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(41, Nauky Ave., Kyiv 03028, Ukraine; e-mail: vvvit@ukr.net)**DEPENDENCE OF THE CdTe MELTING
THRESHOLD ON THE PULSE DURATION
AND WAVELENGTH OF LASER RADIATION
AND THE PARAMETERS OF NON-EQUILIBRIUM
CHARGE CARRIERS**PACS 81.15.Fg, 81.65.Cf,
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The dependences of the melting threshold of CdTe under the pulsed laser irradiation on the radiation wavelength λ and the laser pulse duration τ_p are calculated with regard for the non-equilibrium character of charge carriers. Three components of the energy released at the thermalization of excited carriers under the nanosecond laser irradiation of CdTe in the fundamental absorption region are considered: the component that dominates immediately after the excitation, and the components released at the nonradiative bulk and nonradiative surface recombinations. Together, they determine the depth of heat penetration into the crystal and, therefore, its melting threshold. It is shown that the CdTe melting threshold grows from 2.6 to 4.75 MW/cm², when λ changes from 300 to 800 nm at $\tau_p = 20$ ns. The changes in the non-equilibrium charge carrier parameters (the surface recombination rate, lifetime, and diffusion depth) are found to vary the CdTe melting threshold by at least 30%.

Keywords: cadmium telluride, pulsed laser irradiation.

1. Introduction

Cadmium telluride is a base material for the creation of detectors of x-rays and gamma radiation, as well as infrared sensors. Special attention to CdTe is associated, in particular, with large atomic numbers of its constituting elements and a considerable transverse cross-section for the photo-electric absorption [1–5].

In the formation of CdTe-based barrier structures to detect gamma radiation, one of the effective methods to treat the surface is the application of a nanosecond laser irradiation. In some cases, it allows the required parameters of the surface state to be optimized at the stage of technological treatments, when contacts (Ag, In, Au) are deposited and a barrier is formed [2–4]. A challenging problem also concerns the formation of ordered nanostructures on the CdTe surface under a pulsed laser irradiation (PLI) [6].

For the PLI of semiconductors and semiconductor-based structures, the laser radiation with various

pulse durations τ_p and various radiation wavelength τ_p is used. Ruby ($\lambda = 694$ nm) and neodymium (the first, $\lambda = 1064$ nm, and second, $\lambda = 532$ nm, harmonics) lasers, as well as excimer KrF ($\lambda = 248$ nm) and XeF ($\lambda = 351$ nm) ones, are mainly applied to modify the surface. Typical values of the parameter τ_p for them are 7, 20, 80, 120 ns, and others. Therefore, in this work, we are interested first of all in the pulse duration interval $\tau_p = 5\div 120$ ns.

While solving the problems of this kind, it is often required to know—or at least to estimate in advance—the melting temperature and the melting threshold I_{th} of a CdTe surface subjected to PLI, and their dependences on the laser pulse duration τ_p , radiation wavelength (the optical absorption coefficient $\alpha(\lambda)$), and electrophysical parameters of the CdTe surface, in particular, the surface recombination rate S and the lifetime of nonequilibrium charge carriers (NECs). Those parameters depend on the surface treatment procedure [7] and the concentration of doping impurities. Relevant experimental researches are very labor- and time-consuming. Therefore, the theoretical calculation of I_{th} and the surface temperature

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as functions of τ_p , $\alpha(\lambda)$, and other parameters becomes very useful.

The literature sources contain mismatches concerning the values of CdTe melting threshold and, accordingly, various interpretations of changes in the photo-electric properties under a nanosecond laser irradiation, provided identical experimental conditions. For example, in works [8,9], the melting threshold for the CdTe(111) face under the irradiation using a ruby laser with $\tau_p = 20$ ns was experimentally and theoretically found to equal 2 MW/cm^2 (40 mJ/cm^2). Under the same conditions for the CdTe (111) surface, $I_{\text{th}} = 8 \text{ MW/cm}^2$ in our work [10], 4 MW/cm^2 in work [11], and $10\text{--}12 \text{ MW/cm}^2$ in work [12]. In researches [13–15] carried out for CdTe(111), I_{th} was evaluated to equal 300 mJ/cm^2 (15 MW/cm^2). Under the irradiation with an excimer KrF laser ($\lambda = 248 \text{ nm}$, $\tau_p = 20 \text{ ns}$), $I_{\text{th}} = 2.5 \text{ MW/cm}^2$ [3, 4, 16].

2. Calculation Formulas for the Melting Threshold

Certainly, the rapid (tens or hundreds of nanoseconds) process of heating-cooling of the CdTe surface under a nanosecond laser irradiation is rather complicated for a mathematical description with regard for the temperature dependences of the thermal parameters and the concentration of photo-excited NECs. The threshold value for the laser-induced melting of the crystal surface is determined first of all by the heat diffusion depth $L_H = \sqrt{k\tau_p}$ and the light absorption depth $d \sim \alpha^{-1}$, where α is the absorption coefficient, and k the thermal diffusivity coefficient. The melting threshold for a semiconductor surface under PLI is theoretically determined, as a rule, by solving a non-stationary equation of thermal conductivity with a corresponding boundary condition across the phase interface (the Stefan problem).

The mathematical modeling of phase transitions initiated in cadmium telluride by a pulsed laser radiation on the basis of the heat conductivity equation [4, 11, 16, 17] is complicated due to the account for those parts of the optical radiation energy that are spent on the instant thermalization ($\tau \approx 10^{-12} \text{ s}$) and the excitation of the electron-hole plasma followed by its diffusion and recombination. In work [18], this was done by making allowance for the energy components that are released at the thermalization of

excited charge carriers immediately after their excitation and at the nonradiative bulk and surface recombinations. Taken together, those components determine the depth of heat penetration into the crystal and, accordingly, the melting threshold. In other words, it is the “semiconductor” crystal parameters that are taken into consideration. Therefore, the theoretical calculation of the laser-induced heating process should provide a detailed account for the optical parameters, the NEC and heat diffusion coefficients, and the variation of those parameters with the NEC concentration and the temperature. In the case of uniform irradiation of the surface of a semiinfinite crystal, the power density I that is required to heat up the surface from the initial temperature T_0 to the final temperature T_f is described by the following well-known formula (see, e.g. works [18, 19]):

$$I_0 = \frac{\rho c_P(T_0)\Delta T L_H}{[1 - R(T_0)]\tau_p}, \quad (1)$$

where ρ is the density, c_P the specific heat, R the optical reflectance, τ_p the laser pulse duration, and $\Delta T = T_f - T_0$. Unlike the simple expression $L_H \sim \sqrt{k\tau_p}$, the depth of heat penetration into a material in formula (1) is given by the following expression, in which the NEC parameters have already been taken into account [18]:

$$L_H = \frac{1 - R(T_0)}{c_P(T_0)\Delta T} \int_{T_0}^{T_f} \frac{dT c_P}{(1 - R)\alpha} [\chi_T / (L_T\alpha + 1) + \chi_B^{\text{NR}}(L_T\alpha + L_D\alpha + 1) + \chi_S^{\text{NR}} / (L_T\alpha)]^{-1}, \quad (2)$$

where $\alpha = \alpha_1 + \alpha_2 + \alpha_{FC}$ is the coefficient of total optical absorption; α_1 and α_2 are the coefficients of one- and two-photon, respectively, interband absorption; α_{FC} the absorption coefficient for free charge carriers; $L_D = (D\tau_B)^{1/2}$ is the diffusion depth for free charge carriers;

$$D = \frac{k_B T}{e} \frac{\mu_n \mu_p}{\mu_n + \mu_p}$$

is the coefficient of ambipolar diffusion for charge carriers; k_B the Boltzmann constant; τ_B the lifetime of charge carriers at their recombination in the crystal bulk; μ_n and μ_p are the mobilities of electrons and holes, respectively; and

$$L_T(T) \approx \pi^{1/2} [k(T)\tau_p]^{1/2} (T - T_0) / \Delta T.$$

The parameters χ_T , χ_B^{NR} , and χ_S^{NR} mean those components of the laser radiation energy that are released at the thermalization of excited charge carriers immediately after their excitation at nonradiative bulk and surface recombinations, respectively:

$$\chi_T = \left\{ \left[1 - \frac{\eta_Q E_R}{h\nu} \right] \alpha_1 + \left[1 - \left(\frac{E_R}{2h\nu} \right) \right] \alpha_2 + \alpha_{FC} \right\} / \alpha, \quad (3)$$

$$\chi_B^{NR} = (\tau / \tau_B^{NR})(1 - \chi_T), \quad (4)$$

$$\chi_S^{NR} = (\tau / \tau_S^{NR})(1 - \chi_T) L_D \alpha / (L_D \alpha + 1), \quad (5)$$

where η_Q is a dimensionless parameter equal to the ratio between the photon energy that is thermalized at the absorption depth $1/\alpha$ and the energy of NEC recombination at the diffusion depth, $h\nu$ is the energy of a photon, E_R the recombination energy, $\tau = (\tau_B^{-1} + \tau_S^{-1})^{-1}$ is the effective time of charge carrier recombination, $\tau_S = L_D/S$ is the effective lifetime of NECs at the surface, S the surface recombination rate, and τ_B^{NR} and τ_S^{NR} are the lifetimes of NECs at their nonradiative recombination in the bulk and at the surface, respectively.

3. Results and Their Discussion

In expressions (1)–(5), the temperature dependences of thermal quantities and NEC parameters have to be taken into account. The corresponding data for CdTe are presented in Table, and the dependences $\alpha(\lambda)$ and $R(\lambda)$ are shown in Fig. 1. The temperature dependence of the absorption coefficient arises due to the presence of charge carriers at the surface and is described rather well by the approximation formula $\alpha(\lambda) = \alpha_0 + A(\lambda)T$. In particular, at $\lambda = 694$ nm, we have $\alpha = (3 \times 10^6 + 8400T) \text{ m}^{-1}$. Despite an insignificant two-photon absorption in the interval of fundamental absorption, this dependence will be used below while making calculations in a wavelength interval of 300–800 nm. In a vicinity of the melting temperature, the parameter α becomes approximately 2.5 times as large. We also have to take into consideration the temporal dependence of the reflectance R : according to the results of work [20], this parameter changes from 0.28 to 0.36, when I varies from 0 to 0.2 J/cm^2 , owing to the start of the melting. In general, the value of R affects the magnitude of I_{th} rather substantially.

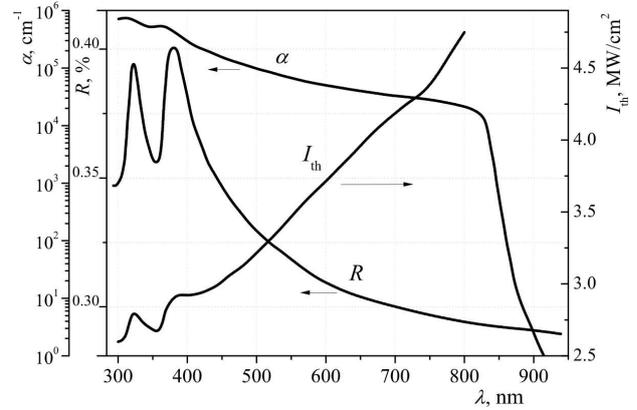


Fig. 1. Calculated dependence of the melting threshold on the laser wavelength, $I_{th}(\lambda)$, at $\tau_p = 20$ ns and experimental dependences $R(\lambda)$ and $\alpha(\lambda)$ for CdTe [22, 24]

It is important to know the NEC lifetime at the surface and in the bulk of CdTe under PLI. In the general case, the NEC lifetime in the CdTe bulk is given by the expression

$$1/\tau_B = 1/\tau_{SR} + 1/\tau_{Rad} + 1/\tau_{Aug},$$

where τ_{SR} , τ_{Rad} , and τ_{Aug} are the lifetimes of charge carrier recombination driven by the Shockley–Read, radiative, and Auger mechanisms, respectively. At high levels of NEC excitation, which takes place at our laser power densities, the Auger recombination [4] dominates. Accordingly, the charge carriers lifetime $\tau_B \approx \tau_{Aug} = 1/\gamma_{Aug}n^2$, where n is the NEC concentration. On the other hand, the lifetime can be divided into the radiative and nonradiative components, as in expressions (3)–(5):

$$1/\tau_{B(S)} = 1/\tau_{B(S)}^{Rad} + 1/\tau_{B(S)}^{NR}.$$

At the same time, bearing in mind that the radiative recombination is low and does not contribute to the lattice heating, the first term can be neglected. As a result, $\tau_B = \tau_B^{NR}$ and $\tau_S = \tau_S^{NR}$. Hence, under the PLI conditions, the NEC lifetime in the CdTe bulk is mainly governed by the Auger recombination, whereas the NEC lifetime at the CdTe surface by the surface recombination rate S .

In our calculations, we used the typical parameters of the detector high-resistance γ -CdTe (Acrorad Co. Ltd) (see Table) [2–5]. At the nanosecond laser irradiation, the measured NEC lifetime amounts to 3×10^{-8} s [5]. The parameter η_Q demonstrates the

Physical parameters of CdTe

Parameter	Value
Density ρ_0 , kg/m ³	5860
Melting temperature, °C	1092
Specific heat c_P , J/(kg·K)	209
Temperature conductivity k , m ² /s	$7.1(5.8) \times 10^{-6}$ [22]
Temperature dependence of density, kg/m ³	$5887 - 0.1165 \times T$ [21]
Temperature dependence of specific heat c_P , J/(kg·K)	$205 + 3.6 \times 10^{-2} \times T$ [16, 17, 22]
Temperature dependence of temperature conductivity k , m ² /s	$2.16 \times 10^{-3}/T$ [22]
Temperature dependence of heat conductivity, J/(m·s·K)	$1507/T$ [16, 17, 22]
Optical and electronic parameters of CdTe at $\lambda = 694$ nm (ruby laser)	
Reflectance R	0.28 [20, 24]
Absorption coefficient α , m ⁻¹	3×10^6 [17, 24]
Coefficient of NEC ambipolar diffusion D , m ² /s	3.9×10^{-4} (300 K), 5.7×10^{-4} (1000 K)
NEC bulk lifetime τ_B (for p -CdTe), s	$(10-30) \times 10^{-9}$ [5, 23]
NEC surface lifetime τ_S , s	$(0.5-1) \times 10^{-9}$ [5, 23]
Constant of Auger recombination γ_{Aug} , cm ⁶ /s	1.7×10^{-30} [22, 23]
Rate of surface nonradiative recombination S , m/s	10^3-10^5 [1, 5, 7, 23]
Dimensionless parameter η_Q	0.3-0.78
Recombination energy E_R , eV	1.5
Electron mobility μ_n , cm ² /(V·s)	1000-1100 (300 K) [1, 22]
Hole mobility μ_p , cm ² /(V·s)	80-100 (300 K) [1, 22]
NEC diffusion depth L_D (for p -CdTe), m	$(0.4-2) \times 10^{-6}$ [23]

ratio between the energy of photons that was immediately thermalized at the absorption depth $1/\alpha$ and the energy of photons that was transferred to NECs and released at the recombination at the NEC diffusion depth L_D (the recombination energy). The parameter η_Q is a little uncertain for CdTe under PLI (see Table). However, the calculations showed that, when it changes from 0.3 to 0.78, the magnitude of I_{th} changes by a factor of 1.08. As a rule, η_Q is close to 1 for some semiconductors (InSb, Si) [18].

Figure 1 demonstrates the results of calculations for the CdTe melting threshold under PLI within a wavelength interval of 300–800 nm and, for descriptive reasons, the literature data for $R(\lambda)$ and $\alpha(\lambda)$ on the logarithmic scale. The calculation was performed, by using formulas (1) and (2) and taking the data from Table into account. At $\lambda > 800$ nm – especially, in the interval of CdTe transparency ($\lambda > 850$ nm) – two-photon absorption and, accordingly, the defect and doping impurity concentrations, which strongly affect I_{th} , have to be taken into account. Usually, the coefficient of two-photon absorption in CdTe $\beta = 0.1 \div 0.2$ cm/MW (at $\lambda = 1.064$ μ m).

One can see that, when λ changes from 380 to 800 nm, the reflectance R changes from 0.4 to 0.29, i.e. it increases by a factor of 1.38. At the same time, the absorption coefficient α changes from 4.4×10^5 to 2.1×10^4 cm⁻¹, i.e. it decreases by more than an order of magnitude. It is evident that, if only the reflectance R decreases, the melting threshold I_{th} also decreases [formula (1)]; but if only α decreases, I_{th} increases, because a larger volume in the near-surface CdTe layer is heated up. Therefore, the dependence $\alpha(\lambda)$ is responsible for the behavior of the dependence $I_{th}(\lambda)$. In an interval of 300–420 nm, the curve $R(\lambda)$ has two peaks, where the variation of R amounts to 5%. The parameter a also has two small peaks in this interval. They are associated with the electron transition into minimum L in the conduction band ($L_{4.5}-L_6$ and L_6-L_6 transitions). Such a characteristic behavior of the curves $R(\lambda)$ and $\alpha(\lambda)$ in the indicated interval reveals itself in the dependence $I_{th}(\lambda)$, where a maximum and a minimum are observed (Fig. 1). Hence, when the wavelength of a pulsed laser radiation changes from 300 to 800 nm, the melting threshold grows from 2.6 to 4.75 MW/cm², i.e. it almost doubles.

Below $\lambda = 300$ nm, the absorption coefficient in CdTe is very high, and light is absorbed at a depth of about 10 nm. For example, for an excimer KrF laser ($\lambda = 248$ nm, $\tau_p = 20$ ns, $\alpha = 1.1 \times 10^6$ cm $^{-1}$), the melting threshold $I_{th} = 2.5$ MW/cm 2 [3, 4, 16]. On the other hand, according to our calculations, $I_{th} = 2.71$ MW/cm 2 , which is quite a reasonable value, because the reflectance R has rather a high value of 0.46 at $\lambda = 248$ nm [22].

Also important is the dependence of the melting threshold on the pulse duration τ_p . This dependence is depicted in Fig. 2. One can see that the power density of the melting threshold falls down rather drastically with the growth of τ_p [in accordance with formulas (1) and (2)]. The corresponding depth and, accordingly, volume of the heated-up layer increase at that. If the pulse duration becomes smaller, the rate of energy delivery becomes higher, and the processes of NEC thermalization and heat transfer into deeper layers owing to the heat conductivity become slower.

The dependence in Fig. 2 provides information about the melting threshold change at the variation of the pulse duration from 7 to 120 ns, because typical lasers operating in the Q -switching mode have a pulse duration of 7, 15, 20, 80, or 120 ns. The plot in Fig. 2, *b* demonstrates the dependence concerned in a wide τ_p -interval from 2 ns to 1.2 μ s on the log-log scale. The growth of τ_p by six orders of magnitude stimulates the decrease of I_{th} by three orders of magnitude from 20 MW/cm 2 to 10 kW/cm 2 .

At $\tau_p = 80$ nm and $\lambda = 694$ nm, the theoretical value of I_{th} for CdTe equals $I_{th} = 1.9$ MW/cm 2 (Fig. 2, *a*). The corresponding experimental value equals 1.5 MW/cm 2 [2], i.e. an agreement takes place. In work [17], the theoretical value for I_{th} at $\tau_p = 100$ ns equals 0.6 MW/cm 2 (60 kJ/cm 2); the calculations take into account the cadmium evaporation, owing to which CdTe begins to melt at a certain depth from its surface. Furthermore, according to our calculations, the CdTe melting threshold equals 9.08 kW/cm 2 at the ruby laser pulse duration $\tau_p = 1.2$ ms. This value is close enough to a value of 4.2 kW/cm 2 (5 J/cm 2), which was obtained theoretically and experimentally in work [25].

One can see that, within the pulse duration interval from 5 to 100 ns, I_{th} changes from 10.5 to 1.46 MW/cm 2 , i.e. by a factor of seven. At $\tau_p = 20$ ns, we have $I_{th} = 4.17$ MW/cm 2 . When the pulse duration changes from 15 to 25 ns, the value of

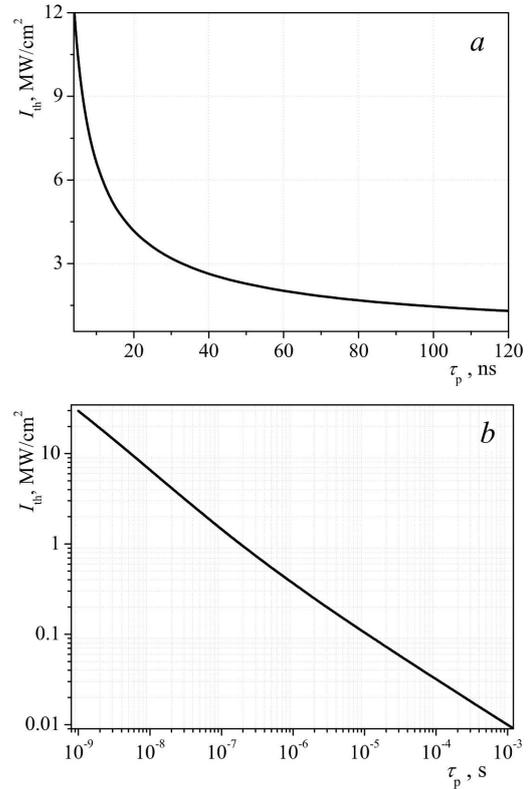


Fig. 2. Dependence of the CdTe melting threshold pulse duration τ_p : $\tau_p < 120$ ns (*a*) and $\tau_p < 1.2$ ms (*b*). $\lambda = 694$ nm

I_{th} changes from 5.05 to 3.6 MW/cm 2 . This means that the variation of the pulse duration from 15 to 25 ns is accompanied by the relative change of the power density $\Delta I_{th}/I_{th}(20 \text{ ns}) = (5.05 - 3.6)/4.17 = 0.35$. This fact is important to be taken into consideration, while treating semiconductors, when τ_p can change a little during a long time interval of the laser application.

Note that, if the pulse duration becomes shorter than 1 ns, the electric field strength grows strongly, so that such processes as ablation, ionization, laser-induced breakdown of vapor over the surface, and so forth start to play a significant role.

The calculation of the CdTe melting threshold by formulas (1) and (2) for a ruby laser with $\tau_p = 20$ ns gave the values $I_{th} = 4.17$ MW/cm 2 (Fig. 3) and $L_H = 0.33$ μ m. One can see that the dependence $T(I)$ is nonlinear due to the strong temperature dependences of the thermal parameters c_p , χ , and ρ , which change by tens of percent, when T grows from 0 $^{\circ}$ C to 1092 $^{\circ}$ C (Table). As a result, the value of I_{th}

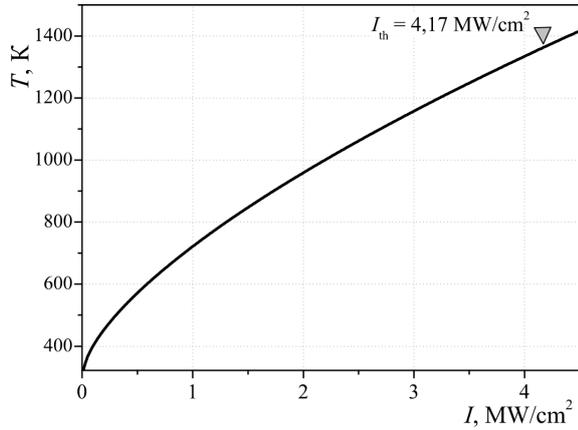


Fig. 3. Dependence of the CdTe surface temperature on the pulse power density of a ruby laser at $\tau_p = 20$ ns

becomes strongly dependent on T . The evaluation of I_{th} according to expression (1), at $L_H = \sqrt{k\tau_p}$, and assuming the thermal parameters to be independent of T gives rise to a value of 6 MW/cm^2 .

Electronic and optical parameters of the surface will be different a little, depending on the specific kind of a surface treatment: cleavage, polishing, etching, annealing in a definite atmosphere, and so on [7]. The change of NEC parameters (τ_B , S , L_D), provided the definite kind of a surface treatment, was found to affect the magnitude of I_{th} as follows. The growth of the surface recombination rate S from 10^3 to 10^5 m/s and the corresponding change of the surface recombination time τ_S^{NR} result in the variation of I_{th} by 5%. The growth of L_D from 0.4 to $2 \mu\text{m}$ stimulates the increase of I_{th} by 19%. This is so, because, in our case, the term χ_S^{NR} [Eq. (5)] in the integrand—this term includes the parameters of the surface recombination—is smaller than the term χ_T [Eq. (3)], and τ_B^{NR} is larger than τ_S^{NR} by an order of magnitude.

Therefore, a certain mismatch in the literature for the values of CdTe melting threshold under the nanosecond laser irradiation can be associated with the application of different kinds of a surface treatment and different (doped/undoped) CdTe crystals, because I_{th} demonstrates its dependence on the NEC parameters.

4. Conclusions

The CdTe melting threshold is calculated as a function of the pulsed laser irradiation wavelength λ and the pulse duration τ_p . It is shown that the

melting threshold I_{th} at $\tau_p = 20$ ns changes from 2.6 to 4.75 MW/cm^2 , when λ grows from 300 to 800 nm . Again, I_{th} changes from 10.5 to 1.46 MW/cm^2 , when the duration of a ruby laser pulse increases from 5 to 100 ns. The theoretically calculated values agree well with literature experimental data.

It is shown that the variation of the ruby-laser pulse duration within a typical interval of 20 ± 5 ns induces the relative change $\Delta I_{th}/I_{th} = 35\%$ of the melting threshold. This fact has to be taken into account when applying PLI of the CdTe surface in various technological tasks. It is found that the increase of the surface recombination rate S from 10^3 to 10^5 m/s changes the melting threshold I_{th} by 5%. The increase of the NEC diffusion depth L_D from 0.4 to $2 \mu\text{m}$ under the ruby-laser irradiation changes I_{th} by 19%. Finally, the modification of η_Q from 0.3 to 0.78 changes I_{th} by 8%. Hence, the variation in the NEC parameters can affect the CdTe melting threshold by at least 30%.

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ЗАЛЕЖНІСТЬ ПОРОГА ПЛАВЛЕННЯ CdTe
ВІД ТРИВАЛОСТІ ІМПУЛЬСУ ТА ДОВЖИНИ ХВИЛІ
ВИПРОМІНЮВАННЯ ЛАЗЕРА І ПАРАМЕТРІВ
НЕРІВНОВАЖНИХ НОСІЇВ ЗАРЯДУ

Р е з ю м е

Розраховано поріг плавлення CdTe в залежності від довжини хвилі випромінювання $I_{th}(\lambda)$ та тривалості лазерного імпульсу $I_{th}(\tau_p)$ з урахуванням параметрів ННЗ. При наносекундному лазерному опроміненні CdTe в області фундаментального поглинання враховано три компоненти енергії, що виділяється при термалізації збуджених носіїв – відразу після збудження (1), при безвипромінювальній об'ємній (2) та безвипромінювальній поверхневій (3) рекомбінації, які в сукупності визначають глибину проникнення тепла у кристал і відповідно поріг плавлення. Показано, що поріг плавлення змінюється від 2,6 до 4,75 МВт/см² при зростанні λ від 300 нм до 800 нм при $\tau_p = 20$ нс. Виявлено, що зміна параметрів ННЗ – швидкості поверхневої рекомбінації, часу життя, глибини дифузії може змінити поріг плавлення CdTe принаймні на 30 відсотків.