Sensitivity of PbSnTe:In films to the radiation of free electron laser

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Abstract. The analysis of experimental data on the observation of photoresponse in narrow gap semiconductor Pb₁₋ₓSnₓTe:In films grown by the method of molecular beam epitaxy, exposing samples to the powerful radiation of the Novosibirsk free electron laser (wavelength range of about 70–240 µm) under different measurement conditions, is presented in the paper. Both the positive and negative photoconductivities were detected. In a magnetic field, the resonance-type photoconductivity was observed. The results are discussed within the framework of the model taking into account the existence of different capture levels in PbSnTe.

1. Introduction

The band gap $E_g$ of the Pb₁₋ₓSnₓTe:In solid solution depends upon the composition and the temperature [1]. Near the liquid helium temperature, the increase in the content of Sn from $x = 0$ (PbTe) to $x \approx 0.35$ leads to a reduction of $E_g$ from $\approx 0.19$ eV to zero and, for $x > 0.35$, it increases again. Within $x \approx 0.24–0.29$ ($E_g \approx 0.04–0.06$ eV), adding In to this compound leads to a “pinning” of the Fermi level in the band gap. Thus, the resistivity of narrow-gap semiconductor PbSnTe:In at $T = 4.2$ K becomes comparable to the resistivity of conventional insulators at $T \approx 300$ K. A giant value of the static permittivity [2] of more than 2000 at $T = 4.2$ K is one of the reasons for the low ionization energy of impurity centers in PbSnTe:In, which corresponds to the submillimeter (terahertz) wavelength range. This applies to the intrinsic point defects in PbSnTe:In (tellurium and metal vacancies) that can create trap levels within the band gap. In addition, the features of PbSnTe as a solid solution (i.e., a disordered system [3]), can lead to a complex energy spectrum of trap levels, the filling of which depends on the magnitudes of applied electric and magnetic fields or the lighting conditions in the fundamental absorption band (far ir region). The possible link of a complex spectrum of traps with the features of the magnetoresistance of PbSnTe:In was pointed, for example, in [4]. The assessment of the trap levels energy distribution within the bandgap by analyzing the current-voltage characteristics (CVC) was carried out in [5]. The aim of this study is to analyze the photoresponse characteristics of the structures based on PbSnTe:In films in the submillimeter...
wavelength range using the free electrons laser (FEL) radiation and a static magnetic field up to 4 T.

2. Samples and experimental setup

The structures based on PbSnTe:In single-crystal films with a thickness of about one to several micrometers, that were grown by molecular beam epitaxy (MBE) on a substrate BaF$_2$ (111), were investigated. The MBE technique is described, for example, in [6]. The typical values of the charge carriers (electrons $n$ or holes $p$) mobility in the samples were $\mu \approx (1–10) \times 10^4$ cm$^2$V$^{-1}$s$^{-1}$ at $T < 30$ K, depending on the film composition, temperature and measurement conditions (presence or absence of lighting). In the selected samples, the concentration of $n$ ($p$) without lighting was decreased upon cooling from room temperature to $T \approx 15–20$ K for the law close to the Arrhenius law. A typical value of the activation energy was $\Delta E \approx 0.01–0.03$ eV. Below the temperature $T \approx 15–20$ K, the measurement of the Hall effect was considerably complicated due to a high resistance of the samples (more than $10^9$ $\Omega$). The minimum measured values of $n$ ($p$) were about $10^{12}$ cm$^{-3}$ in the range of $T \approx 15–20$ K.

The investigation of the terahertz radiation effect on PbSnTe:In film was carried out at the work station of the Novosibirsk free electron laser (NovoFEL). The NovoFEL radiation characteristics are described in detail, for example, in [7]. The laser operates in the quasi-continuous mode with a pulse repetition frequency of about $10^7$ Hz for each pulse duration of about $10^{-10}$ s. The emission wavelength of FEL can be varied in the range of $\lambda \approx 65–235$ $\mu$m at the typical relative line width $\Delta \lambda \approx 0.5–1\%$. The average radiation power of FEL measured by the output window was varied from a few to tens of watts (depending on the wavelength and laser operation mode). However, because of special polarized attenuator was installed between output window and sample setup, the FEL illumination intensity of the sample was about of $0.01–0.1$ W/cm$^2$. It was comparable for different wavelengths and it may not lead to a noticeable change in the temperature (“heating”) of the samples directly immersed in liquid helium. FEL radiation “on” and “off” was carried out by opening and closing a mechanical shutter for the time of about fractions of a second.

The experiments on the interaction of PbSnTe:In and FEL radiation were performed on two types of samples. The sample of the first type is schematically shown in figure 1(a). Indium contacts to the PbSnTe:In film were separated by a narrow (30–60 $\mu$m) and long ($\approx 0.2$ cm) gap. The contacts were made using vacuum deposition and photolithography. The structures of this type are used for measurements in the injection and the current limited by the space charge (SCLC) mode. The sample of the second type for the Hall effect measurements under different conditions is shown schematically in figure 1(b).

Optical measurements were performed directly in liquid helium inside the Dewar at $T = 4.2$ K. The samples were shielded from the uncontrolled background radiation by a metal chamber. FEL radiation was introduced into the sample chamber through the black plastic window nontransparent to the radiation in the fundamental absorption band of PbSnTe:In (roughly in the region of $\lambda \lesssim 30$ $\mu$m). For adjustable samples lighting in the fundamental absorption band
within the chamber, the light emission diode (LED) with a wavelength of about $\lambda \approx 0.65 \, \mu m$ was located near the sample. LED was connected to a voltage source through 10 kΩ load resistance. When the measurements were carried out in the magnetic field, the sample chamber was placed inside a superconducting coil, which provided a magnetic field $B \leq 4 \, T$.

3. Experimental results

3.1. Sensitivity of PbSnTe:In to the FEL radiation in the current limited by the space charge mode at the absence of a magnetic field

In this mode measurements were carried out both in the absence and presence of additional lighting within the PbSnTe:In fundamental absorption band. The samples of the first type were used, see figure 1(a).

3.1.1. Sensitivity to the FEL radiation without additional short-wavelength light  

The dc voltage within range $U = 0.01$–3.0 V was applied to the sample. The current variation range, at the same time, was about $I = 10^{-12}$–$10^{-5}$ A. The CVC was corresponded to the SCLC mode and was similar to that described in [5]. The series of time dependences of sample current increase and decrease, when turning on and off the FEL at different wavelengths and bias voltages, are shown in figure 2. In all graphs, the FEL radiation was turned on at $t = 0$. The turning-off moments are shown with arrows. The portions of the respective dependencies close to the moments of turning on and off are presented in figure 2(b, c, f) on a larger time scale. For easy comparison, the radiations off points for different curves are shifted in time so that the beginnings of the current reduction on them are at the same time moment. The main features of the above-mentioned results are as follows:

- both the decrease in current after switching off the FEL, and, especially, the increase have a long-term nature with the characteristic times from a few to tens and hundreds of seconds;
- the relative change in the current, when illuminated, is substantially different for various $\lambda$ and reaches about 30–1000 times for $\lambda = 123 \, \mu m$, ~100 times for $\lambda = 68 \, \mu m$ and no more than 3 times for $\lambda = 205 \, \mu m$;
- the relative change of current amplitude and, especially, the complex dynamics of its increase and decrease, when turning on and off the FEL, respectively, are highly dependent on the bias voltage and the FEL wavelength.

For example, at $\lambda = 123 \, \mu m$ (a, b) there are expressed stages on curves where the photocurrent oscillates ($t \approx 0$–25 s, $U_{bias} = 0.096$, 1.5 V). At the same voltages less pronounced stages are observed in the current recession regions. Such features are not visible at $\lambda = 123 \, \mu m$, when the relative change in the current under FEL radiation is also high. At the maximum voltage $U_{bias} \approx 3 \, V$ and $\lambda = 123$; 68 $\mu m$ after FEL turn on the short section of the negative photocurrent is traced, but when $\lambda = 205 \, \mu m$ (c, d) at the same voltage, the photocurrent remains negative until the FEL shutdown. At the same wavelength, there is a transformation of the initial section of complex shape dependences ($t \leq 10 \, s$). At low voltages ($U_{bias} = 0.047$–0.096 V), there are sections of negative photocurrents which disappear at $U_{bias} = 0.41$–1.5 V. The voltage boost to $U_{bias} = 2.9 \, V$, again, leads to a negative photocurrent.

3.1.2. Sensitivity to the FEL radiation with additional lighting in the PbSnTe:In fundamental absorption band

The measurements were made under supplementary lighting background radiation ($T_b \approx 300 \, K$) in the samples of the first type, too, see figure 1(a). When lighting the PbSnTe:In film, from which samples were made, the conductivity was changed from the electronic to the hole type. The CVCs in such conditions are shown in figure 3(a). It is seen that, at a voltage $U_{bias} \leq 0.2 \, V$, the current dependence on voltage is nearly linear with the
resistance of the sample of about 300 Ω. When $U_{\text{bias}} \approx 0.2 \text{ V}$, the current was abruptly decreased of about three times with a further complicated behavior depending on $U_{\text{bias}}$. The current, changing under the FEL radiation with wavelength $\lambda_{\text{FEL}} = 198 \text{ µm}$, is shown in figure 3(b). It can be seen that, up to $U_{\text{bias}} \approx 0.2 \text{ V}$, the current is decreased under the FEL radiation (negative photoconductivity) and increased (normal photoconductivity) over this voltage.

### 3.2. Sensitivity to the FEL radiation in the ohmic regime under strong illumination in the fundamental absorption band and in the presence of a magnetic field

The samples of the second type for measuring the Hall effect were studied, see figure 1(b). The CVCs were linear at all levels of the used LED and FEL lighting, and any magnetic field. A typical value of sample resistance under LED radiation was $10^3$–$10^4 \Omega$. We measured the longitudinal and transverse voltage on the structure at the current $I = 7.5 \times 10^{-5} \text{ A}$. The longitudinal $U_{\text{long}}$ and transverse $U_{\text{trans}}$ voltage dependences of one of the samples with thickness $d = 1.7 \text{ µm}$ on the magnetic field at switched off (“off”) and on (“on”) FEL radiation with wavelength $\lambda = 205 \text{ µm}$ are shown in figure 4(a). For each curve the magnetic field was increased from zero to $B = 4 \text{ T}$ at a rate $0.033 \text{ T/s}$. In the region of weak magnetic field $B \leq 0.1 \text{ T}$, the $U_{\text{long}}(B)$ dependence was weak, the $U_{\text{trans}}(B)$ dependence was close to linear law. Defined in this region of $B$, the electron concentration and mobility were $n \approx 1.3 \times 10^{13} \text{ cm}^{-3}$ and $\mu \approx 4.9 \times 10^4 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, respectively. The dependences of the relative change in the longitudinal and transverse voltages $\Delta U(B)/U(B)$ under the FEL radiation on the magnetic field are shown in figure 4(b). It is evident that these relations have a resonance character with their maxima located close to $B = 1.3 \text{ T}$. The dashed lines show similar dependences at a lower sample illumination (LED supply voltage was reduced from 10.2 to 8.0 V). The dependences obtained in the same conditions at FEL wavelength $\lambda = 138.5 \text{ µm}$ and two levels of sample illumination by LED are shown in figure 4(c, d). The two pairs of closely spaced curves, see figure 4(d), correspond to the two independent measurements and show a good reproducibility of the results, not only near the maxima, but also near the lower amplitude features reflected on the larger scale insets.

### 4. Discussion

The photosensitivity of PbSnTe:In samples to THz radiation was observed in a number of papers, for example [8, 9]. The dependence of the THz signal on the magnetic field is associated by the authors [10] with the presence of unusual localized states, which energy position under non-equilibrium conditions depends on the position of the quasi-Fermi level. The complex of experimental results presented in this paper indicates the presence of a wide variety of trap levels in PbSnTe:In, as described, for example, in [5]. On the qualitative level experimental results could be explained by the change of trap levels population under external actions, including the injection of charge carriers from the contacts, the illumination in different spectral ranges (including THz) and the application of the magnetic field. Thus, in figure 2, the photoresponse is significantly different for different wavelengths, and minimal for $\lambda_{\text{FEL}} = 205 \text{ µm}$. According to the estimates [5], the energy of photons with this wavelength exactly corresponds to the region of trap energy spectrum where the levels density is low. In fact, FEL radiation leads to the traps release in a narrow range of its ionization energy. In other words, the corresponding levels are as if they were “removed” from a complex energy spectrum of the traps. The total change of charge at the respective levels under the FEL radiation highly depends on their initial population without illumination.

In terms of SCLC, this population is given by the applied voltage. Moreover, the current (or free charge carriers concentration) is defined not by a total charge captured on different levels, but by its distribution over the levels. It can be assumed that the change of charge distribution over the levels under the FEL radiation can leads not only to the current increase,
Figure 2. Time dependences of the current \((a, b, e, f)\) and the normalized current \(I(t)/I_{\text{dark}}\) \((c, d)\) through the first-type sample, see figure 1\((a)\). \(I_{\text{dark}}\) is the current value before switching on the FEL radiation \((t < 0)\). Wavelength \(\lambda_{\text{FEL}} = 123\) \((a, b)\), 205 \((c, d)\) and 68 \(\mu\)m \((e, f)\). The bias voltage \(U_{\text{bias}}\) values for the corresponding curves are shown. The moments of FEL turning off are indicated with arrows. \((a, c, e)\) Initial dependences. \((b, d, e)\) The points of turning off are combined (zoomed in on time) for a better comparison of the dynamics of current reduction for various \(U_{\text{bias}}\) values.

but (in certain conditions) to its reduction observed in the experiment. We also note that, at \(T = 4.2\) K, the charge redistribution associated with the emission of carriers from the levels to the permitted bands and the subsequent capture by other levels is a slow process. This corresponds to the experimentally observed longtime current relaxation. Since at different wavelengths and
Figure 3. Dependences on the voltage: current—when the sample is illuminated by background radiation with the temperature of $T_b = 300$ K $(a)$; change of current—when the sample is additionally illuminated by FEL at wavelength $\lambda_{\text{FEL}} = 198$ $\mu$m $(b)$. The sample is of the first type, see figure 1$(a)$.

voltages the different levels are involved in such processes, this leads to the observed relaxation (transition) processes diversity.

The results shown in figure 3 can be explained as follows. In terms of a strong continuous illumination in the interband transition region, the electrons generated by background radiation fill almost all trap levels, including the most “shallow” ones. The ohmic photoconductivity at $U_{\text{bias}} < 0.2$ V is defined by the free holes, which concentration is determined by the total concentration of captured electrons. The electron injection in this region is insignificant. When $U_{\text{bias}} \approx 0.2$ V, the injection increases sharply, and the interband recombination leads to the change of $p$-type conductivity for the $n$-type. In turn, in the SCLC mode the electron concentration depends on the voltage [11], which leads to further CVC nonlinearity. Threshold character of the current switching in the vicinity of $U_{\text{bias}} \approx 0.2$ V may be due to a sharp increase in the electric field close to the injection contact. It is the result of the most intense recombination of the injected electrons and holes in this area. The FEL illumination at $U_{\text{bias}} < 0.2$ V leads to the electron emission from the traps to the conduction band, their recombination with nonequilibrium holes and some reduction ($\sim 1–10\%$) of the conductivity (negative photoconductivity). When $U_{\text{bias}} > 0.2$ V, the sample has the $n$-type conductivity, and a similar process leads to an increase in the current (positive photoconductivity) which, in the SCLC mode, has a complex non-linear dependence on $U_{\text{bias}}$.

Let us now turn to the results shown in figure 4. The illumination conditions in this case are similar to those described above. Without FEL illumination, ohmic conductivity is also determined by the concentration of captured charge carriers, which defines the concentration of free charge carriers of the opposite sign. In this case, initially the sample had a $p$-type conductivity, and it was changed to an $n$-type under the LED illumination (holes were captured). As seen in figure 4, at the FEL illumination, the signal was observed at the different values of magnetic field $B$. In some regions of $B$, it was matched to the normal positive photoconductivity with the negative $\Delta U/U$ value, for example at $B < 1.5$ T in figure 4$(c)$, and in some regions—negative photoconductivity (close to the main maximum on all curves in figure 4). Based on the above-described model, the change not only of the amplitude, but also the sign of the signal looks as if the magnetic field changes the type of traps interacting with FEL at a selected wavelength. This assumption can be explained with the assistance of the “magnetic freezing” effect associated with a change of the trap level ionization energy $\Delta E_t$ in the magnetic field.
Figure 4. The dependences of longitudinal ($U_{\text{long}}$) and transverse ($U_{\text{trans}}$) voltages ($a$), and their relative $\Delta U/U$ changes under the FEL radiation ($b$–$d$) on the magnetic field. Wavelength $\lambda_{\text{FEL}}$ ($\mu$m): 205 ($a$, $b$) and 138.5 ($c$, $d$). The supply voltage of additional lighting $U_{\text{LED}}$ (V): 10.2 ($a$); 8.0 and 10.2 ($b$); 10.2 ($c$); 18.2 ($d$). In the figure ($a$) dashed curves were obtained without FEL illumination, solid—under illumination. In the figure ($d$) “dual” curves represent two independent measurements and the areas with additional reproducible features are shown on the larger scale insets.

Indeed, the condition of cyclotron resonance $\mu B > 1$ for the sample already began to be fulfilled in the field $B > 0.2$ T. The estimate of $\hbar \omega_c/2$, where $\omega_c$—cyclotron frequency, gives a value of $\sim 0.01$ eV in magnetic field $B = 1$ T (effective mass $m^* = 0.01m_e$). In order of magnitude it is comparable to the energy of FEL quantum. It should be emphasized that the given value is a rough estimate, and the real change of $\Delta E_i$ may greatly differ from it and be different for various trap levels. However, the change of magnetic field under the cyclotron resonance conditions should also result in the change of $\Delta E_i$. In other words, at a different magnetic field, the quanta of FEL radiation interact with different trap levels. The amplitude and shape of the signal, depending on $B$, should depend on the FEL wavelength and specific energy spectrum of trap levels, which transforms into the magnetic field. In such representations, the dependences, see figure 4($b$), look like a superposition of the broadened maxima in the vicinity of $B \approx 1.3$ T and $B \approx 2.6$ T ($\lambda_{\text{FEL}} = 200$ $\mu$m), which are a consequence of the features of the traps energy spectrum in the corresponding energy range. In figure 4($c$, $d$) ($\lambda_{\text{FEL}} = 138.5$ $\mu$m), the maxima in the region of “negative photoconductivity” also look like a superposition of at least two maxima with the positions in the vicinity of $B \approx 2$ T and $B \approx 2.5$ T. A series of additional features of the curves $\Delta U_{\text{trans}}$, see figure 4($d$), is observed in $B \approx 3$–$4$ T. It can also be interpreted as a consequence of the trap levels spectrum features.
5. Conclusion
Thus, the results of comprehensive studies of PbSnTe:In film sensitivity to the FEL radiation can be explained within the concept of a complex energy spectrum of the trap levels described in [5]. In the framework of the qualitative model both the signal amplitude and its dynamics are determined by the ratio between the energies of the FEL photon, ionization energies and the density of the corresponding trap levels and their filling degree. These parameters are sensitive to the applied voltage (space charge-limited current), to the illumination in the region of interband generation (background or additional illumination) and to the magnetic field, which can change the levels ionization energy due to the effect of “magnetic freezing”. This is the reason of the complex photoeffects in PbSnTe:In at the THz wavelengths described in this paper.

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