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Scattering mechanisms of electrons in monocrystalline PbTe, PbSe and PbS

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Abstract. The theoretical analysis of carrier scattering mechanisms in electronic lead chalcogenide crystals was carried out. The calculation of carrier mobility in wide temperature (4.2–300 K) and concentration (10^{16}–10^{20} cm^{-3}) ranges is carried out from the viewpoint of interaction of conductivity electrons with deformation potentials of acoustic and optical phonons, polarizing potential of optical phonons, screening Coulombic and short-range potentials of vacancies. It has been shown that the agreement of theoretical and experimental results takes place when taking into account the carrier scattering both on phonons and ionized vacancies.

Keywords: vacancies, optical and acoustic phonons, lead chalcogenides, scattering mechanism, interaction.

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1. Introduction

The exceeding scattering mechanisms in lead chalcogenides crystals of n-type conductivity were investigated in various papers [1-25], but the separation and difference at the obtained results requires an essential correction. The analysis of experimental data upon transport phenomena (mobility, thermal-e.m.f., Nernst-Ettinhshausen effect, to an electronic thermal conductivity) shows that, in lead chalcogenide crystals the carrier scattering caused by interaction with proper point defects and thermal oscillations of a crystalline lattice [1-7] are dominating. The relative contribution of these mechanisms is reduced to the following.

According to majorities of the researches carried out, in the very wide temperature range (beginning from 1 K and to higher), with the greater or smaller exactness, it is possible to limit ourselves by the carrier scattering on acoustic phonons [1-10]. This conclusion is made by comparison between calculated and experimental dependences of kinetic coefficients on temperature and concentration. In accord to [1,11], the mobility limited by scattering on acoustic phonons should vary under the $T^{-5/2}$ law, which is observed in experiment. The same conclusion about significant role of scattering electrons on acoustic phonons, down to low temperature fields, indicate existence of superconductivity properties of $A^{IV}B^{VI}$ compounds [8,9]. In the vicinity of the room temperature, the analysis of data on thermoelectromotive is possible to explain it by a combination of scattering on acoustic and optical phonons [1,5,8,9].

The scattering on optical phonons is considered taking into account two components: polar and deformational optical phonons.

The scattering on short-wave phonons with passage of carriers between equivalent valleys is an improbable process for electronic crystals. This aspect of scattering is realized with a higher probability in lead chalcogenides of the *p*-type conductivity [1].

In the field of low temperatures, the carriers of a current are dispersed mainly on charged vacancies [1,2]. Both the scattering on screening Coulomb potential of vacancies predominates in the field of low concentration and dominating becomes scattering carriers on a short-range potential of vacancies (interior part of Coulomb potential, which operates on lengths close to a constant of lattice) in the field of high concentration.

The exceeding of scattering mechanisms in semiconductors, as a rule, determine from associations of relaxation time from an energy, carriers concentration, or tem-

perature [1,2]. At a simultaneous realization of different alternative mechanisms scattering the summarized relaxation time is determined by Mattisen rule [13]:

$$\tau^{-1} = \sum_i \tau_i^{-1}, \tag{1}$$

where τ_i is relaxation time for i-type mechanism of scattering.

At the calculated used Cane's laws of dispersion:

$$\varepsilon(k) = \frac{\hbar^2 k^2}{2m^*}, \tag{2}$$

$$\varepsilon(k) = \frac{\hbar^2 k^2}{2m^*} + \frac{1}{2}\varepsilon_G + \sqrt{\varepsilon_G^2 + \frac{4\hbar^2}{m^{*2}} k^2 p^2}, \tag{3}$$

where (2) correspond to parabolic law of dispersion and (3) correspond of nonparabolic law of dispersion, accordingly. At these expansion k is the wave vector, $\hbar$ is Plank's constant, m^* – the effective mass of the charge carrier, ε_G is forbidden gap.

The analyses of received results led from compare theoretical and experimental data of current mobility, what calculated by:

$$\mu_H = \frac{e}{m^*} \tau I^m_{n,k}(z;\beta), \tag{13}$$

where $I^m_{n,k}(z;\beta)$ are two-parameter Fermi's integrals, tabulated in [13], τ is the relaxation time for each separate mechanism of scattering or when taking into account contribution of several alternative mechanisms simultaneously, according to (1), e is the electron charge.

2. Theoretical representations of carriers scattering

2.1. Relaxation time for scattering on acoustic phonons

The relaxation time for electrons dispersed on the deformational potential of acoustic phonons, when using the Cane law of dispersion, is determined by the relation [4]:

$$\tau_a(x) = \frac{\tau_{0,v}(x+\beta x^2)^{-1/2}}{(1+2\beta x)[(1-A)^2 - B]}, \tag{4}$$

where $\tau_{0,a} = 2\pi\hbar^4 C_l / E_{oc}^2 (2m^* k_0 T)^{3/2}$,

$A = \beta x(1-K)/(1+2\beta x)$, $B = 8\beta x(1+\beta x)K/3(1+2\beta x)^2$.

Here E_{ac} – deformational potential of the conductivity band, C_l – combination of elastic constants, $K = 1.5$ is ratio of deformational potentials of acoustic phonons for conductivity and valence bands, m^* – carrier effective mass, T – temperature, k_0 – the Boltzmann constant, $x = \varepsilon_0 / k_0 T$ – reduced energy, $\beta = \varepsilon / \varepsilon_G$ – parameter of nonparabolicity, ε_G – width of forbidden band, ε – carrier energy (according to the law of variance).

2.2. The scattering of carriers on polar optical phonons

In the case of a simple isotropic parabolic model, the relaxation time for polar longitudinal optical phonons has the form [5]

$$\tau_{opt}^{-1} \sim \frac{1}{vk^2} \int_0^{2k} q\,dq, \tag{5}$$

where v – velocity of carriers, if the integral takes into account all phonons with a wave vector q,

$$\tau_{opt}^{-1} = \frac{2k_0 T e^2 (\varepsilon_\infty^{-1} - \varepsilon_0^{-1})}{\hbar^2 v}, \tag{6}$$

where ε_∞ and ε_0 – high-frequency and static dielectric constants, accordingly. The difference $\varepsilon_\infty^{-1} - \varepsilon_0^{-1}$ is determined by polarity of crystal and determines a longwave part of a potential polar mode of lattice oscillations.

Significant difference between values ε_0 and ε_∞ is motive necessity of the registration of screening effects [1-4].

The registration screening effects is carried out with the factor [5]

$$1 - \delta_\infty \ln(1 + \delta_\infty^{-1}) \tag{7}$$

where $\delta_\infty = (2kr_\infty)^{-2}$.

The account of nonparabolicity in the two-band model gives the following expression for the radius of screening r_∞

$$r_\infty^{-2} = \frac{2^{5/2} e^2 m_d^{*3/2}}{\pi\hbar^3 \varepsilon_\infty} \int_0^\infty \left(-\frac{\partial f}{\partial \varepsilon}\right) \left(\varepsilon + \frac{\varepsilon^2}{\varepsilon_G}\right)^{1/2} \left(1 + 2\frac{\varepsilon}{\varepsilon_G}\right) d\varepsilon. \tag{8}$$

In cases that concern values $kr_\infty \approx 1$, the screening effects carry on to a diminution of appropriate values of a relaxation time by the indicated factor twice. Besides the power association of a relaxation time also varies through a power association δ_∞.

Combining the screening effects and nonparabolic ones, the authors [1,5] have received an expression of a relaxation time for the case of polar scattering

$$\tau_{opt}^{-1} = \frac{2^{1/2} e^2 k_0 T m_{d1}^{*1/2}}{\hbar^2 \varepsilon^{1/2}} \left(\varepsilon_\infty^{-1} - \varepsilon_0^{-1}\right) \frac{1 + (2\varepsilon/\varepsilon_G)}{[1 + (\varepsilon/\varepsilon_G)]^{1/2}} \times$$
$$\times \left\{ \left[1 - \delta_\infty \ln(1 + \delta_\infty^{-1})\right] - \right.$$
$$\left. - \frac{2\varepsilon(\varepsilon_G + \varepsilon)}{(\varepsilon_G + \varepsilon)^2} \left[1 - 2\delta_\infty + 2\delta_\infty^2 \ln(1 + \delta_\infty^2)\right] \right\}. \tag{9}$$

In the formulae that determine polar optical phonons, there are no adjustable parameters, therefore, their contribution can be estimated rather precisely.

2.3. Scattering on polar optical phonons at the Debye temperature

Near Debye temperatures, relaxations cannot be used. However, account of nonparabolicity and the use of a quasielastic approximation gives the following expression for the mobility [3]

$$\mu = \frac{k_F \hbar^3 \left(e^z - 1\right)^2}{2ek_0 T m_0^{*2} \left(1 + \frac{2\varepsilon_F}{\varepsilon_G}\right)\left(\varepsilon_\infty^{-1} - \varepsilon_0^{-1}\right) e^z z^2 d_{00}}. \qquad (10)$$

Such labels are used here:

$$d_{00} = S_1 - \frac{2\varepsilon_F \left(\varepsilon_G + \varepsilon_F\right)}{\left(\varepsilon_G + 2\varepsilon_F\right)^2} S_2,$$

$$S_1 = \frac{\left(e^z - 1\right)^2}{e^z \left(2k_F r_e\right)^2} \int_0^{(2k_F r_e)^2} \frac{\exp\left[z\left(\frac{t}{t+1}\right)^{1/2}\right]\left(\frac{t}{t+1}\right)^2 dt}{\left\{\exp\left[z\left(\frac{t}{t+1}\right)^{1/2}\right] - 1\right\}^2}.$$

S_2 is obtained with S_1 by multiplication integral of expression on $\frac{2t}{\left(2k_F r_e\right)^2}$, ε_F – carriers energy on the Fermi level. The changeable integration t arises through replacement of an element of integration for q on $t = \left(qr_e\right)^2$. Parameter $k_F = \left(\frac{3\pi^2 n}{N}\right)^{1/3}$ – wave vector on the Fermi level, $r_e = \left(\frac{\varepsilon_\infty}{4\pi e^2 \rho(\varepsilon_F)}\right)^{1/2}$ – radius of screening, $\rho(\varepsilon_F)$ – density of states on the Fermi level (in all ellipsoids).

2.4. Scattering of carriers on the deformational potential of optical phonons

To recent time deformational optical phonons in crystals lead chalcogenides were not practically taken into account. For the first time, some attention was paid to them by Zayachuk [2] when analyzing the carrier mobility in *n*-PbTe crystals at 300 K. He found that this mechanism can be dominating in a definite concentration range.

A possible contribution of the deformational potential of optical phonons can be represented according to (3), using relations for τ_0 [2] in the following form:

$$\tau_{0,o} = 2\hbar^2 \alpha^2 \rho (\hbar\omega_0)^2 / \pi (2m_n k_0 T)^{3/2} E_{oc}^2, \qquad (11)$$

where $K_0 = E_{ov} / E_{oc}$, α is the lattice constant, ρ – density of crystal, ω_0 – frequency of an optical phonon, E_{ov} and E_{oc} – constants of deformational potential for valence and conductivity bands, accordingly.

2.5. Scattering of carriers on screening Coulomb potential of vacancies

The relaxation time of carrier scattering on the screening Coulomb potential inherent to vacancies can be calculated according to [2]:

$$\tau_{Coul}(x) = \frac{\varepsilon_0^2 (2m^*)^{1/2} (k_0 T)^{3/2}}{\pi (Ze^2)^2 N_v \left[Ln(1+\xi) - \xi/(1+\xi)\right]} \times$$

$$\times \frac{(x + \beta x^2)^{3/2}}{(1 + 2\beta x)}, \qquad (12)$$

where $\xi = (2kr_v)^2$, k is the wave vector of carriers, r_v – is radius of screening of vacancies potential, $x = \varepsilon / k_0 T$ – reduced energy, $\beta = \varepsilon / \varepsilon_G$ – parameter of nonparabolic. The concentration of vacancies is assumed to be equal $N_v = 2 \cdot 10^{18}$ cm^{-3} for $n < 4 \cdot 10^{18}$ cm^{-3}, and $N_v = n/2$ for $n \geq 4 \cdot 10^{18}$ cm^{-3}.

2.6. Scattering of carriers on the short-range potential of vacancies

The scattering of carriers on a short-range potential of vacancies, as well as scattering on deformational potential of acoustic phonons, can be expressed as in (4) with a replacement τ_{0a} on τ_{core}:

$$\tau_{0,core} = \pi \cdot \hbar^4 / m^* (2m^* k_0 T)^{1/2} U_{VC}^2 N_v. \qquad (13)$$

3. Results

From Figs 1, 2 and Table one can see that the account of all mentioned scattering mechanisms gives a good accordance between theoretical and experimental results, and correctly describes appearances of transposition in lead chalcogenides electronic crystals.

Carrier scattering on vacancies (Fig. 1, curves 2, 3, Fig. 2, curves 3, 4, Fig. 2, curve 5, table) and their short-range potential in the part that operates on lengths close to the lattice constant (Fig. 1, Fig. 2, curve 1, the table)) gives a noticeable contribution at the temperatures 77–300 K only in the of high concentrations (~10^{19} cm^{-3} and higher). At the liquid helium temperature, scattering on vacancies is dominating and to a great extent determines appearances of transposition. Such conclusion coincides with results of antecedent researches, in particular, with those by Ravych [3,4] and Zayachuk [2]. The scattering

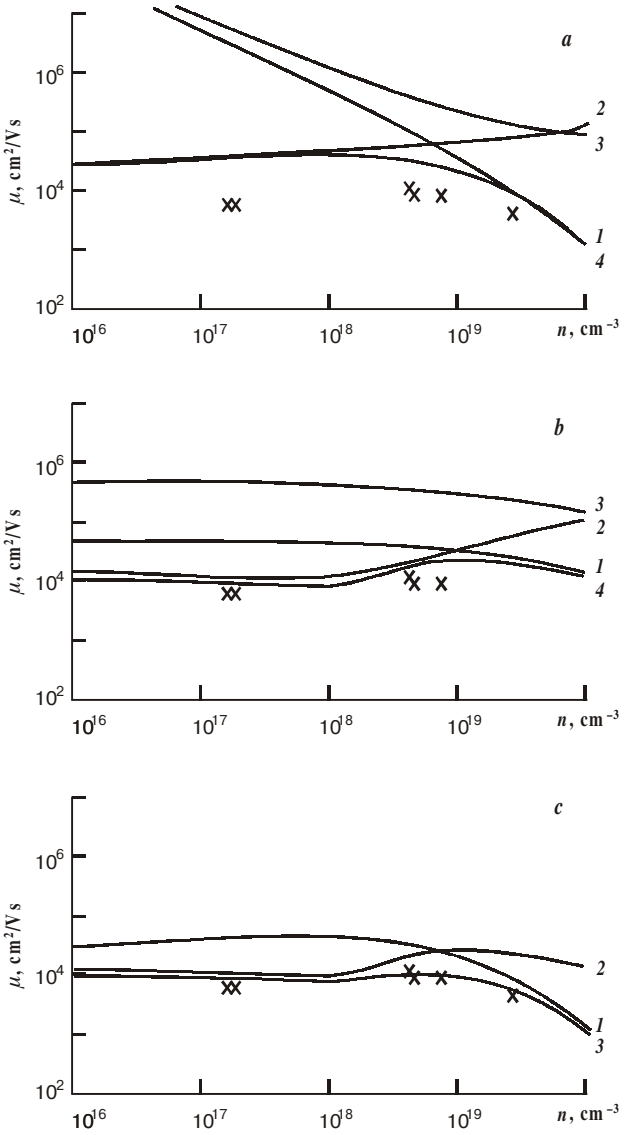

Fig. 1. Relation between the current carrier mobility and their concentration in PbS crystal at the temperature 77 K: *a* – in view of scattering carriers on: *1* – short-range potential of vacancies, *2*, *3* – Coulomb potential of vacancies, *4* – summarized scattering on *1–3*; *b* – in view of scattering free carriers on phonons: *1* – on deformational potential of acoustic phonons, *2* – on polarizing potential of optical phonons, *3* – on deformational a potential of optical phonons, *4* – summarized scattering on *1–3*; *c* – for summarized mechanisms of scattering: *1* – registration of summarized scattering of carriers on potentials of vacancies, *2* – registration of summarized scattering through an electron-phonon interaction, *3* – simultaneous registration of all aspects of scattering (till curve *1–2*). Experimental values from [15].

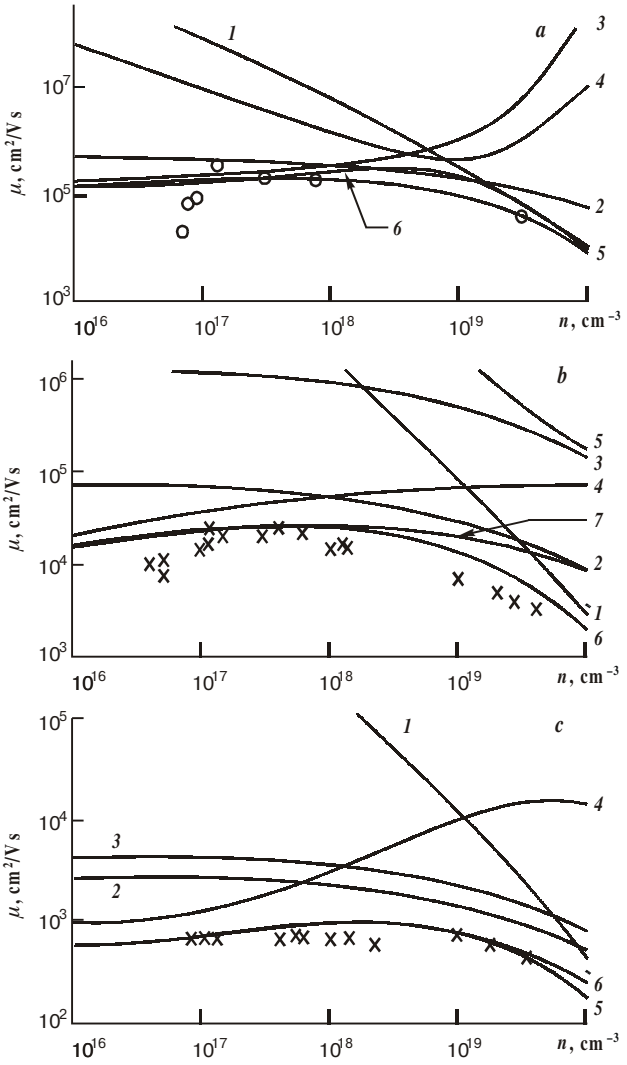

Fig. 2. Relation between the current carrier mobility and their concentration in PbS crystal at temperatures: *a* – 4.2 K, in view of scattering free carriers on: *1* – short-range potential of vacancies, *2* – deformational potential of acoustic phonons (and together with it summarized scattering on oscillations crystalline lattice), *3* – coulomb potential of vacancies (at $N_v = 2 \cdot 10^{18}$ cm^{-3}), *4* – Coulomb potential of vacancies (at $N_v = n/2$), *5* – simultaneous registration of all aspects of scattering, *5* – summarized scattering on vacancies (till a curve *1*, *3*, *4*); *b* – 77 K, in view of scattering carriers on: *1* – short-range potential of vacancies (and together with its summarized scattering on vacancies), *2* – deformational potential of acoustic phonons, *3* – deformational potential of optical phonons, *4* – polarizing potential of optical phonons; *5* – coulomb potential of vacancies (at $N_v = 2 \cdot 10^{18}$ cm^{-3}), *6* – simultaneous registration of all aspects of scattering (till a curve *1–5*), *7* – summarized scattering of carriers on phonons (till a curve *2–4*); *c* – 300 K, in view of scattering carriers on: *1* – short-range potential of vacancies (and together with it summarized scattering on vacancies), *2* – deformational potential of acoustic phonons, *3* – deformational potential of optical phonons, *4* – polarizing potential of optical phonons; *5* – simultaneous registration of all aspects of scattering (till a curve *1–4*), *6* – summarized scattering of carriers on phonons (till a curve *2–4*). Experimental values are taken from [15,16].

Table. Concentration and temperature limits of dominant separate scattering mechanisms on lead chalcogenides electronic crystals

	4.2 K	77 K	300 K
	Screening Coulomb potential of vacancies		
PbS	$1\cdot10^{16}$-$5\cdot10^{18}$, $5\cdot10^{18}$-$1\cdot10^{19}$	$1\cdot10^{16}$-$5\cdot10^{17}$	–
PbSe	$1\cdot10^{16}$-$2\cdot10^{18}$, $7\cdot10^{18}$-$1\cdot10^{19}$	–	–
PbTe	$1\cdot10^{17}$-$2\cdot10^{17}$	–	–
	Short-range potential of vacancies		
PbS	$1\cdot10^{19}$-$1\cdot10^{20}$	$2\cdot10^{19}$-$1\cdot10^{20}$	$8\cdot10^{19}$-$1\cdot10^{20}$
PbSe	$2\cdot10^{19}$-$1\cdot10^{20}$	$5\cdot10^{19}$-$1\cdot10^{20}$	$8\cdot10^{19}$-$1\cdot10^{20}$
PbTe	$1\cdot10^{19}$-$1\cdot10^{20}$	$8.5\cdot10^{19}$-$1\cdot10^{20}$	$>1\cdot10^{20}$
	Polar potential of the optical phonons		
PbS	–	$1\cdot10^{16}$-$8\cdot10^{18}$	$1\cdot10^{16}$-$1\cdot10^{18}$
PbSe	–	$1\cdot10^{16}$-$8\cdot10^{17}$	$1\cdot10^{16}$-$6\cdot10^{17}$
PbTe	–	$1\cdot10^{16}$-$8.5\cdot10^{17}$	$1\cdot10^{16}$-$7\cdot10^{18}$
	Deformational potential of optical phonons		
PbS	–	–	$5\cdot10^{18}$-$1\cdot10^{20}$
PbSe	–	–	$1\cdot10^{19}$-$2{,}5\cdot10^{19}$
PbTe	–	–	$8\cdot10^{18}$-$1\cdot10^{20}$
	Deformational potential of acoustic phonons		
PbS	$5\cdot10^{18}$-$5\cdot10^{19}$	$1\cdot10^{16}$-$1\cdot10^{20}$	$5\cdot10^{18}$-$1\cdot10^{20}$
PbSe	$1\cdot10^{16}$-$8\cdot10^{19}$	$1\cdot10^{17}$-$7\cdot10^{19}$	$6\cdot10^{17}$-$1\cdot10^{20}$
PbTe	$1\cdot10^{16}$-$1\cdot10^{20}$	$8\cdot10^{17}$-$1\cdot10^{20}$	$\sim 1\cdot10^{20}$

of carriers on polar optical phonons is dominating in the field of low and average concentrations at temperatures 77 and 300 K (Figs 1 and 2, curve 4, table). At higher carrier concentrations their influence on appearance of transposition decreases, and mechanisms of scattering on the deformational potential of acoustic and optical phonons predominate. The relative share of scattering on optical polar phonons decreases with lowering the temperature below the nitrogen one. At $T < 10$ K polar optical phonons are not excited, therefore polar scattering is insignificant. The account of this mechanism gives the actual agreement between the theory and experiment in the temperature range 20–300 K. The essential role of optical polar phonons at these temperatures is confirmed by an experimentally observable diminution of the carrier mobility at low concentrations (Figs 1, 2).

The carrier scattering on the deformational potential of optical phonons up to date was not practically taken into account, though, as was shown in [2], at 300 K it is rather essential. The researches carried out by us confirm the important role of this scattering mechanism at room temperatures for lead chalcogenide crystals. The use of expression (10) for a considered relaxation time in a combination with polar optical and acoustic phonons enables to reach the good agreement between the theory and experiment (Fig. 2, curve 3). With this purpose, to describe τ_0, using the expressions (10) magnitude E_{oc} should be taken equal to 26 eV. The contribution of the optical deformational potential, as seen from Fig. 2 (curve 3), at the room temperature is rather essential, and at high concentrations is even dominating in carrier mobility. At temperatures of liquid helium, scattering on optical deformational phonons is less essential (Fig. 1b, Fig. 2b,c, curve 3).

Acoustic phonons influence on kinetic parameters of lead chalcogenides crystals in all the considered temperature (4.2–300 K) and concentration (10^{16}–10^{20} cm^{-3}) ranges. Let us note that, in the low concentration range, some deviation of experimental results from data of calculations (Fig. 1, 2) takes place. Therefore, it is possible to assume the necessity of considering inelastic effects when evaluating scattering mechanisms.

4. Conclusions

1. The dominating mechanisms of carrier scattering in electronic lead chalcogenide crystals are both scattering on vacancies and on thermal oscillations of the lattice. In the range of low temperatures, carrier scattering on vacancies predominates, while at high temperatures – on thermal oscillations of crystalline lattice.

2. Role of optical polar phonons are significant at temperatures 77 and 300 K for concentrations 10^{16}–

10^{18} cm^{-3}. At magnification of concentration the scattering on optical phonons is hindered due to screening.

3. At high concentrations (higher than 10^{19} cm^{-3}), the scattering on optical phonons is exhibited due to their deformational potential, the role of which in the total scattering at definite concentrations (see Table) becomes most essential at the room temperature, in particular, for lead sulphides and tellurides.

4. The scattering of carriers on acoustic phonons becomes essential at all considered temperatures in the whole researched concentration range. In the field of low concentrations at helium temperatures, for the agreement of theoretical calculations with experimental data, it is necessary to take into account inelastic effects at scattering of carriers on acoustic phonons.

5. The scattering on a short-range potential of vacancies is realized at high concentrations, and with increase of temperature the prevalence of this mechanism is displaced into the side of higher concentration values and decreases in the series PbS-PbSe-PbTe.

6. The Coulomb potential of vacancies dominates at low concentration and it payment in the total scattering, as in the case of a short-range potential of vacancies, decreases in the series PbS-PbSe-PbTe.

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