Temperature quenching of the terahertz photoluminescence of shallow acceptors in HgCdTe ternary alloy

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The capture times of holes to the shallow excited levels of neutral mercury vacancy via acoustic phonon emission are calculated for $Hg_{1-x}Cd_xTe$, as well as the transition times of holes from shallow localized levels to the continuum of the valence band at different temperatures. Due to the redistribution of carriers in the valence band with temperature, the time of carrier capture to the localized levels of the neutral vacancy increases, and the time of reionization to the continuum decreases. Based on the calculation results, a model is proposed to describe the temperature quenching of photoluminescence caused by radiative transitions between the localized states of holes on a neutral mercury vacancy

Keywords: HgCdTe, photoluminescence, shallow acceptor.

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1. Introduction

Solid solutions $Hg_{1-x}Cd_xTe$ (CMT) have been studied for about six decades [1,2]. Interest in such structures is caused by the possibility of changing the width of band gap of the material by selecting the composition of the solid solution: from 1.6 eV in pure CdTe to 0 in a solid solution with Cd concentration Cd $< 16.5\%$. This fundamental property of CMT solid solutions makes such materials promising for far infrared optoelectronics [3]. One of the main problems for the creation of interband detectors and sources designed for the long-wavelength range is the reduction of carrier lifetime due to recombination by the Shockley–Reed–Hall (SRH) mechanism through impurity-defect centers [4–6].

The most common defect of HgCdTe is the vacancy of mercury, which occurs in these materials due to the weakness of the mercury–tellurium chemical bond. The mercury vacancy is a doubly charged or double acceptor. Such an acceptor can be in three charge states: a neutral A_2^0-center with which two holes are connected, a once ionized A_2^{-1}- center with which one hole is connected, and, finally, a completely ionized A_2^{-2}-center free of holes. Although mercury vacancies are always formed in HgCdTe solid solutions, their energy spectrum is still insufficiently studied. Relative consensus was reached only regarding the fact that the vacancy is a double acceptor and the ionization energy of the A_2^0-center is ~ 10 MeV for ~ 0.2 [7–9] and 14 MeV for $x \sim 0.3$ [10–12].

Determining the energy spectrum of vacancy is of considerable interest, since they can play a significant role in recombination via the SRH mechanism. Such recombination is a competing process with respect to both interband radiative recombination and nonradiative Auger recombination. Usually, the capture of carriers at centers in the band gap during recombination by the SRH mechanism is nonradiative: electrons and holes give up their energy to phonons during capture. However, in some cases the capture process is accompanied by intracenter transitions with the emission of photons. Thus, the SRH process may turn out to be partially radiative. These radiative processes were observed in studies of photoluminescence (PL) of a number of semiconductors and low-dimensional heterostructures [13–15].

For the first time, long-wavelength PL in the far infrared (IR) range associated with acceptor states in bulk CMT epitaxial layers and HgTe/CdHgTe heterostructures with quantum wells was observed in papers [16,17]. Lines were found in the PL spectra near quantum energies of 10 and 20 meV (the latter value is close to the ionization energy A_2^{-1} of the center). A distinctive feature of such „impurity“ lines is that, in contrast to the line of interband transitions, they do not shift to shorter wavelengths with temperature increasing, which leads to increase in the width of the band gap.

The PL studies in the paper [16] were carried out on a structure with electronic type of conduction, in which all acceptor centers were completely ionized. In this case, all mercury vacancies in the absence of illumination were in A_2^{-2}-charge state, and A_2^{-1}-centers, due to which the longest-wavelength PL is observed, appeared due to the capture of holes excited by illumination from the valence conduction band to partially ionized mercury vacancies with the formation of A_2^0-centers. As already noted, measurements of the CMT PL spectrum of the structure

were carried out in a wide temperature range in [16]. A non-monotonic dependence of the intensity of the long-wave band of the PL was found with increasing temperature. It was shown in [16] that the non-monotonic nature of the dependence of the intensity of the CMT PL structure on temperature is associated with a sharp increase in the number of A_2^{-1}-centers involved in the formation of the PL signal with an increase in temperature > 40 K. As for the decreasing sections of the dependence of the intensity of the PL line on temperature, such signal damping has not yet been described in detail. It was shown in [18] that the PL spectrum of the CMT structure is formed by the transitions of holes between the excited states of holes in mercury vacancies and the ground level. The occupation of the upper energy states is ensured by the rapid nonradiative capture of holes into small excited levels of the neutral mercury vacancy during the emission of acoustic phonons. The accumulation of holes at such levels makes it possible to observe radiative transitions with the largest matrix element in the terahertz PL structure. With this process competes the process of capturing media to deep levels with the emission of optical phonons [19]. Such capture occurs without the participation of small excited states and does not lead to the occurrence of PL.

It is shown in [19] that the rate of capture of holes with the emission of optical phonons increases rapidly with increasing temperature. Such an increase in the rate of emission of optical phonons in [19], was associated with the presence of a decreasing section of the dependence of the intensity of the long-wavelength PL band on temperature in the range from 20 to 40 K. However, with a further increase in temperature, the frequency of emission of optical phonons changes little. This is a slight increase in the speed of the process 2 cannot explain the rapid attenuation of PL with an increase in temperature from 70 to 100 K. This work is devoted to the study of the causes of PL quenching of narrow-band CMT structures. Since the cause of the appearance of terahertz PL is the accumulation of holes in the excited states of the neutral vacancy of mercury, in order to understand the causes of the temperature quenching of PL, it is necessary to describe the processes of non-radiative filling and emptying of such levels.

2. Calculation method

It should be noted that the frequencies of transitions of holes from the continuum of the valence band to localized states of the neutral vacancy of mercury during spontaneous emission of acoustic phonons were calculated in [18]. As it was shown in this work, the capture times of holes into small ones with a binding energy of < 1 MeV, the states of the neutral vacancy of mercury in the CMT layer with a fraction of cadmium in the solution of $\sim 20\%$ with spontaneous emission of acoustic phonons are several picoseconds. There is a state among such small levels

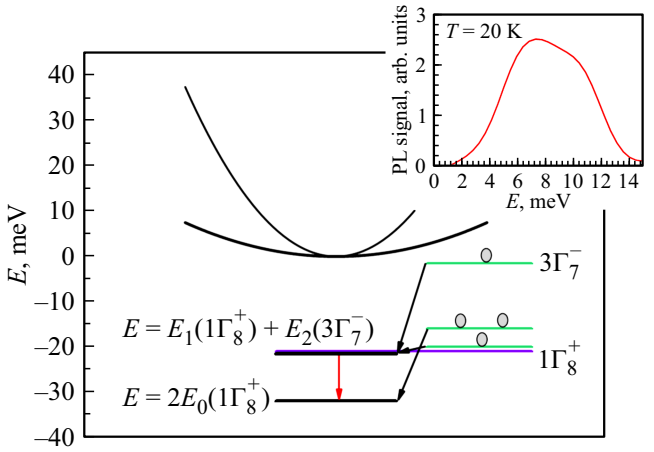

Figure 1. Energy diagram of 2- and 1-partial states of mercury vacancies in the HgCdTe layer with a fraction of cadmium in the solution $x = 0.19$. The energy is counted from the edge of the valence band. Purple line — the energy of the partially ionized A2-1 center (single-particle state). The energy of the neutral center state (black lines) consists of the energies of two particles on the acceptor, indicated by green lines. The diagram shows the ground state of the center and the excited state from which optical transitions of holes to the ground level are observed (red arrow). The insert shows the photoluminescence spectrum caused by such transitions. (A color version of the figure is provided in the online version of the paper).

from which optical transitions manifest themselves in the PL spectrum (state $3\Gamma_{7-}$ in the classification [18], Figure 1).

The bond energy of this state is 0.42 MeV [18], and the time of its filling with carriers during spontaneous emission of acoustic phonons, at the temperature of liquid helium and at the concentration of holes in the valence band $5 \cdot 10^{14}$ cm^{-3} is ~ 2 ps [19]. The filling time of this state through the transitions of holes from the valence band during the emission of acoustic phonons and the emptying of this level through the absorption of phonons and the transition of holes into the continuum will be calculated in a wide temperature range in this paper.

Consider the transitions of holes between the states of the valence band and the localized states of the neutral vacancy of mercury during the emission or absorption of an acoustic phonon. The intensity of hole transitions between continuum states and localized levels of neutral mercury vacancy during interaction with acoustic phonons was calculated on the basis of the Fermi golden rule. Phonons cause displacements of atoms and thereby distort the ideal periodic potential of the lattice. Let us denote the correction to the crystal potential due to lattice vibrations as δV. The potential δV can be decomposed into an electrostatic macropole $\delta\bar{V}$ and a deformation micropole $\delta\tilde{V}$, $\delta V = \delta\bar{V} + \delta\tilde{V}$ [20]. A macrofield arises only in polar semiconductors, in which polarization occurs when atoms of the lattice are displaced. It was shown in [20] that in polar semiconductors the interaction of carriers with the

macrofield usually turns out to be more significant than with the deformation potential, so we will take into account scattering only on the macrofield.

It is possible to calculate the probability of absorption or emission of a phonon per unit of time during the transition of a hole between acceptor states using the following expression according to [18]:

$$W_{i\to f} = \int \left[\frac{2}{M\omega_\phi}\right] \frac{e^4}{a} |\langle \Psi_{\text{cont}} | e^{i\mathbf{qr}} | \Psi_f \rangle|^2 \left(N_\phi + \frac{1}{2} \pm \frac{1}{2}\right) \times \delta(\varepsilon_i - \hbar\omega_\phi - \varepsilon_f) d^3\mathbf{q}, \quad (1)$$

where N_ϕ — the number of phonons, $\Psi_i, \Psi_f, \varepsilon_{i,f}$ — the wave functions and energies of the initial and final states of the neutral mercury vacancy, respectively „+“ corresponds to the emission of a phonon „–“ corresponds to phonon absorption, ω_ϕ — phonon frequency, which can be expressed as $\omega_\phi = sq$, where s — the speed of sound. The magnitude of this velocity can be estimated from the dispersion curves for longitudinal and transverse acoustic phonons in HgTe and CdTe from the works [21,22]. The velocities of HgTe-like phonons are 723 and the velocities of transverse and longitudinal phonons are 1987 m/s, respectively; the velocities of CdTe-like phonons are 1376 and 2522 m/s. A spherical approximation was used as in the calculation of the wave functions of acceptors, i.e. the phonon dispersion law was assumed to be isotropic, a — the lattice constant of the solid solution $Hg_{1-x}Cd_xTe$. Its value — linear interpolation between the values of constant lattices for HgTe (6.46 Å) and CdTe (6.48 Å) [23], M — sum of the atomic masses of the unit cell (the mass of the mercury atom plus the mass of the tellurium atom), e — electron charge.

The hole capture frequency (inverse capture time) from a continuous spectrum can be obtained from the expression (1) integration over the states of a continuous spectrum taking into account the distribution of holes:

$$\frac{1}{\tau_1} = \sum_{\text{cont}} W_{\text{cont}\to f} f. \quad (2)$$

The frequency of the transition of a hole into a continuous spectrum from localized states of holes has the form

$$\frac{1}{\tau_2} = \sum_{\text{cont}} W_{\text{cont}\to f} (1 - f). \quad (3)$$

The free run time for scattering holes on piezoacoustic phonons is calculated in *Appendix* 1. This calculation showed that such a time is < 0.1 ps, which is much less than the lifetime of carriers in the zone, so the distribution function f can be considered quasi-equilibrium:

$$f = \frac{1}{1 + \exp\left(\frac{\varepsilon_{\text{cont}} - F}{T}\right)}, \quad (4)$$

where F is the quasi-Fermi level, the position of which it depends on the concentration of holes in the valence band and temperature, according to the expression:

$$p = \frac{2}{(2\pi)^3} \left(\int \frac{d^3\mathbf{k}}{1 + \exp\left(\frac{E_V + \frac{\hbar^2 k^2}{2m_{hh}} - F}{T}\right)} + \int \frac{d^3\mathbf{k}}{1 + \exp\left(\frac{E_V + \frac{\hbar^2 k^2}{2m_{lh}} - F}{T}\right)} \right). \quad (5)$$

Here m_{hh} and m_{lh} — the effective mass of heavy and light holes in the solid solution of the CMT, respectively. Note that the mass of light holes is by order of magnitude smaller than the mass of heavy holes. Therefore, the second term in expression (4) turns out to be much less than the first, and it can be neglected.

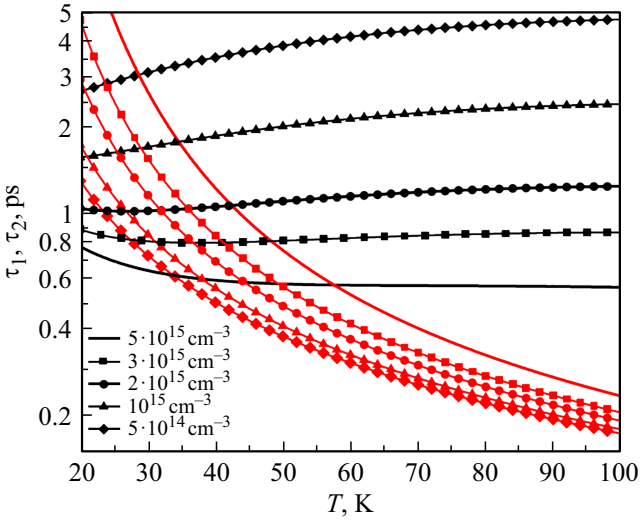

Figure 2. Dependences on the temperature of the hole capture time during the emission of acoustic phonons on the state of $3\Gamma_{7-}$ neutral mercury vacancy in the epitaxial layer $Hg_{1-x}Cd_xTe$ with a fraction of cadmium in solution $x = 0.19$ (τ_1, black lines) and the time of departure of holes into the valence band at the position of acoustic phonons (τ_2, red lines), calculated at different values of carrier concentration in the zone (indicated in the legend).

3. Results and discussion

Figure 2 shows the temperature dependences of the hole capture time on the state $3\Gamma_{7-}(\tau_1)$ and the time of the carrier's departure to the continuum from this state (τ_2). The calculation was carried out for the parameters of the CMT structure from [13] for different carrier concentrations in the zone.

It can be seen that the filling time of the state τ_1 increases with increasing temperature, the emptying time

of the state τ_2 — decreases. This is due to a change in the hole distribution function in the zone (see expressions (2) and (3)). For the frequency of capture of holes, summation occurs by the states of the valence band filled with holes, and for the frequency of emptying — by empty ones.

It should be noted that there are limiting energies of acoustic phonons. This is 10.56 MeV for an HgTe-like phonon and 13.4 MeV for a CdTe-like phonon. This circumstance limits the kinetic energy of carriers involved in transitions to localized states from the valence band and, conversely, the kinetic energy of holes transitioning from localized levels during phonon absorption. This energy should not exceed $\hbar\omega_{\text{lim}} - E_i$, where $\hbar\omega_{\text{lim}}$ is the limiting energy of the phonon, and E_i is the ionization energy of the state to which the carrier passes. Thus, carriers with kinetic energy > 12.98 MeV, cannot participate in the capture and emission of phonons. The distribution function „spreads“ with increasing temperature, which means that fewer and fewer carriers turn out to have energy < 12.98 MeV. Therefore, the frequency of capturing holes should decrease with increasing temperature based on the expressions (2) and (3), and the frequency of leaving the hole in the continuum, on the contrary, should grow.

We do not know the concentration of free carriers in the valence band created by illumination. It can be seen from Figure 2 that, depending on the concentration of holes in the valence band, the ratio between the filling time of the state $3\Gamma_{7-}$ and its emptying into the valence band will be different. Thus, the emptying time will be noticeably (2 times at low temperature and 20 times at high) less than the filling time of small excited states in the entire temperature range at a low concentration of holes ($5 \cdot 10^{14}$ cm^{-3}), at which PL can be detected (from 20 to 100 K). This means that such small states will not be filled, and relaxation of holes from the valence band will occur through the emission of optical phonons. The time of such a process at high temperature for the concentration of holes $5 \cdot 10^{14}$ cm^{-3} is ~ 15 ps [19].

The time of filling with carriers of the state $3\Gamma_{7-}$ at low temperatures turns out to be significantly less than the time of emptying this state at low temperature at a higher concentration of holes (see the line without symbols in Figure 2). These times become comparable at high temperature, then the time of emptying the state $3\Gamma_{7-}$ becomes less than its filling time with a further increase in temperature.

It is necessary for the occurrence of PL that a hole from the valence band be captured at the A_2^{-1}-center with the formation of a neutral vacancy in a state in which one of the holes is at one of the excited levels. It is possible to write a balance equation between the number of partially ionized A_2^{-1}-centers and neutral A_2^0-centers, given that the processes of transitions between the continuum and small excited states of the neutral vacancy are much faster than the transitions of holes between localized states [18]. In this case, the balance equation between the concentration of A_2^{-1}-centers and A_2^0-centers with holes at small excited

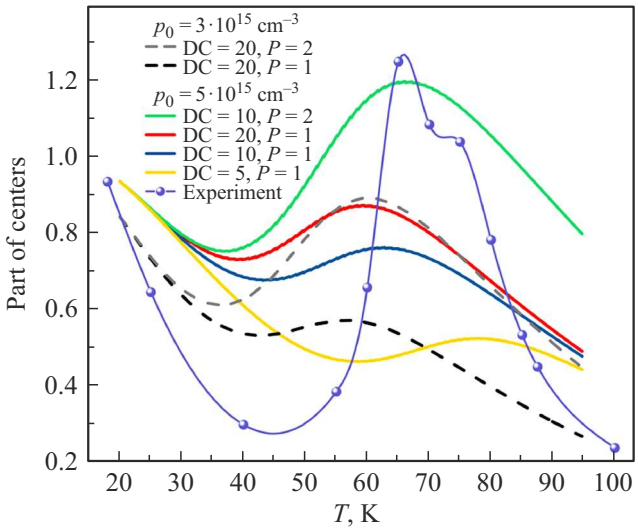

Figure 3. Dependence of the number of neutral mercury vacancies with populated fine excited states, attributed to the total number of neutral and partially ionized centers on the temperature in the epitaxial layer $Hg_{1-x}Cd_xTe$ with a fraction of cadmium in solution $x = 0.19$ and the total concentration mercury vacancies $2 \cdot 10^{14}$ see^{-3}.

levels looks like this:

$$\frac{N_{A^0}}{\tau_2} = \frac{N_{A^{-1}}}{\tau_1}. \tag{6}$$

As it was shown above, the times of emptying and filling of small excited states of the neutral mercury vacancy vary depending on the concentration of holes in the valence band. This concentration is determined by the rate of carrier generation due to illumination and the times of capture of holes from the valence band to deep acceptor levels, from which reverse ionization into the continuum is unlikely. Such capture occurs, firstly, by means of rapid capture of holes into small acceptor states (when emitting an acoustic phonon), followed by a „slow“ transition from these states to deep levels of the acceptor. The speed of such a process is determined by the slow relaxation of carriers in localized states and, according to [18], is units of nanoseconds. The second, faster process is the capture of holes when emitting an optical phonon. The time of such a process is from 10 to 70 ps at a temperature of > 20 K [19]. Thus, the time of capturing holes to deep levels is determined by transitions with the emission of an optical phonon.

Then we can write the balance equation for the concentration of holes in the valence band:

$$g = \frac{1}{\tau_0(p, T)}. \tag{7}$$

Here g is the rate of generation, and $\tau_0(p)$ is the emission time of an optical phonon, depending on the concentration of free carriers and temperature. Given that the rate of generation is constant, from the equation (7) we obtain a

relation linking the concentration of carriers in the zone and the temperature:

$$\tau_0(p, T) = \text{const} = \tau_0(p_0, T = 20\,\text{K}), \qquad (8),$$

where p_0 — the concentration of holes at a temperature of 20 K. The emission time of the optical phonon was calculated using the method developed in [19]. Values τ_1 and τ_2 were found from the expression (8) concentrations for each temperature value. Then the proportion of neutral mercury vacancies was calculated using the expression (6).

Neutral vacancies (A_2^0-centers) are involved in the formation of PL, which are formed when the hole is captured both on partially ionized (in nonequilibrium conditions of intense interband illumination) centers and on equilibrium A_2^{-1}-centers that arise due to a temperature other than zero. The contribution of these centers should be summed up. The number of equilibrium partially ionized mercury vacancies was calculated as a function of temperature in [16] (function $N_{A^{-1}P}(T)$). The calculated dependence of the number of neutral mercury vacancies with populated small excited levels should be multiplied by $1 + PN_{A^{-1}P}(T)$, while P is a fitting parameter that was selected for better agreement of the dependence of the population of small acceptor levels on temperature with the dependence of the intensity of the PL line due to these centers (see [16]). The degree of compensation (DC) is an additional fitting parameter. Recall that the structures under study have electronic conductivity. Figure 3 shows the dependences of the population of excited A_2^0-centers on temperature at different values of the parameter P and the degree of compensation. The best conformance of the form of the experimental dependence of the PL intensity on temperature and the model used was achieved at a compensation degree of 10% and at a value of $P = 1$, and the concentration of free holes $5 \cdot 10^{15}\,\text{cm}^{-3}$ at $T = 20\,\text{K}$.

It should be noted that the measured PL goes off faster with increasing temperature [16] than predicted by the calculations shown in Figure 3 predict. Apparently, this is due to a decrease in the number of free holes in the valence band unaccounted for in this model due to the capture of carriers at A_2^{-2}-centers, the concentration of which is high in the *n*-type material. Since the ionization energy of the A_2^{-2}-center slightly exceeds the maximum energy of the optical (CdTe-like) phonon, transitions of holes to the ground state of the A_2^{-2}-center with the emission of optical phonons are possible only from small localized levels. However, as shown in this paper, the occupation of these levels can occur quite quickly due to the emission of acoustic phonons. The difference from the situation discussed above is that transitions with the emission of an optical phonon between localized states are much faster than radiative ones. As a result, the accumulation of carriers in excited states and reverse ionization into the continuum will be weaker, which will lead to a noticeable decrease in the concentration of carriers in the valence band. The intensity of transitions with the emission of optical phonons will increase with increasing temperature, leading to additional quenching of PL.

4. Conclusion

In this paper, for different temperature values, the capture times of holes on small excited levels of neutral mercury vacancy are calculated, as well as the transition times of holes from small localized levels to the continuum of the valence band when emitting an acoustic phonon in a solid solution of $Hg_{1-x}Cd_xTe$. Due to the redistribution of carriers along the valence band with temperature, the time of carrier capture to localized levels of the neutral vacancy increases, and the time of departure to the continuum decreases. The model proposed on the basis of the calculation results satisfactorily describes the temperature quenching of PL observed in the experiment due to radiative transitions between localized states of holes on a neutral mercury vacancy.

Appendix

Expression for the probability of transition rates (probability of transitions per unit time) for piezoacoustic scattering during spontaneous phonon emission, according to [16], has the form

$$W_{\mathbf{k}\to\mathbf{k}'} = \frac{4\pi^2 e^4}{V}\left[\frac{1}{Msqa}\right]\delta_{\mathbf{k},\mathbf{k}'\pm\mathbf{q}}\delta(\varepsilon_1 - \varepsilon_f \mp \hbar sq). \quad (\Pi.1)$$

Here $\mathbf{k}$ and $\mathbf{k}'$ — quasi-wave vectors of the initial and final states of the hole in the valence band, $\mathbf{q}$ — phonon wave vector, $\varepsilon_{i,f}$ — the energies of the initial and final states, e — electron charge, s — speed of sound, a — lattice constant of solid solution $Hg_{1-x}Cd_xTe$. M — sum of the atomic masses of the unit cell (the mass of the mercury atom plus the mass of the tellurium atom), V — sample volume.

Then the expression for the free run time has the following form

$$\frac{1}{\tau} = \sum_{\mathbf{k}'} W_{\mathbf{k}\to\mathbf{k}'} = \frac{V}{(2\pi)^3}\int W_{\mathbf{k}\to\mathbf{k}'}\mathbf{d}^3\mathbf{k}. \qquad (\Pi.2)$$

It should be noted that the value turns out to be independent of the characteristics of the particle state (energy, quasi-pulse) and temperature. Substituting the constants characterizing the material, we get $\tau = 0.085\,\text{ps}$.

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Conflict of interest

The authors declare that they have no conflict of interest.

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