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# Microwave Photoconductivity in $Pb_{1-x}Sn_xTe(In)$ Semi-Insulating Solid Solutions

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We study photoconductivity in $Pb_{1-x}Sn_xTe(In)$ semi-insulating solid solutions under 48 GHz radiation at temperatures 4.2−50 K. We found qualitative differences between the microwave photoconductivity and previously observed photoconductivity induced by terahertz radiation with frequencies $f \geq 0.6$ THz. We show that in contrast to the terahertz photoconductivity the microwave photoconductivity at low temperatures has negative sign and does not manifest long-term relaxation. Possible mechanisms responsible for the microwave photoresponse are discussed.

**Keywords:** photoconductivity, lead−tin telluride, microwave radiation.

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## 1. Introduction

The interest in the study of semiconductor solid solutions of $Pb_{1-x}Sn_xTe$ is mainly related to their high photosensitivity in the infrared and terahertz ranges [1–3]. These narrow-gap semiconductors are also considered as promising materials for creating highly efficient thermoelectric devices [4–7].

A number of unique features of the optoelectronic properties of $Pb_{1-x}Sn_xTe$ are attributable to the specific structure of the energy spectrum of these materials [8]. An increase in the molar content of tin telluride in a solid solution is accompanied by a transition from the direct order of the energy bands at $x < 0.35$ through the gapless state to the inverse order at $x > 0.35$. Under the conditions of such a restructuring of the band structure with an increase of $x$, a transition from a trivial state to the phase of a topological crystalline insulator is realized, the discussion about the topological properties of which has remained open over the past years [9–11]. In particular, the formation of topological electronic states of higher order is discussed [12,13].

Doping of solid solutions of PbTe-SnTe with indium leads to the formation of a system of levels corresponding to different charge states of impurity centers. The manifestation of the mixed valence of impurity indium determines the effect of Fermi level stabilization. In the composition range $0.22 < x < 0.29$, a deep indium level stabilizes the Fermi level in the band gap, ensuring the implementation of a semi-insulating state.

Long-term relaxation processes and persistent photoconductivity (PC) are observed due to the correlation interaction in the crystal lattice−impurity center in $Pb_{1-x}Sn_xTe(In)$ at low temperatures of $T <\sim 25$ K [14]. These nonequilibrium phenomena in systems based on lead telluride are well studied, basic information can be found in the reviews [3,8]. Persistent positive PC in the terahertz range is one of the most interesting photoelectric effects observed in $Pb_{1-x}Sn_xTe(In)$. It is important that the generation processes and the positive PC are preserved with a decrease in the energy of the exciting quantum up to 2.5 meV (corresponds to the frequency of $f = 0.6$ THz) [15], including even under conditions of electron gas degeneracy [16,17]. The practical absence of the red boundary of the effect made it possible to associate it with specific local states tied to the position of the Fermi quasi-level [17]. At the same time, it is noted in Ref. [18] that the transition of the stabilized Fermi level through the middle of the band gap with an increase in the SnTe content in a solid solution $x \geq 0.26$ and a change in the type of conductivity from electronic to hole are accompanied by suppression of the positive persistent PC.

It is interesting to study PC stimulated by longer-wavelength radiation and compare its properties with the previously observed features of terahertz PC. PC was studied in this paper in semi-insulating solid solutions of $Pb_{1-x}Sn_xTe(In)$ of both *p*- and *n*-type under conditions of exposure to microwave radiation.

## 2. Studied samples and experimental methods

The studied single crystals of $Pb_{1-x}Sn_xTe(In)$ in the vicinity of composition $x = 0.25$ were grown by the modified Bridgman method. The indium content in the mixture did not exceed 1%. Samples in the form of rectangular parallelepipeds with characteristic dimensions of $1 \times 1 \times 4\,mm^3$ were cut from monocrystalline wafers. The contacts were applied with a micro-soldering iron using an indium-based alloy.

Two low-temperature inserts were used for measurements. The first of them was designed for measurements under conditions of complete shielding of the sample from the background heat illumination. The sample was placed in a closed low-temperature chamber located at the bottom of the insert. At the same time, special measures were taken to prevent any „dripping“ background radiation, including through the holes for the supply wires. The second insert was intended for measurements of microwave PC. The source of the microwave radiation was a Gunn diode with a frequency of 48 GHz and an output power of 10 mW, located in the upper part of the insert at room temperature. The sample was located in the lower, low-temperature part of the insert. The radiation was applied to the sample through a stainless steel tube. In this case, the sample was additionally exposed to thermal background radiation from the upper part of the insert at room temperature.

The transport properties of the samples were studied in the temperature range of 4.2–300 K in magnetic fields up to 0.05 T. All galvanomagnetic and photoelectric measurements were carried out using a standard four-probe technique in the geometry of the Hall bar. The current through the sample was set using a constant voltage source connected in series with the sample and a load resistance, the value of which was much higher than the resistance of the sample. All measurements were carried out for two opposite directions of current through the sample to exclude any impact of thermo- and photovoltaics. The type of conductivity was determined by the sign of the Hall coefficient.

The study of microwave PC was carried out in *n*- and *p*-type samples at temperatures of 4.2–50 K using the modulation technique. The frequency of modulation of the diode radiation flux was 13 Hz. The amplitude and phase of the voltage drop change at potential or Hall contacts induced by microwave radiation at the modulation frequency were recorded.

## 3. Experimental results

Figure 1 shows the dependence of the resistance of the studied samples on the reciprocal temperature in dark conditions (curves *1* and *2*) and under background illumination (curves *3* and *4*). The high-temperature activation regions in the presented curves correspond to the regions of intrinsic (at $T >\sim 150$ K) and impurity (at $\sim 30\,K < T <\sim 150$ K) conductivity. The exposure to a thermal background reduces the resistance values of the sample by orders of magnitude at sufficiently low temperatures of $T <\sim 25$ K. The temperature dependences obtained are typical for semi-insulating compositions of $Pb_{1-x}Sn_xTe(In)$ [8]. It should be noted that the *p*-type samples had several orders of magnitude higher resistance compared to the *n*-type single crystals.

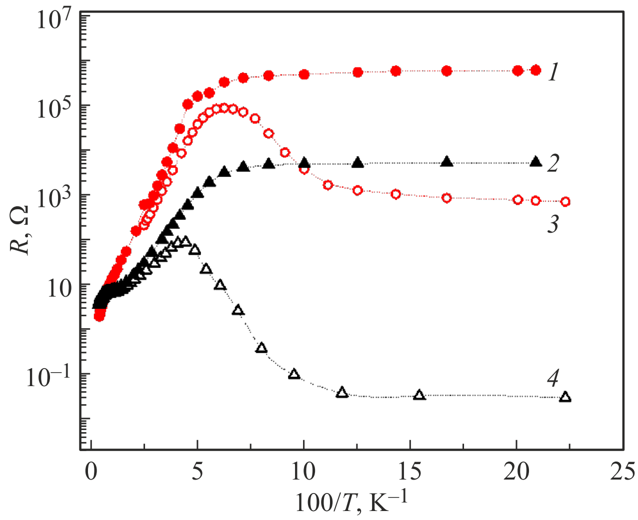


**Figure 1.** Dependences of the resistance of the studied *p*- and *n*-type samples in dark conditions (curves *1* and *2*, respectively) and under background illumination (curves *3* and *4* respectively) on the reciprocal temperature.

Microwave PC was detected in the studied samples at $T <\sim 40$ K. Figure 2 shows the temperature dependences of resistance $R$ with background illumination (*a*) and photoresistance $\triangle R$ with additional microwave excitation (*b*) for *p*- and *n*-type samples. Despite the fact that the absolute values of $\triangle R$ in high-resistance *p*-type samples and relatively well-conducting *n*-type samples are very different, nevertheless, the main features of the dependencies $\triangle R(T)$ are qualitatively similar in them. Negative PC is observed in the low temperature region ($\triangle R > 0$ at $T < 25$ K and at $T < 17$ K for *n*- and *p*-type samples, respectively). As the temperature increases, the amount of photoresistance increases, reaches a maximum, and then drops quite rapidly, demonstrating a change in sign. With a further increase in temperature, the effect of positive PC ($\triangle R < 0$) is suppressed, practically ceasing to be recorded at $\sim 40$ K. A change in the sign of the PC is observed at temperatures of 17 and 25 K in samples of *p*- and *n*-type, respectively. It should be noted that there were no indications of persistent PC. In particular, the resistance of the sample before and after the end of the microwave radiation supply coincided at all temperatures.

A comparison of the dependencies $R(T)$ in Figure 2,*a* and $\triangle R(T)$ in Figure 2,*b* shows that the sign of the photoresistance practically coincides with the sign of the derivative of the resistance with respect to temperature. The sign change $\triangle R$ is observed at a temperature corresponding to the maximum on the curve $R(T)$. The results obtained make it possible to link the registered PC with the bolometric effect.

The Hall photovoltage $\triangle U_H$ was measured to find out to what extent the generation processes and the concentration change factor during microwave excitation are responsible for the formation of the observed PC. The Hall photo response $\triangle U_H$ was recorded in both *n*- and *p*-type samples. In this case, the sign $\triangle U_H$ corresponds to a decrease in the concentration of the main carriers at $T < 25$ K for samples of *n*-type and at $T < 17$ K for samples of *p*-type. Experiments have shown that the change in the Hall photoresponse with temperature for each of the samples repeats with a high degree of accuracy the features of the corresponding dependence $\triangle R(T)$, demonstrating a similar sign change. The temperatures of the inversion of the sign $\triangle U_H$ and PC are practically the same. Figure 3, *a* and *b* show the temperature dependences of the Hall concentration under background illumination and the excess Hall concentration of the main carriers under additional exposure to radiation from the Gunn diode, respectively. The punctured symbols in Figure 3, *b* indicate the points corresponding to the negative values of the excess concentration of electrons $\triangle n$ and holes $\triangle p$ in samples *n*- and *p*-type, respectively. The dotted line shows trend lines for clarity. The observed correlation of the change in signs of photoresistance and excess concentration with an increase in temperature shows that microwave PC is associated with the processes of generation of nonequilibrium carriers.

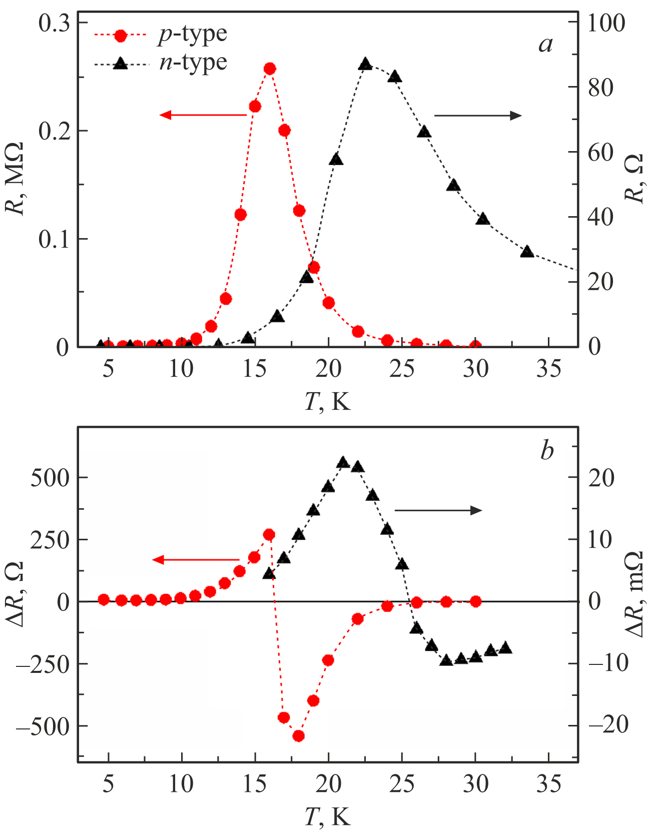


**Figure 2.** Temperature dependences of the resistance when illuminated by background radiation (*a*) and the photoresistance induced by radiation from the Gunn diode (*b*) of the studied samples. It is necessary to pay attention to the different scale on the right and left scales.

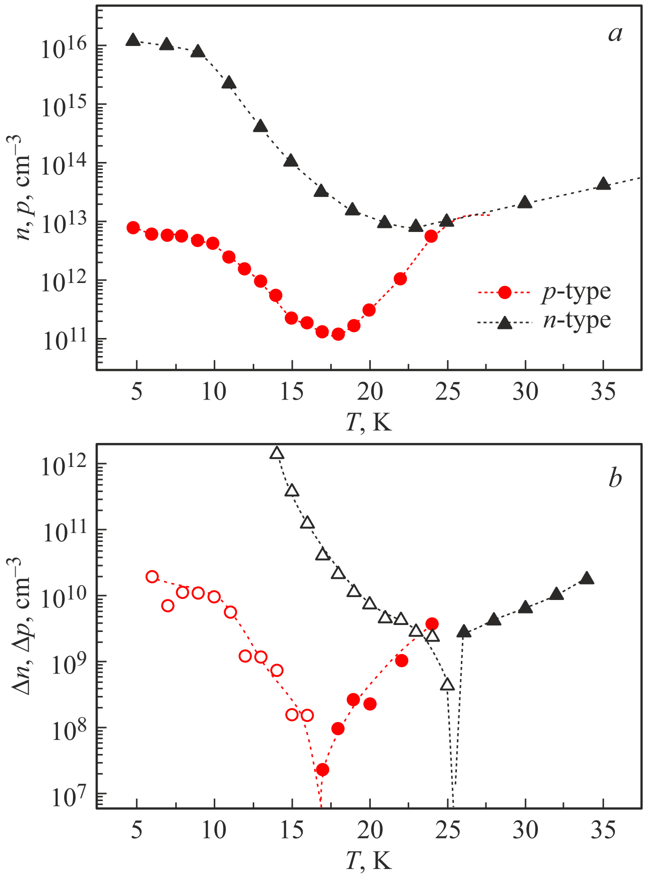


**Figure 3.** Temperature dependences of the concentration under background illumination (*a*) and the excess concentration of the main carriers under the influence of microwave radiation from the Gunn diode (*b*) in the studied samples. Negative values are indicated by punctured characters. The dotted line shows trend lines for clarity.

## 4. Discussion of the results

The presented experimental results demonstrate that microwave excitation stimulates bolometric PC in the studied samples. It is important that PC is negative at low temperatures and is not persistent.

The sign of the PC of systems based on lead telluride can be either positive or negative, while in some cases the contributions of both signs coexist. Usually, in the solid solutions under consideration, PC is characterized by complex kinetics reflecting the participation of nonequilibrium processes with various time constants.

Negative PC in such semiconductors has often been observed before. It manifested itself in Refs. [19–22] as quenching of the background infrared PC under conditions of excitation by quanta with energy exceeding the band gap of the semiconductor. When exposed to terahertz laser radiation with low quantum energy, which practically eliminates interband generation, the negative sign of the PC is usually due to a decrease in the mobility of free carriers as a result of their heating. In particular, processes of this type were observed in $Pb_{1-x}Sn_xTe(In)$, for example, in Ref. [15].

In turn, the persistent positive PC in $Pb_{1-x}Sn_xTe(In)$ and PbTe(Ga), excited by quanta with energy significantly lower than the band gap, is usually associated with the formation of DX-like impurity centers, ensuring long relaxation times for nonequilibrium carriers. Under conditions of high-power terahertz radiation, special local states may be responsible for persistent positive PC in $Pb_{1-x}Sn_xTe(In)$ at low temperatures of $T < 25$ K, whose energy levels are tied to the position of the quasi-Fermi level [16], controlled in turn by the background illumination intensity. In particular, a positive PC with slow relaxation to the equilibrium state was observed with a decrease in the radiation frequency down to 0.6 THz, corresponding to the quantum energy $\sim 2.5$ meV [15]. This energy is significantly less than all the characteristic energies of the electronic spectrum $Pb_{1-x}Sn_xTe(In)$, such as the band gap, the activation energy of the impurity state, and the energy distance from the bottom of the conduction band to the quasi-Fermi level. This circumstance allowed assuming the absence of the threshold of the photoinduced process, leading to a positive persistent PC [17]. At the same time, the results of this work show that when microwave radiation with a frequency of 48 GHz is excited at low temperatures, a negative PC is recorded that does not include a delayed component. Therefore, there is reason to believe that the red border of the effect of the previously observed positive delayed PC exists, while the threshold frequency value lies in the range of 48–600 GHz.

However, it should be noted that the sign and kinetics of the PC may depend on the position of the quasi-Fermi level, determined by the intensity of the background radiation. In particular, it was shown in Ref. [23] that PC is negative and is not persistent in PbTe(Ga) at a low background level. An increase in the intensity of the background radiation leads to the appearance of a positive persistent PC. Thus, the observed negative sign and rapid relaxation kinetics of PC may be associated with a low level of background excitation.

The results of the Hall effect study convincingly prove that microwave radiation leads to a change in carrier concentration. Analysis of temperature dependences of relative changes in conductivity $\triangle\sigma/\sigma_0(T)$ and carrier concentration $\triangle n/n_0(T)$, $\triangle p/p_0(T)$ ($\sigma_0$, $n_0$, $p_0$ — conductivity and concentration of electrons and holes at the same temperature in the absence of a microwave pulse) under photoexcitation conditions indicates that recombination processes can be considered a factor determining negative PC in the low-temperature region. When the PC sign changes to positive with increasing temperature, there is a noticeable difference between the values $\triangle\sigma/\sigma_0(T)$ and $\triangle n/n_0(T)$ ($\triangle p/p_0(T)$). This indicates that, along with the generation processes, when the temperature rises, there is a significant change in mobility due to the heating of carriers by microwave radiation.

## 5. Conclusion

Thus, it is shown in this study that PC stimulated by microwave radiation with a frequency of 48 GHz is observed in single crystals $Pb_{1-x}Sn_xTe(In)$ in the vicinity of the composition $x \sim 0.25$ at low temperatures. PC is non-persistent and negative. The results obtained may indicate the presence of a frequency threshold of the previously observed positive terahertz PC effect due to the excitation of states near the quasi-Fermi level.

However, it is important to keep in mind that the intensity of background thermal radiation can affect the sign and kinetics of the PC. It is possible that long-term relaxation processes caused by specific electronic states near the quasi-Fermi level may manifest themselves in conditions of elevated background concentrations.

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### Conflict of interest

The authors declare that they have no conflict of interest.

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