The effect of impurity and intrinsic defects on the energy structure and dynamic of electronic processes CdTe:V and $Cd_{1-x}Hg_xTe:V$ crystals

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Received September 30, 2007

The electron trapping and detrapping processes in the semi-insulating CdTe:V crystals have been studied using a new time-resolved photoelectric spectroscopy technique. It has been shown that the electron processes in such crystals involving the impurity centers and intrinsic defects are fast and occur in the nanosecond range. The information on the nature and energy structure of the anisotropic impurity centers has been obtained. Two different photogeneration mechanisms of free electrons have been revealed: the direct photoionization of electrons from the ground impurity states and their self-ionization from the excited impurity states which are in resonance with the conduction band. It was found that the photosensitivity region for $Cd_{1-x}Hg_xTe$ crystals (x=0.018) at 300 K is in the range from 0.9 to 1.7 μ m.

Исследованы процессы захвата и высвобождения электронов в полуизолирующих кристаллах CdTe:V и Cd $_{1-x}$ Hg $_x$ Te:V с использованием метода фотоэлектрической спектроскопии с временным разрешением. Показано, что электронные процессы в таких кристаллах с участием примесных центров и собственных структурных дефектов являются быстрыми и происходят в наносекундном диапазоне. Получена информация о природе и энергетической структуре анизотропных примесных центров. Установлено наличие двух механизмов фотогенерации электронов: прямая фотоионизация электронов из основного состояния примеси и их автоионизация из возбужденных состояний, находящихся в резонансе с зоной проводимости. Установлено, что область фоточувствительности кристаллов Cd $_{1-x}$ Hg $_x$ Te (x=0.018) при T=300 К охватывает спектральную область от 0,9 до 1.7 мкм.

One of the most urgent tasks of modern optoelectronics is the development of fast semiconductors, including the photorefractive IR-materials (0.9-1.7 $\mu m)$ [1-6]. Today, various materials are used for photodetectors in the near IR-region (Ge, PbS, InGaAs, PbSe, CdHgTe, etc.). The photosensitivity spectral region in such materials is defined by the band gap width of the crys-

tals. A drawback of those materials is the necessity for their cooling (cryogenic or thermoelectric). In the case of CdTe:V and $Cd_{1-x}Hg_xTe:V$ crystals, the photosensitivity is defined by the impurity photoconductivity [2, 3]. Therefore, these materials can be used to develop a new type of non-cooled IR-photodetectors. The materials are also of good promise for novel effective adaptive

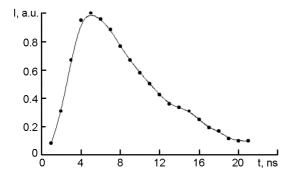


Fig. 1. The shape of pulse photocurrent normalized to the maximum of pulse amplitude for CdTe:V, $N_V=10^{19}~{\rm cm^{-3}}$ at 300 K at the electric field strength 3.3 kV/cm.

photodetectors [4]. It should be noted that the doped Cd_{1-x}Hg_xTe alloys, in contrast to commonly used photorefractive CdTe:V crystals, make it possible to tune the photoionization energy and expand the photosensitivity region up to 1.5 µm [3]. Since the CdTe:V and $Cd_{1-x}Hg_xTe:V$ crystals are $(\rho = 10^{7}$ semi-insulating materials $10^9 \ \Omega \cdot \text{cm}$), they can be considered as promising materials for the non-cooled detectors of X- and γ -rays. It is well known that the incorporation of 3d elements in A^{II}B^{VI} semiconductors results in formation of the strong localized electronic states and, respectively, to the appearance of deep impurity states [3-7]. The AIBVI semiconductors doped with vanadium, in particular, of the CdTe and ZnTe crystals as well as their $Cd_{1-x}Zn_xTe$ solid solution were studied in [1-7]. Basing on EPR measurement results, it was shown [7-10] that in the $Cd_{1-x}Zn_xTe$ crystals, anisotropic impurity centers can be formed in contrast to the binary compounds.

In this work, the complex optical and photoelectric studies of semi-insulating CdTe: V and Cd_{1-x}Hg_xTe: V crystals have been carried out with the aim to define the nature and energy structure of the deep impurity centers, including the anisotropic impurity centers, as well as their role in the electronic processes in the nanosecond range. The vanadium doped CdTe and $Cd_{1-x}Hg_xTe$ crystals were grown by the vertical Bridgman technique. Doping was provided by vanadium additive into the melt. The initial concentration of the impurity atoms was about 10^{19} cm⁻³. All studied samples had n-type conductivity. The timeresolved photoelectric spectroscopy measurements were carried out using the appara-

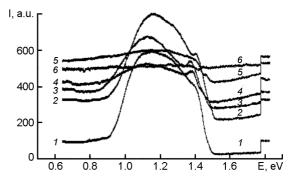


Fig. 2. Spectral dependences of the pulse photocurrent intensity at $t_d=5$ ns under additional stationary monochromatic illumination for CdTe:V crystal, $N_V=10^{19}~{\rm cm}^{-3}$ at 300 K. Curves 1-6 correspond to the electric field strength 3.3 kV/cm, 4.7 kV/cm, 5.3 kV/cm, 6.0 kV/cm, 6.7 kV/cm and 7.3 kV/cm, respectively.

tus and technique presented in [2]. The absorption and photodiffusion current (PDC) spectra were measured using a KSVU-23 spectrophotometer.

In a CdTe:V crystal excited by a nitrogen laser ($\lambda=337.1$ nm, t=9 ns, P=5 kW), free charge carriers are photogenerated in the near-surface layer. The stationary illumination of the crystal surface with the different wavelengths was carried out using a high-aperture MDR-12 monochromator. This makes it possible to measure the photocurrent spectra caused by an increase in the current pulse component due to detrapping of the charge carriers from the traps. If the illuminated electrode has negative polarity, mostly electrons move through the crystal. In this work, the electronic conductivity of crystals is investigated.

Fig. 1 shows the shape of photocurrent pulse (PPC) measured with the 0.1 ns time resolution in the absence of additional illumination. In this case, the PPC has the same shape as the laser pulse. Fig. 2 shows the intensity of the photocurrent pulse corresponding to the laser pulse maximum for the delay time $t_d=5\,\,\mathrm{ns}$, as a function of the stationary monoch "romatic illumination energy. The spectral dependence at different values of electric field strength $(F = (3.3 \text{ to } 7.3) \cdot 10^3 \text{ V/cm})$ was measured. The wide band with a maximum at 1.17 eV is due to the photoionization transitions involving the photoinduced V^{2+}_{ind} impurity centers as a result of the self-ionization of electrons from the excited ${}^4T_1(P)$ state being in resonance with the conduction band. This mechanism of photoionization of trapped electrons is the main one. The direct photoionization of electrons from the ${}^4T_1(F)$ state is less effective. It should be noted that when the laser pulse was off, the PPC is caused by the photoionization of V²⁺-centers was equal to zero, as the stationary photocurrent itself was not revealed.

The spectrum structure in its shortwavelength wing is associated with the photoionization transitions of electrons from the traps of the intrinsic defects. The results obtained indicate the presence of V^{3+} ions, neutral cadmium vacancies and acceptor complexes in CdTe:V crystals, which are the traps for the electrons photogenerated by a nitrogen laser pulse. The maximum of the broad band at 1.17 eV is due to the photoionization transitions of V^{2+} ions caused by the self-ionization of electrons from the ${}^{4}T_{1}(P)$ excited state to the conduction band. The trapping and detrapping of electrons photogenerated by a nitrogen laser pulse may be presented as:

$$\mathsf{V}^{3+} + e_{cb} \rightarrow \mathsf{V^{2+}}_{ind} \; , \tag{1}$$

$$({\rm V^- cd} + {\rm D^+})^0 + e_{cb} \rightarrow ({\rm V^{2-} cd} + {\rm D^+})^- \;_{ind} \; , \eqno(2)$$

$$V^{0}_{cd} + e_{ch} \rightarrow (V^{-}_{cd})_{ind}$$
, (3)

where V^{2+}_{ind} , $(V^{2-}_{Cd} + D^{+})^{-}_{ind}$ and $(V^{-}_{Cd})_{ind}$ are the photoinduced impurity and intrinsic defects appearing due to trapping of free electrons. The processes of the electron photorelease from the traps due to stationary monochromatic illumination can be presented as:

$$V^{2+}_{ind} + h\nu(0.90-1.35eV) \rightarrow V^{3+} + e_{cb},$$
 (4)

$$(V^{2-}_{cd} + D^{+})^{-}_{ind} + hv(1.41eV) \rightarrow (5)$$

 $\rightarrow (V^{-}_{cd} + D^{+})^{0} + e_{ch},$

$$(V^{-}cd)_{ind} + hv(1.45eV) \rightarrow V^{0}cd + e_{cb}.$$
 (6)

Consideration of the PPC spectra shape, intensity, and the energy position indicates that the electron trapping efficiency depends on the electric field strength. For $F>6.7 \text{ kV}\cdot\text{cm}^{-1}$, the broad impurity PPC band practically disappears. In this case, the electron trapping is absent.

As shown above (Fig. 2) the PPC shape and intensity is very sensitive to the IR illumination by the pulse Nd:YAIG laser (no additional infrared illumination taking place). The excitation energy (1.17 eV) cor-

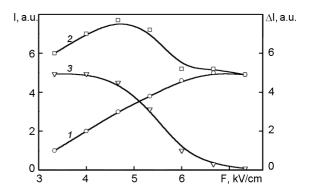


Fig. 3. Dependences of the pulse photocurrent intensity at $t_d=5$ ns on the electric field strength for CdTe:V, $N_V=10^{19}~{\rm cm}^{-3}$ at 300 K. Curve 1 and curve 2 correspond to the excitation by N₂ pulse laser only and the additional illumination by the Nd:YAIG laser, respectively. Curve 3 corresponds the difference between the PPC values (ΔI_{ppc}) presented by the curve 2 and curve 1.

responds to that the transitions from the ground to the ${}^4T_1(P)$ excited state of V^{2+} ions.

In Fig. 3, curve 1 corresponds to the PPC intensity variation as a function of the electric field strength in the absence of the additional illumination. The illumination by a pulse Nd:YAIG laser for low F values results in a sharp increase of PPC as is presented by curve 2. For F = 7.3 kV/cm, the PPC intensity is almost independent of the the electric field strength. Fig. 3 (curve 3) shows the difference between the intensity presented by curve 2 and the curve 1 and presents to a certain degree the change of the electron capturing efficiency in the deep impurity traps, i.e. V^{3+} ions as a function of the applied electric field strength.

It is obvious that the PPC intensity increase under pulse illumination with the energy 1.17 eV is caused by the detrapping of electrons from the deep impurity V^{2+}_{ind} centers. Curve 2 corresponds to the pulse photocurrent originated from two sources. The first source is the excitation of free electrons. The second one is the electron excitation from the ground state to the excited one and their self-ionization to the conduction band. The consideration of the dependences mentioned above evidences that the pulse photocurrent caused by the electron detrapping F>4.0 eV/cm. This is caused by the decrease of the electron capturing efficiency in the deep impurity traps $(V^{3+} ions)$ under such applied electric field. In this case, the concentration of V²⁺_{ind} centers de-

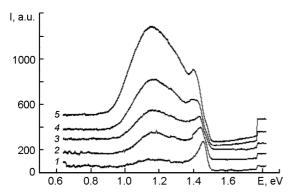


Fig. 4. Spectral dependences of the intensity of pulse photocurrent at $t_d=5$ ns under the different intensities of additional stationary monochromatic illumination for CdTe:V crystal, $N_V=10^{19}~{\rm cm^{-3}}$ at 300 K. Curves I-5 correspond to the different light intensity values equal to I_0 , $2I_0$, $8I_0$, $16I_0$ and $40I_0$, respectively.

creases, thus resulting in the decrease of the PPC intensity caused by the detrapping of electrons from these centers. Free electrons are not captured substantially by V^{3+} ions at $F \ge 6.7$ kV/cm and the V^{2+}_{ind} centers are not formed.

Fig. 4 shows the spectral dependences of the pulse photocurrent intensity for CdTe:V crystal at $t_d=5$ ns. Curves 1-5 correspond to different intensity values of the stationary illumination. At the lowest intensity, the broad impurity band is very weak and the band corresponding to the photoionization of $(V^-_{Cd})_{ind}$ centers becomes the most intense. The increase of the light illumination intensity (curves 2-5) results in decrease of PPC band at 1.45 eV as well as its sharp drop to 1.41 eV.

These results indicate that the electron detrapping efficiency from the traps differing in energy depends on the illumination intensity. At low intensity (curve 1 and curve 2), the electrons are trapped in the traps with energies $E_v + 0.05$ eV and $E_v +$ 0.06 eV. These energies correspond to the singly charged cadmium vacancies and possibly to the charged acceptor centers caused by the presence of the residual atoms such as Na or Li, respectively, in CdTe:V crystal [10]. The increasing light intensity (curve 5) results to the effective de-trapping of the electrons from the singly charged acceptor $(V^{2-}_{Cd} + D^+)^-_{ind}$ complexes with the energy $E_v + 0.09$ eV. In this case, the broad impurity band becomes the most intense. This may be caused by the higher concentration of the deep impurity traps, i.e. V^{3+} ions, in

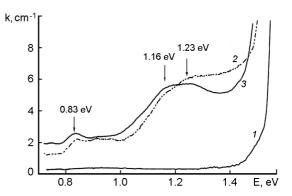


Fig. 5. Absorption spectra of CdTe (curve 1), CdTe:V (curve 2) and $Cd_{1-x}Hg_xTe:V$ (x = 0.014, curve 3) at 78 K.

comparison with other electron traps, namely, the neutral cadmium vacancies and the neutral acceptor complexes. The V³⁺ ion concentration in CdTe:V crystals usually is about $10^{16}~\rm cm^{-3}$ [8]. Therefore, the concentration of the neutral acceptors in the CdTe:V crystals must be lower than $10^{16}~\rm cm^{-3}$.

The absorption spectra of $Cd_{1-x}Hg_xTe$ crystals doped with vanadium as well as pure CdTe are shown in Fig. 5. A longwavelength shift of the absorption edge is observed with growing Hg atom concentration. In this case, the band gap of solid solution is narrower than in CdTe. Therefore, the photoionization energy of V²⁺ impurity centers will increase for the solid solution as compared to CdTe crystal due to decreasing conduction band bottom. The position of the absorption bands corresponds to the intracenter absorption of the V impurity ions in various charge states as well as to the photoionization spectra. The intracenter absorption near 0.8 eV is due to transitions between ${}^4T_1(F)$ and ${}^4A_2(F)$ states of V^{2+} ions [11]. The additional absorption bands at 0.91 and 0.97 eV indicate the existence of V^{2+} and V^{3+} ions [9]. The wide absorption band near 1.20 eV is due to both the intracenter transitions between the ${}^4T_1(F)$ and ${}^4T_1(P)$ states of V^{2+} impurity ions and their overlap with the photoionization absorption.

Information on the energy position of the levels of deep impurity centers and intrinsic defects relative to the crystal energy bands can be obtained by measuring the PDC spectra [1, 3]. In the PDC spectra of these crystals (Fig. 6), the positive bands are caused by photoionization transitions of electrons from impurity or defect levels to the conduction band, while the negative bands are

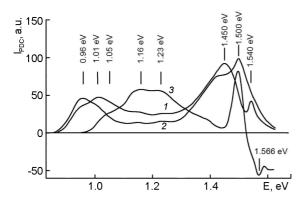


Fig. 6. PDC spectra of the $Cd_{0.986}Hg_{0.014}Te:V$ crystal (curves 1 and 2, the light propagation in the <111> and <110> directions, respectively) and CdTe:V (curve 3, <111> direction).

attributed to the excitation of valence-band electrons to discrete levels positioned in the crystal band gap [3]. In Fig. 6, there are the PDC spectra corresponding to the light propagation along the <111> and <110> directions in $Cd_{1-x}Hg_xTe$ crystals (curves 1, 2, respectively) as well as the PDC spectrum of CdTe:V crystal for <111> direction (Fig. 6, curve 3). The most intense band near 1.500 eV is caused by intrinsic defects of acceptor type in the investigated crystals. Such acceptors are the singly charged centers consisting of doubly charged cadmium vacancy (V^{2-}_{Cd}) and singly ionized donor (D⁺), i.e. the ($V^{2-}_{Cd} + D^+$)-centers. Their energy is in the range $E_V + (0.10$ to 0.12 eV) [12]. In [13], the complex centers which include the cadmium vacancy and donor associated with the Cl impurity at anionic sites were detected. In our case, the

ionized donors might be accidental impurities of Group III atoms (Ga, In, or Al) positioned at cationic sites in the <110> direction. The negative band at 1.566 eV is probably caused by the transition from the valence band to the donor level at E_c -0.035 eV formed by the singly ionized donor (vacancy of Te atom) [14].

When light propagates in the <111> direction, the PDC spectrum consists of two positive bands near 1.450 and 1.540 eV. The high-energy band at 1.540 eV is probably caused by electron transition from the acceptor level $E_V + 0.028$ eV [14] corresponding to a singly charged Na atom (residual impurity) positioned at a cationic site (Na-Cd) to the conduction band. The other band at 1.450 eV is caused by photoionization of a singly charged acceptor $(V^{2-}_{\mbox{Cd}} + \mbox{D}^+)^-$. When light propagates in the <110> direction, the intense 1.500 eV band is observed in the PDC spectrum (curve 2), which is observed in the thermostimulation conductivity spectra of the In doped CdTe crystals with atoms [15]. The PDC spectrum of the CdTe:V crystals also exhibits a broad structured band in the long-wavelength spectral range. The long-wavelength wing of the PDC band reveals an inflexion near 1.05 eV which is due to the direct photoionization energy of V^{2+} centers. This energy is close to the value (~1.0 eV) obtained by photoconductivity measurements [3]. The observed maxima of the PDC bands at 1.16 and 1.23 eV coincide with the energy positions of the broad absorption band in CdTe:V (Fig. 6, curve 2). This suggests that the PDC bands are caused by the excitation of

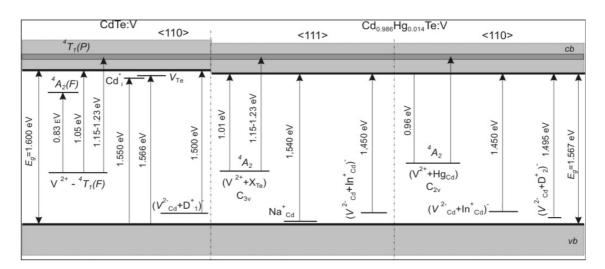


Fig. 7. Combined energy-level diagram showing $d-d^*$ intracenter transitions and ionization levels of impurity centers and intrinsic defects in CdTe:V and Cd_{0.986}Hg_{0.014}Te:V crystals.

 V^{2+} ions into the $^4T_1(P)$ state, which is in resonance with the conduction band. The double-peaked bands near 1.00 eV are probably caused by anisotropic V^{2+} centers of trigonal and orthorhombic symmetry in the $Cd_{1-x}Hg_xTe$ crystals. Those centers of C_{2v} symmetry may be caused by formation of the complex ($V^{2+}Cd + ZCd$) centers with axis oriented along one of the <110> directions.

In Fig. 7, the combined energy-level diagram is presented showing $d-d^*$ intracenter transitions and ionization levels of V impurity centres and intrinsic defects in these crystals. It should be noted that such scheme was constructed basing on the experimental results obtained at liquid nitrogen temperature. In our case, the PPC measurements were carried out at T = 300 K. The difference between the band gap energy of CdTe crystals at 77 K and 330 K equals to $0.10~{\rm eV}.$ Therefore, in investigated Cd_{1-x}Hg_xTe:V (x = 0.014) crystals, the impurity photoionization energy for the trigonal and orthorhombic centers will be 0.91 eV and 0.86 eV, respectively. Fig. 7 shows that the energy of intracenter transitions to the excited ${}^{4}A_{2}(F)$ state is 0.83 eV, i.e. somewhat lower than the direct photoionization enfor $_{
m the}$ orthorhombic centers (0.86 eV). Therefore, in this case, the excited ${}^{4}A_{2}(F)$ state is still within the band gap. However, for somewhat higher mercury concentrations, this state will be in the resonance with the conduction band. In this case, both the direct photoionization and the self-ionization from the ${}^4T_1(P)$ and ${}^4\!A_2(F)$ states to the conduction band occur. This makes it possibly to increase the crystal photosensitivity and to tune the spectral range. To that end, the PPC spectra of the $Cd_{1-x}Hg_xTe:V$ (x = 0.018) crystals have been studied in this work.

Fig. 8 shows the spectral dependence of pulse photocurrent for $Cd_{1-x}Hg_xTe:V$ (x =0.0183) crystal at different delay times (curves 1-3 for 4.5; 0.4, and 7.0 ns, respectively). At $t_d = 4.5$ ns and 0.4 ns, the shape of PPC spectra is the same. The maximum of PPC spectra corresponds to $\lambda = 1080 \text{ nm}$ (1.15 eV), similar to the CdTe: V crystals; that is associated with the electron selfionization from the excited ${}^4T_1(P)$ states of both the orthorhombic and trigonal centers. In the short-wavelength wing, two specific features are revealed as inflexions ($\lambda = 900 \text{ nm}$ and 970 nm). The first band corresponds to the photoionization of singly charged cadmium vacancies (V-Cd) while the other one, to that of acceptor complexes $(V^{2-}_{Cd} + D^{+})^{-}$,

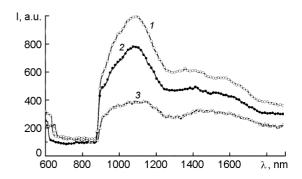


Fig. 8. Spectral dependences of the pulse photocurrent intensity at $t_d=4.5$ ns, 0.4 ns and 7 ns (curves 1--3, respectively) under additional stationary monochromatic illumination for Cd $_{1\text{--}x}$ Hg $_x$ Te:V $(x=0.0183,\ N_V=10^{19}\ \mathrm{cm}^{-3})$ crystal at $T=300\ \mathrm{K}.$

i.e. the nature of the intrinsic defects is the same for both the CdTe:V and the $Cd_{1-x}Hg_xTe:V$ crystals. The long-wavelength wing is extended up to $\lambda = 1700 \text{ nm}$ (0.73 eV). This wing also reveals the structure of PPC spectrum at 1400 nm and 1550 nm wavelengths. This structure is caused by the resonance of the excited ${}^{4}A_{2}(F)$ -state with the conduction band. At the delay time $t_d=7\,\mathrm{ns},$ the PPC spectrum structure (Fig. 8, curve 3) is less pronounced. This situation is caused, in particular, by the decreased intensity of bands as a result of the lowered intensity of the pulse photocurrent at $t_d = 7$ ns. In this case, the self-ionization of electron from the excited ${}^4A_2(F)$ state takes place for the orthorhombic centers only. As to the trigonal centers, such excited state is in the band gap. Therefore, the optical transitions from the ${}^4\!A_2(F)$ state will occur involving several LO-phonons only, and the probability of such photoionization transitions is very low.

It has been shown that the electronic processes in the semi-insulating (ρ - $10^8\Omega$ ·cm) CdTe:V crystals involving the impurity centers and intrinsic defects are fast and occur in the nanosecond range. The nature and the energy positions of defects in the band gap, which act as traps for free electrons photogenerated by the laser pulse have been established. The relations describing the electron trapping and detrapping mechanisms in the traps are presented. In particular, it has been shown that the electron trapping causes the formation of the photoinduced impurity V²⁺ and the acceptor (V-Cd)_{ind} as well as (V²-Cd + D⁺)- $_{ind}$. The electron trapping efficiency by the deep impurity centers and intrinsic defects de-

pends from the strength of applied electric field and monochromatic stationary illumination. For the vanadium doped $Cd_{1-x}Hg_xTe$ crystals, the information on the nature and energy structure of anisotropic impurity centers and intrinsic defects has been obtained. The combined energy-level diagram showing $d-d^*$ intracenter transitions and ionization levels of V impurity centers and intrinsic defects is presented. The photosensitivity region of such crystals which is defined both by the direct impurity photoionization and the autoionization of electrons from the excited impurity states to the conduction band is extended up to $\lambda = 1700$ nm. The influence of the anisotropic impurity centers on the fast electronic processes in such materials has been established.

Acknowledgment. This work has been supported by Grant CRDF No.UKE2-2855-KV-07.

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Вплив домішкових і власних дефектів на енергетичну структуру та динаміку електронних процесів кристалах CdTe:V та $Cd_{1-x}Hg_{1-x}Te:V$

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Досліджено процеси захвату та звільнення електронів у напівізолюючих кристалах CdTe:V та $Cd_{1-x}Hg_xTe:V$, використовуючи методику фотоелектричної спектроскопії із розділенням за часом. Показано, що електронні процеси у таких кристалах із участю домішкових центрів та власних структурних дефектів є швидкодіючими і відбуваються у наносекундному діапазоні. Отримано інформацію щодо природи та енергетичної структури анізотропних домішкових центрів. Встановлено наявність двох механізмів фотогенерації електронів: пряма фотоіонізація електронів із основного стану домішки та їх автоіонізація із збуджених станів, що знаходяться у резонансі із зоною провідності. Встановлено, що область фоточутливості кристалів $Cd_{1-x}Hg_xTe:V$ (x=0.018) при T=300 K охоплює спектральну область від 0.9 до 1.7 μm .