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Elastic waves induced by pulsed laser radiation in a semiconductor: effect of the long-range action

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Abstract
We observed the photosensitivity increase and the change of electrical properties of CdMnTe crystals which were photo-excited by pulsed ruby laser radiation ($\lambda = 0.694$ μm, $t_L = 20$ ns) with pre-threshold energy density. Changes took place not only in the irradiated part of the samples investigated, but also outside. Analysis of the heat diffusion and the point defect diffusion has shown that these factors cannot be responsible for the laser effect of the long-range action. It could be connected with propagation of the surface elastic wave induced by pulsed laser irradiation. The possible mechanism of the optical excitation of the elastic wave in the semiconductor was analysed. It was determined that decreasing of the energy of the point defects generation takes place as a result of the laser processing.

1. Introduction

High photo-excitation by laser radiation is a good tool for driving materials into non-equilibrium states and for material properties transformation. Now laser technology has proved to be a powerful method in the area of material processing and characterization [1, 2]. It has been shown previously that the application of pulsed laser irradiation to photoconductive semiconductors has resulted in the crystal photosensitivity increasing, modifying their electrical properties and dislocation structure [3–5]. Pulsed laser treatment shows promise as an effective procedure for cleaning and well ordering the crystal surface along. However, the change of the crystal parameters occurs not only in the irradiated part of samples, but also outside [4]. This phenomenon signifies the unpredictability of laser treatment and requires investigation.

The preliminary analysis has shown the connection between this so-called effect of the long-range action and propagation of surface elastic waves induced by the action of pulsed laser radiation. In this paper, we present the result of an investigation into the semiconductor crystal properties change as a consequence of laser processing. We also discuss the possibility of material properties change outside the irradiated part of the crystal. In this connection the role of the surface elastic wave, which is optically excited in a solid due to the intrinsic absorption of the short duration light pulse available from a Q-switched laser, is discussed. We have analysed the wave generation in GaAs and CdTe single crystals as a result of the thermal expansion of the crystal lattice and of the change in the equilibrium density of the substance during a process of photogeneration and recombination of non-equilibrium carriers.

2. Experimental observation of the photo-stimulated long-range effect

The object of the investigation was the diluted magnetic semiconductor $Cd_{1-x}Mn_xTe$ (x = 8–10%) grown from the melt using the Bridgman method. The dimensions of the single crystal samples were $6 \times 4 \times 1$ mm^3. The $Cd_{1-x}Mn_xTe$ samples used for measurements were n-type with a resistivity of 5–25 kΩcm at a temperature of 300 K. The investigated surface was mechanically polished and etched in a KOH methanol solution. After the etching, the samples were washed in distilled water.

A Q-switched laser source ($\lambda = 694$ nm) with a typical Gaussian pulse of $t_L = 20$ ns was employed for irradiation of the samples. The energy density in a light pulse E_L did not

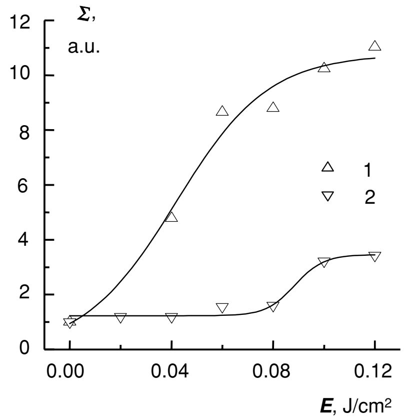

Figure 1. Dependence of the integral photosensitivity on the energy density of the laser irradiation for the irradiated (curve 1) and protected (curve 2) parts of the sample of the typical $Cd_{0.9}Mn_{0.1}Te$ crystal investigated. The solid lines are a guide to the eye.

exceed 0.12 J cm^{-2} and was below the threshold of damage or melting of the material. Optical radiation was focused onto the crystal surface in a spot of $a = 2$ mm in diameter. One half of the crystal surface was irradiated. A metal plate protected the other half of the crystal surface. Magnitudes of the dark resistivity and photoconductivity were measured before and after laser irradiation in the irradiated and protected parts of samples investigated. Photoconductivity measurements were performed in a spectral range of 0.6 μm and 1 μm at normal conditions.

The initial magnitude of dark resistivity in both parts of the samples investigated was nearly equal, which confirms the homogeneity of initial crystals. We observed an increase of the photosensitivity and dark resistivity of $Cd_{1-x}Mn_xTe$ (x = 8–10%) crystals after laser irradiation. Typical dependence of the integrated photosensitivity in $Cd_{0.9}Mn_{0.1}Te$ crystal on the energy density of a light pulse E_L is presented in figure 1. It was found that under $E_L > 0.08$ J cm^{-2} the photosensitivity rise has taken place not only in the irradiated parts of the samples, but also outside (figure 1, curve 2). It is necessary to point out that the change of the electric characteristics and the dislocation structure modification in the irradiated and in the protected part of CdTe crystals occurs under the same mode of laser processing. This is described in [4].

Further growth of dark resistivity took place in the irradiated and also in the protected area of the crystal at further increase of the energy density above $E_L = 0.12$ J cm^{-2}. At the same time, photoconductivity quenching occurs under $E_L > 0.12$ J cm^{-2}.

3. Discussion

First of all, let us consider the processes that take place in the irradiated part of the crystal as a result of laser treatment. The change of the photoconductivity and dark resistivity of CdMnTe samples could be explained by the laser stimulation of the surface depletion of cadmium atoms similarly to a phenomenon which occurs in cadmium telluride after laser processing [6]. Really, generation of cadmium vacancies, which are acceptors and centres of photosensitivity for CdTe and CdTe-based alloys [7], can result in an increase in dark resistivity and photoconductivity values in the illuminated part of the sample.

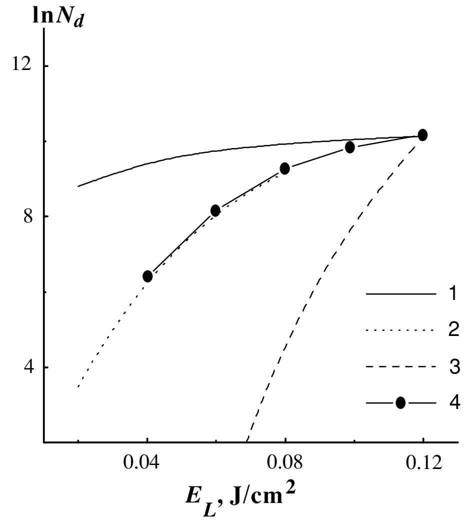

Figure 2. Dependence of the concentration of point defects generated in the irradiated part of the sample during laser processing from the energy density in a light pulse. Curve 1 was calculated at $E_A = 0.1$ eV, curve 2 at $E_A = 0.5$ eV, curve 3 at $E_A = 2.75$ eV. Curve 4 is a result of the experiment, $\Delta S(E_L)$.

The process of defect formation, resulting from the laser irradiation of semiconductors, is determined by a number of interrelated laser-induced phenomena, such as excitation of the electronic subsystem, heating and by the generation of elastic waves in a crystal. The point defect generation in the irradiated part of the sample during laser processing could be described by [8]

$$\frac{\partial N_d}{\partial t} = \text{const} \cdot \exp\left(-\frac{E_A}{k(T_0 + \Delta T)}\right) \tag{1}$$

where N_d is the defect concentration, k is Boltzmann's constant, E_A is the activation energy of the point defect generation process, T_0 is the initial temperature of the sample and ΔT is the laser heating.

The energy of an optical quantum ($h\nu \sim 1.78$ eV) exceeds the band gap value of CdMnTe. Therefore the fundamental absorption is realized and the defect generation occurs in the surface layer with a thickness of $\sim\alpha^{-1} \sim 10^{-6}$ m (α is an absorption coefficient) for crystals investigated. It is known that the increase of the defect concentration close to the crystal surface results in the increase of the surface recombination rate S [9]. Dependences of the surface recombination rate from the number of laser pulses $S(N)$ at a fixed energy density in a light pulse E_L = (0.04 J cm^{-2}; 0.08 J cm^{-2}; 0.12 J cm^{-2}) were determined from the photoconductivity measurements. The value of the surface recombination rate reaches saturation $S = S_{st}$ at a certain value of N. On the basis of this data we plotted the dependence $\Delta S(E_L)$ (see figure 2, curve 4), where $\Delta S = S_{st} - S_0$, S_0 is an initial value of the surface recombination rate at $E_L = 0$. Since the value of ΔS is proportional to the concentration of the surface centres of the recombination induced by laser irradiation [10], we calculated $N_d(E_L)$ dependence on the basis of equation (1) and compared it with an experimental dependence $\Delta S(E_L)$. The best agreement

Table 1. Properties of the CdTe and GaAs crystals used in the calculation.

	CdTe	GaAs
ρ, material density (kg m^{-3})	5860	5316
c_R, velocity of the surface acoustic wave (m s^{-1})	2×10^3	3.1×10^3
c_t, velocity of the transverse acoustic wave (m s^{-1})	2.3×10^3	3.6×10^3
c_l, velocity of the longitudinal acoustic wave (m s^{-1})	3.3×10^3	5×10^3
β, volumetric thermal expansion coefficient (K^{-1})	1.65×10^{-5}	1.5×10^{-5}
c, specific heat (J kg^{-1} K^{-1})	209	163.3
χ, thermal diffusivity, m^2 s^{-1}	7.1×10^{-6}	5.3×10^{-5}
R, optical reflection coefficient	0.436	0.34

between the experimental and calculated dependences was obtained at $E_A = 0.5$ eV (see figure 2).

Such a value essentially differs from the typical value of the thermal generation energy of intrinsic point defects for this class of semiconductors. For example, the thermal generation energy of cadmium vacancies is 2.75 eV [11]. Thus, a supposition about the decrease of the generation energy of intrinsic point defects as a result of laser processing and the influence of the laser-induced phenomena could be made.

As was noted above, the laser irradiation of the semiconductor crystal is accompanied by the excitation of the electronic subsystem, by the heating and by the generation of elastic waves. In the protected part of the crystal, the photoelectric emission (photoeffect) is ruled out by the experimental conditions. The influence of a shock wave generated under certain conditions by a laser pulse can also be excluded because of its propagation to the crystal bulk. The effective value of the heat diffusion length along a crystal surface was estimated by $l_h \sim (ct_L)^{1/2}$ within 0.37 μm. The diffusion length of the photo-excited electrons $l_{pee} \sim (Dt_L)^{1/2}$ amounts to 4 μm, where $D = 3.85 \times 10^{-4}$ m^2 s^{-1} is a diffusion coefficient of the photo-excited electrons. We also determined that the dominating diffusion flux of point defects generated in consequence of laser processing is directed into the crystal bulk and the diffusion length of the point defects along a crystal surface does not exceed several microns [12].

Thus, photo-excited charge carriers, heat and point defects generated as a result of the laser processing are localized within the illuminated field of the crystal. The mentioned factors cannot be responsible for the properties modification over the irradiated part of the crystal surface. It is thought that the effect of the 'long-range' action should be connected with propagation of the surface acoustic wave induced by the laser impulse.

3.1. Photo-excited surface acoustic wave

According to [13], the process of elastic wave photo-excitation in a solid absorbing the energy of the light is determined by the thermal expansion of the crystal lattice during spatially non-uniform heating. For a semiconductor crystal the process of the thermalization between hot carriers and the lattice causes a competitive mechanism of elastic wave excitation. At the same time the efficacy of the thermalization process depends on carrier diffusion and carrier recombination [14]. It is necessary to find the correlation between characteristic frequencies of the noted processes for the determination of the dominant mechanism.

The physical properties of the $Cd_{1-x}Mn_xTe$ with a low content of Mn are close to cadmium telluride physical properties. Therefore further calculation will be made for CdTe. Furthermore, we shall also discuss the excitation of the surface elastic wave in GaAs, which is a typical crystal from the A_3B_5 semiconductor group for which the laser effect of the 'long-range' action was observed [15].

The frequency of the photo-excited surface elastic wave may be determined as $\omega \sim \min\{t_L^{-1}, c_R/a\}$, where t_L is the light pulse duration, c_R is the surface elastic wave velocity (see table 1) and a is the diameter of the laser spot. In our experiment the value of ω is determined by the time of the surface elastic wave propagation across the region of the photo-excitation a/c_R. It is equal to $\sim 7.5 \times 10^5$ Hz and $\sim 10^6$ Hz for GaAs and CdTe crystals, respectively. Moreover, an experimental study of the photoconductivity kinetics of GaAs and CdTe original single crystals has allowed us to estimate the characteristic frequency of the charge carrier recombination ω_R which is much greater than ω. Thus, as a consequence of the theory presented in [14], the thermo-elastic mechanism of acoustic wave excitation is dominant.

We have also estimated the characteristic frequency of the process of carrier diffusion from the region of the photo-excitation as $\omega_1 = \omega_R\chi/D \sim 5 \times 10^4$ Hz. It is worth underlining that $\omega_1 \ll \omega$ and the shape of the elastic pulse induced by the laser irradiation contains some information about the dynamics of photo-excited carriers.

One of the aims of our work is determination of the possibility of the material properties change outside the irradiated part of the crystal stimulated by photo-excited elastic waves. In this connection, we have estimated the value of the displacement and the effective pressure accompanying the elastic wave propagation as a function of the time at the location of some distance from the irradiated field of a solid $r \gg a$, where a is a linear size of the laser spot. In the frame of the thermo-elastic mechanism of the generation and a non-heat-conducting medium the amplitude of the surface elastic pulse $u(r, t)$ can be calculated by [16]

$$u(r,t) = \frac{(1-R)I\tau_L\beta}{\rho c_p}\left(1 - \frac{4c_t^2}{3c_l^2}\right)\left(\frac{a}{\lambda}\right)^2\left(\frac{\lambda}{2r}\right)^{1/2} \times \Phi\left[\frac{c_R t - r}{\lambda}\right] \qquad (2)$$

$$\Phi(\xi) = \int_0^\infty k^{1/2}\exp(-k^2/4)\cos(k\xi + \pi/4)\,\mathrm{d}k$$

where $k = 2\pi/\lambda$ is the wave vector of the photo-excited elastic pulse, λ is the wavelength, $\Phi(\xi)$ is the shape of the acoustic

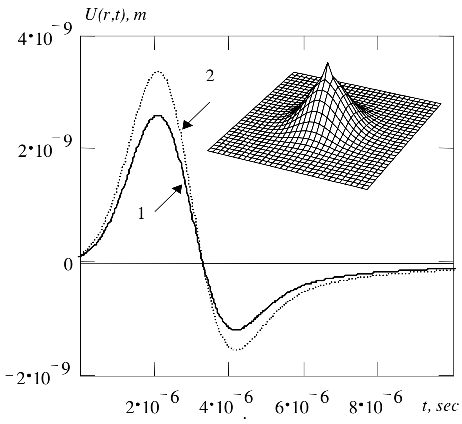

Figure 3. Surface elastic pulse calculated at the detection distance $r \sim 4$ mm according to equation (2) for CdTe (curve 1) and GaAs (curve 2) single crystals. Inset: 3D-simulation of the surface displacement of the irradiated field of GaAs crystal during pulsed laser processing.

pulse and I is the intensity of the incident radiation; the other parameters are shown in table 1.

Elastic pulses calculated according to equation (2) for CdTe and GaAs semiconductor crystals are shown in figure 3. The amplitude of the surface displacement at the detection distance r increases up to ~4 nm and the value of the effective pressure accompanying the acoustic wave propagation does not exceed 2–3 MPa at maximum used light pulse energy density 0.12 J cm^{-2}. We also made a 3D-simulation of the surface displacement sharp of a crystal during pulsed laser irradiation (see inset in figure 3).

According to the model of [17], interaction between elastic waves and defects could result in structural reconstruction of the defective region of the crystal already under the effective pressure $p > 0.06$ MPa. Hence, the energy transferred by elastic wave at some distance from the irradiated field of the crystal is sufficient for the beginning of the process of the point defect structure transformation. Such a model is confirmed by the change of photoconductivity and dark resistivity of CdMnTe samples experimentally observed in the protected part of the crystal.

Finally, let us revert to a discussion of the dependence of the integral photosensitivity on the energy density of laser irradiation. As seen from figure 1, the photosensitivity change is threshold in character for the protected part of the crystal in contrast to the irradiated part of the crystal. It is thought that such behaviour of crystal properties outside the irradiated region could be related with elastic wave propagation.

Earlier, we investigated the thermo-optical mechanism of the generation of acoustic waves in single crystals under similar conditions of laser processing [18]. The efficiency of the optical energy transformation to the acoustic energy was determined and a direct registration of acoustic waves was made. It was found that the efficiency of the wave generation is not significant under used conditions of laser processing. But it can be increased by the laser beam focusing or decreasing of the laser pulse duration. In any case, we deal with increasing the energy density in a light pulse. We have determined that the signal detected with a typical Rayleigh waveform is observed outside the irradiated region when $E_L > 0.06$ J cm^{-2} only.

On the other hand, the threshold character of the material properties change could be connected with the overcoming of a certain barrier for the beginning of the defect system transformation and confirms the activation character of the defect generation. It is necessary to note that a rise of the photosensitivity outside the irradiated region took place also under $E_L < 0.08$ J cm^{-2} and it relaxed to its original value during 10–20 min. At the same time, under $E_L > 0.08$ J cm^{-2}, the material properties change had a permanent character.

Thus, the threshold behaviour of the photosensitivity change is determined by both the efficiency of the acoustic wave generation and the activation character of the defect generation. However, more experiments are clearly necessary.

4. Conclusion

In conclusion, we have observed the photosensitivity increase and modification of the electrical properties of CdMnTe crystals, which were photo-excited by pulsed ruby laser irradiation ($\lambda = 0.694$ μm, $t_L = 20$ ns) with a pre-threshold energy density. Changes took place not only in the irradiated part of the samples, but also outside. It was determined that decreasing of the energy of the point defect generation takes place as a result of laser processing and the influence of the laser-induced phenomena.

The analysis of laser-induced heating and the point defects diffusion has shown that these factors cannot be responsible for the laser effect of the 'long-range' action. We connected it with photo-excited elastic wave propagation to the protected part of the crystal. It was shown that the energy transferred by elastic waves at some distance from the irradiated field of the crystal is sufficient for the beginning of the point defect structure transformation.

It was determined that the thermo-elastic mechanism of the surface acoustic wave excitation is dominant for the nanosecond regime of the laser irradiation ($\lambda = 694$ nm). The amplitude of the surface displacement and the value of the effective pressure accompanying the acoustic wave propagation were estimated for CdTe and GaAs single crystals.

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