A distance R_1 through N^{DAP}=$0.5R^{-3}$, we found R_2 that corresponds to decreasing N^{DAP}_{D2} by k times. After that we built a $\Delta S/\Delta R(R)$ curve, where $\Delta R = R_2 - R_1$. The intersection of this curve with theoretical dS^{DAP}/dR dependence gives an approximate value of N^{DAP} in the dose range D_1-D_2. From such evaluation we have found the values $N^{DAP} \leq 2\cdot10^{15}$cм$^{-3}$ in the range of g-irradiation dose 1-10 kGy. This value is in agreement with results reported in [30, 31], where the upper edge ~$6\cdot10^{15}$cm^{-3} for V_{Cd} concentration in CdTe:Cl was determined.

5. Conclusions

We have studied PL properties of CdTe:Cl crystals subjected to γ-irradiation with different doses. The "low-dose effect" was found in Cl-doped and undoped CdTe crystals which manifests in an enhancement of the exciton PL intensity (by ~4 times) for exposure dose $D\leq10$ kGy. At further dose increase a quenching of the near-edge PL occurs due to generation of non-radiative recombination centers.

A non-monotonic dependence of the exciton PL intensity on D was accompanied by a redistribution of the relative intensity of the individual lines: PL intensity of excitons bound to centers which include Cl_{Te} ((D^0,X), G and W) decreased more rapidly as compared to the (A^0, X) line connected with background Cu impurity. Intensity of the 1.35-1.50 eV line also decreased with increasing D which is explained by a decrease in concentration of the DAP consisting of Cl_{Te} donors and A-center (V_{Cd}-Cl_{Te}) acceptors. This conclusion is supported by observed increase in the Huang-Rhys factor S^{DAP} for the DAP recombination at the γ-irradiation dose increasing.

It was shown that the DAP concentration can be estimated using the $S^{DAP}(D)$ dependence and the calculated dependence dS/dR, where R is the mean distance between donor and acceptor. The DAP concentration in investigated CdTe:Cl samples was estimated to be $N^{DAP} \le 2\cdot10^{15}$ cm^{-3}.

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