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(De)localization of nonequilibrium electrons in $Pb_{1-x}Sn_xTe(In)$ in high magnetic field

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We report on high-field ($H < 38$ T) magnetoresistance and Hall effect measurements of initially dielectric $Pb_{0.75}Sn_{0.25}Te(In)$. The data taken at 4.2 K show a magnetoresistance increase in time (localization) for a relatively low concentration of nonequilibrium electrons n. For a higher level of photoexcitation the localization disappears, and the Hall effect measurements, on the contrary, reveal an increase of n with field. The localization is attributed to energy band modulation. The delocalization is discussed in terms of field-induced electron transitions between the metastable local and extended states.

In recent years lead–tin telluride doped with indium has received a lot of attention as a new promising candidate for photodetection in the far infrared. The nontrivial effects observed in these materials – Fermi level pinning [1] and persistent photoconductivity [2] – originate from unusual properties of impurity states in $Pb_{1-x}Sn_xTe(In)$ [3]. The Fermi level pinning effect provides the electrical homogeneity of $Pb_{1-x}Sn_xTe(In)$ to be even higher than in the undoped material [4]. As far as we know, there exists only an approximate estimation of the remaining energy band modulation (EBM) amplitude in $Pb_{1-x}Sn_xTe(In)$ [5], and no effects definitely due to the EBM existence have been observed until now.

Low free carrier concentration resulting in absence of inhomogeneity screening is an important condition for the observation of effects due to EBM in semiconductors. Therefore we have chosen an alloy with tin content $x = 0.25$. In this material the Fermi level is pinned within the band gap, and initially the sample is in the dielectric state [6]. The external infrared illumination allows to change smoothly the free electron concentration and to observe the effects due to EBM screening.

We report on the high-field ($H < 38$ T) magnetoresistance and Hall effect measurements of $Pb_{0.75}Sn_{0.25}Te(In)$ in the photomemory regime for different levels of initial photoexcitation at the temperature $T = 4.2$ K.

The samples used in this investigation were monocrystals with the indium content varying between 0.2 and 0.5 at%. They were mounted in a special chamber that screened completely the background radiation. The chamber was cooled by liquid helium. An internal thermal source of infrared radiation provided the controlled change of free carrier concentration.

The high-field magnet allows field pulses of different shapes up to 40 T. The pulse length is up to 1.5 s. As it is virtually impossible to commutate the field direction in the magnet, a complete Hall effect measurement (i.e. for both field directions) cannot be performed. Such experiments were performed in a superconducting magnet generating fields up to 5 T.

The temperature dependence of the resistivity of all the $Pb_{0.75}Sn_{0.25}Te(In)$ samples has the well-known characteristics [6]. There are two pronounced activation parts; one (70–300 K) corresponds to the thermal electron–hole generation via the band gap, the other (15–50 K) to

the activation from impurity centers. For the alloys with $N_{In} < 0.4$ at%, the lowering of the temperature below 15 K results in thermal "freezing" of electrons in the conduction band. The amount of "frozen" electrons depends on N_{In} and on the cooling rate [7]. For the sample with $N_{In} = 0.5$ at% the "darkness" free electron concentration does not exceed 10^6 cm^{-3} at $T =$ 4.2 K [6].

For all the samples it is possible to change the free electron concentration n (or zero-field resistance ρ_0) by means of infrared radiation. The effects we observe do not depend on whether the nonequilibrium electrons are photogenerated or "frozen". The results are defined mainly by the value of n, and not by N_{In}.

The typical magnetic field and magnetoresistance time profiles are shown in fig. 1. It was found that for $\rho_0 > 1\ \Omega$ cm, the sample resistance increases in time after the moment t_0 of magnetic field stabilization. Arrows in fig. 1 correspond to t_0 for the respective $\rho(t)$ curves. The resistance rises exponentially in time during the pulse (fig. 2),

$$\rho = \rho_{(t0)} + \Delta\rho[1 - \exp(-(t - t_0)/\tau)) . \qquad (1)$$

Here $\Delta\rho = \rho_{(t\to\infty)} - \rho_{(t_0)}$. For every fixed ρ_0 the characteristic time τ depends exponentially on the magnetic field applied (fig. 3),

$$\tau = \tau_{(H=0)} \exp(H/H_0) , \qquad (2)$$

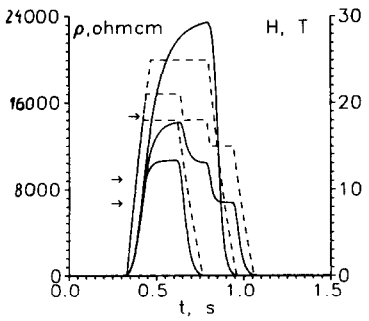

Fig. 1. Magnetoresistance and magnetic field time profiles. The solid lines correspond to the magnetoresistance, the dashed lines correspond to the magnetic field. The arrows indicate the moment of magnetic field stabilization t_0 for the respective $\rho(H)$ curves. $\rho_0 = 25\ \Omega$ cm.

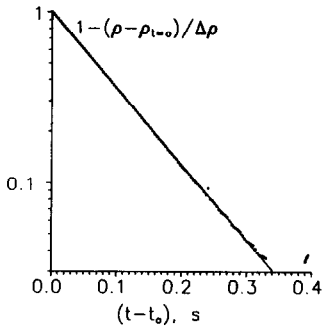

Fig. 2. Dependence of $[1 - (\rho - \rho(t = t_0))/\Delta\rho]$ on time $(t - t_0)$ after magnetic field stabilization, on a semilog scale; $H = 25$ T, $\rho_0 = 25\ \Omega$ cm.

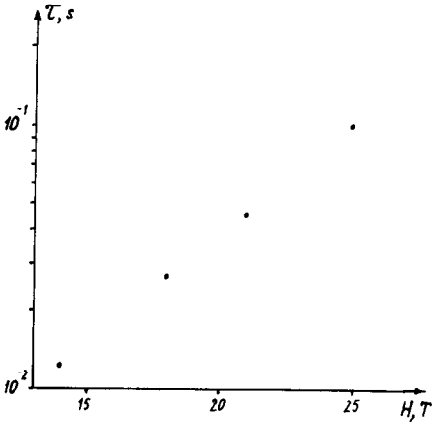

Fig. 3. Magnetic field dependence of the characteristic time τ. $\rho_0 = 25\ \Omega$ cm.

where $\tau_{(H=0)}$ and H_0 are constants for a given ρ_0.

The measurements show that for every given H the value of τ decreases if the free electron concentration rises (or ρ_0 drops). If $\rho_0 < 1\ \Omega$ cm, then the effect described above is not observed even for the highest values of $H = 38$ T and $\partial H/\partial t = 80$ T/s that we have used in our experiment.

This leads us to conclude that we have observed the localization of nonequilibrium free carriers in the magnetic field, its characteristic time τ being defined by ρ_0 and H. However, from the magnetoresistance measurements it remained unclear whether the localization is also present in relatively low fields, where τ is much smaller than the time of magnetic field increase or decrease (~0.1 s). The answer could be given

by measurements of the field dependence of the Hall coefficient. It turned out, however, that these measurements can be performed only if $\rho_0 < 1\ \Omega$ cm. For higher ρ_0 the contribution of the magnetoresistance U_{mr} to the sensed voltage at the Hall contacts is much higher than the contribution of the Hall voltage U_H itself, and accurate measurements are impossible.

For $\rho_0 < 1\ \Omega$ cm, $U_{mr} < U_H$ as a result of the Hall mobility rise, and the measurement accuracy (~10%) becomes satisfactory. Although under these conditions the magnetoresistance relaxation in time is not already observed, we expected to detect a "fast" localization of nonequilibrium electrons. In this case the field dependence of U_H should be superlinear. It turned out, however, that the $U_H(H)$ curve is sublinear, and, moreover, it seems to saturate near a field of ~38 T (fig. 4). This means that the free electron concentration increases in a magnetic field, for a relatively high degree of photoexcitation.

In order to detect more carefully the localization–delocalization transition described above, we have measured the field dependence of the Hall coefficient R_H for different ρ_0 in the superconducting magnet. It was observed that if the free electron concentration is small enough, then R_H initially drops (fig. 5). Subsequently R_H reaches a minimum at $H = H_{min}$ and rises with further increase of H.

With decreasing ρ_0 the amplitude of the initial R_H drop becomes smaller, and the value of H_{min} increases. At $\rho_0 < 10\ \Omega$ cm only a slight decrease

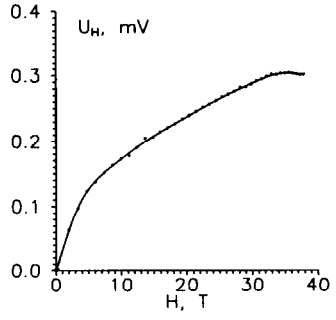

Fig. 4. Dependence of the Hall voltage U_H on the magnetic field H. $\rho_0 = 0.5\ \Omega$ cm.

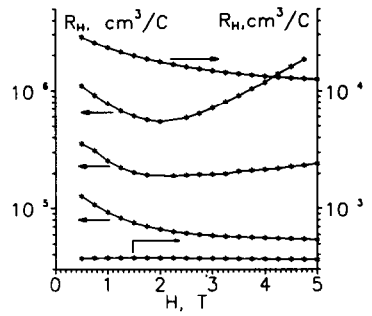

Fig. 5. Dependence of the Hall coefficient R_H on the magnetic field H for different degrees of initial sample photoexcitation.

of the Hall coefficient is detected for the magnetic field range available (fig. 5).

In summary, we have observed three types of galvanomagnetic-effect peculiarities: (i) a steep decrease of R_H in a small field, (ii) localization in a strong field, and (iii) sublinear $U_H(H)$ dependence for low ρ_0, on the contrary, reflecting the free electron concentration rise in the field.

In our opinion the localization is the first observation of effects resulting from energy band modulation in $Pb_{1-x}Sn_xTe(In)$, notably from the growth of its amplitude E_{EBM} with magnetic field. The shift of the localization region to higher fields for higher degree of photoexcitation is the most strong argument confirming this statement. Indeed, the nonequilibrium free carriers screen the inhomogeneities, and therefore E_{EBM} decreases as ρ_0 drops.

It is known that the activation energy of the ground impurity state E_2 depends linearly on H, at least, in fields up to 5 T [9]. The value of E_{EBM} is defined by the fluctuations of the E_2 position with respect to the conduction band edge. Therefore it is natural to consider E_{EBM} as depending linearly on H. If the localization process is activation-like, then

$$\tau = \tau_0 \exp[(E_{EBM} + \alpha H)/k_B T], \qquad (3)$$

where $\alpha = \partial E_{EBM}/\partial H$, T is the sample temperature (4.2 K) and $\tau_0 = \text{const.}$ for a given ρ_0. Comparing (2) and (3) one obtains

$$\alpha = kT/H_0 . \qquad (4)$$

For $\rho_0 = 25\ \Omega$ cm (figs. 1–3) corresponding to the free electron concentration $n = 10^{14}$ cm^{-3}, $\alpha = 0.065$ meV/T. The value of α is about one order of magnitude smaller than $\partial E_2/\partial H = (0.5–0.6)$ meV/T [8]. Consequently, one may assume that for a given n, $E_{EBM}(H=0)$ is smaller by approximately the same factor than the activation energy of the ground impurity state $E_2 = 20$ meV in $Pb_{0.75}Sn_{0.25}Te(In)$ [6].

More precise estimation of E_{EBM} requires a measurement of the temperature dependence of τ for different values of ρ_0.

Let us now consider other peculiarities of galvanomagnetic effects. In the case of free carrier degenerate statistics and monopolar conductivity, the nonlinear $U_H(H)$ dependence indicates the existence of quasilocal states with an energy equal to the Fermi energy E_F. The change in the energy position of these states induced by a magnetic field may lead to both localization and delocalization of electrons.

It is known that there exist two types of impurity states in $Pb_{1-x}Sn_xTe(In)$. The ground two-electron state E_2 provides Fermi level pinning effect. The metastable one-electron state E_1 [9] manifests itself in a range of effects [9–11]. In particular, the nature of the giant negative magnetoresistance effect in dielectric $Pb_{1-x}Sn_x$-Te(In) is defined by the motion of the metastable level with respect to the conduction band bottom for relatively low $H < 0.5$ T. The states E_1 act as traps for the injected electrons. The ejection of electrons from the traps to the conduction band in a magnetic field results in this effect [10]. The initial Hall coefficient drop in a small magnetic field (fig. 5) is likely to have the same origin. Some difference with the effect observed in ref. [10] results from the fact that not only injected but also part of the photogenerated electrons may be trapped. As the concentration of free photo(thermo)excited electrons grows, the contribution of devastating electrons to the conductivity decreases, and the initial drop of R_H diminishes.

The high-field $U_H(H)$ nonlinearity also results from the electron transitions between the E_1 level and the conduction band. The electron concentration rise has in this case several origins. First of all, the E_1 level may shift with respect to the conduction band bottom in a magnetic field. Secondly, the magnetic field may influence the E_1 width, and the change of the level density of states at the quasi-Fermi level also may lead to the electron delocalization. Finally, the conduction band density of states rises substantially in the magnetic field. The existence of some quasilocal levels with the energy close to E_F may result in the electron transitions from this level to the band. Effects of this kind have been observed in $Pb_{0.8}Sn_{0.2}Te(In)$, where the Fermi level is pinned within the conduction band [12].

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