

# CONCENTRATION MECHANISM OF PIEZOPHOTOCONDUCTIVITY

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The results of the studies of piezophotoconductance of semiconductors, i.e. a component of electroconductance which is proportional to the light intensity times the mechanical strain, are reported. The experimental researches have been carried out making use of crystals of the layered structure  $\text{In}_4\text{Se}_3$ . The oscillation character of the piezophotoconductance spectrum at the light frequency corresponding to interband absorption has been revealed. The phase shift of piezophotoconductance with respect to the phase of the modulating factor, which may amount to  $\pi/2$ , has been established. On the basis of quantum-mechanical calculations of the probability of transitions under the combined action of light and a time-dependent mechanical strain, the piezophotoconductance spectra have been analyzed. It has been shown that the oscillation character of the piezophotoconductance spectra and retardation effects in their dynamics are caused by peculiarities of the generation of nonequilibrium current carriers and that the piezophotosensitivity depends on the lifetime of the latter.

## 1. Introduction

The electrical conductance  $\sigma$  of a semiconductor that undergoes the combined action of light and mechanical strain may contain a component which is proportional to the product of the intensity of light and the amplitude of the mechanical strain. In this case, it can be formally represented as a sum of the components proportional, respectively, to the intensity of light  $J$ , the mechanical pressure  $p$ , and the product of those two quantities, namely,

$$\sigma = \sigma_0 + \pi p + \beta J + \chi p J, \quad (1)$$

where  $\pi$ ,  $\beta$ , and  $\chi$  are the piezo-, photo-, and piezophotoconductance coefficients, respectively.

In this work, we will report the results of experimental researches of the piezophotoconductance of some semiconductors and discuss the possible mechanisms of appearance of such a component of the conductivity.

We point out that, contrary to piezomodulation spectroscopy [1], no limitations are imposed on the pressure and light intensity magnitudes, as well as on the ratio between them.

From the viewpoint of revealing the mechanisms of the piezophotoconductivity appearance, the spectra of the quantity  $\chi$  are obviously of most interest. In this connection, we should emphasize the particularly important information concerning the dynamics of such spectra which, as will be ascertained below, can serve as a basis for a new method of experimental research of a fine structure of the electron energy spectrum.

Measuring the piezophotoconductance component and its spectral dependence is not faced with any substantial difficulties, if the intensity of light and the mechanical strain depend on time harmonically. In this case, the measurements are worthwhile to be carried out at the summary or difference frequencies with respect to the modulation frequencies of the light intensity and the mechanical strain.

Concerning the choice of materials for experimental researches, we have confined ourselves to semiconductors with high photo- and piezosensitivity and required mechanical properties. Among other semiconductors, crystals of the compound  $\text{In}_4\text{Se}_3$  have been chosen, taking into account their mechanical properties [2] and the results of the previous researches of their piezophotoconductivity [3]. The results of experimental researches of the piezophotoconductance of exactly this semiconductor are mainly dealt with in this work, because they appeared typical of the others and most illustrative among those that were at our disposal ( $\text{Tl}_2\text{S}$ ,  $\text{InSe}$ ,  $\text{GaSe}$ ).

## 2. Research Method and Experimental Results

The study of piezophotoconductance spectra was carried out in a quasi-stationary regime, which was achieved by using the necessary frequencies of modulating factors. As a rule, these frequencies were selected on the assumption that the product of either the current carrier thermalization time or the relaxation time of photoconductivity and the modulation frequency should be always much smaller than unity. With this purpose in view, the mechanical strain of the lowest

possible frequency in the specimen was used, and the overwhelming majority of results were obtained in the constant light illumination regime.

The measuring equipment consisted of a source of monochromatic radiation; a block for registering the piezophotocurrent which included a resonance voltmeter; a device for measuring the phase difference; and an electromechanical device for creating mechanical strains. Specimens were placed into a cryostat and fixed by a special holder. In so doing, the holder allowed the specimen to be arranged in such a manner that either the specimen's free surface or the specimen's surface oriented normally to the created pressure was illuminated. In both the cases, the specimen was fixed between two plane-parallel quartz plates. The variable mechanical strain of low frequency was created in the specimen by applying a unidirectional pressure developed by a piezoelectric or electromechanical device.

Specimens were plane-parallel plates 0.1–0.5 mm in thickness with ohmic contacts on their end faces. The specimens' surfaces, which were to be illuminated, underwent only mechanical treatment. In the case of crystals with layered structure, the specimens under investigation had the surface of a natural chip.

The specimens were inserted into a circuit fed by constant voltage. The modulated component of the current was measured with a resonance millivoltmeter as the voltage drop across a calibrated resistor. The orthogonal components of the current were determined by synchronous detection. In so doing, the piezoconductivity signal of an unilluminated specimen, which was in phase with the current, served as a reference one for the in-phase component.

As was mentioned above, the results of researches of piezophotoconductivity in the  $\text{In}_4\text{Se}_3$  crystals turned out the most typical and illustrative ones. Those crystals possess a distinctive layered structure, which allows one to produce plane-parallel specimens with perfect natural surfaces. In addition, they have mechanical properties necessary for such researches and high piezo- and photosensitivity [4]. The width of the energy gap is 0.62 eV, the electron mobility  $500 \text{ cm}^2/(\text{V s})$ , and the hole one  $120 \text{ cm}^2/(\text{V s})$ . The lifetime of nonequilibrium current carriers does not exceed  $10^{-4} \text{ s}$  [5].

Regular studies of piezophotoconductivity in these materials revealed the following results which allowed, in general, the spectra and the  $\chi$  values to be unequivocally interpreted. The piezophotosensitivity  $\chi$  substantially depends on the impurity composition in the specimens and is maximal for specimens with intrinsic conductivity. It does not strongly depend on the piezosensitivity

and manifests itself most of all, as turned out, in the specimens, where mechanical and light excitations cooperatively affect the same electron states.

The value of the piezophotosensitivity and its spectral dependence are essentially influenced by the specimen temperature. There is a definite interval of low temperatures depending on the impurity composition and the conductivity type, where the sensitivity of the  $\text{In}_4\text{Se}_3$  crystals is maximal.

The piezophotoconductance spectra, as a rule, have oscillatory behavior with various oscillation periods. The values of the latter for the  $\text{In}_4\text{Se}_3$  crystals may differ by an order of magnitude. The structure of the piezophotoconductance spectra substantially depends on measuring conditions. In particular, their form depends not only on the temperature of the specimen, but also on the values of the mechanical strain and the initial pressure exerted upon the specimen. In Fig. 1, the spectra of the piezophotoconductance amplitude obtained for the same sample at various temperatures are presented.

The phase of the piezophotocurrent is shifted with respect to the phase of the modulating pressure by an angle which depends on the energy of a light quantum and can reach the maximal value of  $\pi/2$ . The value of the phase shift does not depend on the modulation frequency within the interval of sound frequencies.

The spectra of the orthogonal components relate to each other as a function and its derivative with respect to the energy of a light quantum. In Fig. 2, one photoconductance and two (in-phase and  $\pi/2$ -shifted) piezophotoconductance spectra of a thin  $p$ - $\text{In}_4\text{Se}_3$  specimen measured at the temperature of 200 K are shown.

The results of measurements allow us to make a preliminary conclusion about the independence of the piezophotoconductance spectrum profile of the modulation frequency within the interval of sound frequencies.

### 3. Concentration Mechanism of Piezophotoconductivity

The experimental results presented below can be interpreted on the basis of the concentration mechanism of the piezophotoconductivity considered as one of the most probable. It is based on the model describing the dynamics of the free current carrier concentration under the combined action of the light and non-stationary mechanical excitations. In this connection, let us pass to formulating the equation describing the dependence

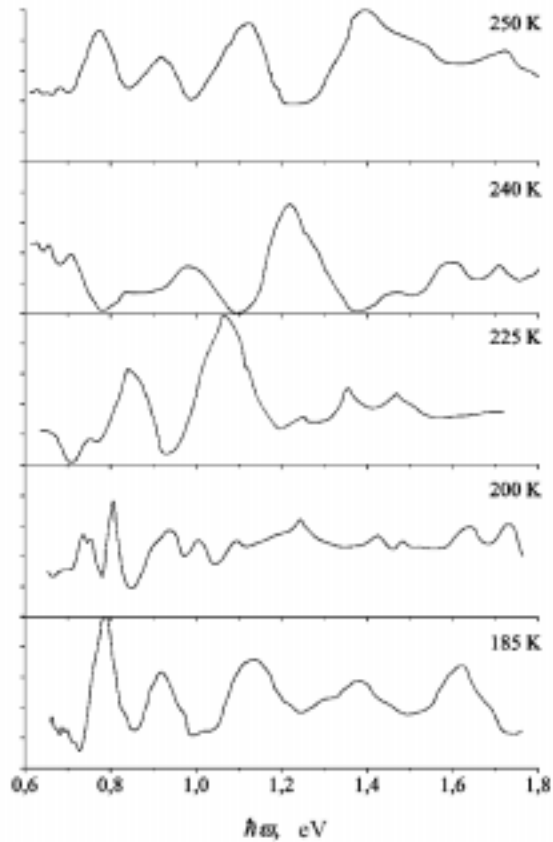


Fig. 1. Spectra of the piezophotoconductance amplitude obtained at various specimen temperatures

of the free current carrier concentration on time under those conditions. In so doing, we confine ourselves to a somewhat simplified model of the monopolar semiconductor and neglect possible variations of the current carrier mobility.

It is known [6] that a mechanical strain created in a semiconductor can result in a variation of the concentration of free charges. In the case of the ac voltage, whose oscillation period is much longer than the carrier thermalization time, and if the concentration increment depends linearly on pressure, the current carrier concentration changes in time synchronously with the pressure variation and without any substantial shift of the phase. Then the equation for the forced oscillations of the concentration  $n_p$  looks like

$$\frac{d^2 n_p}{dt^2} + \frac{1}{\tau_p} \frac{dn_p}{dt} + \Omega_0^2 n_p = -\Omega^2 g_p \sin \Omega t, \quad (2)$$

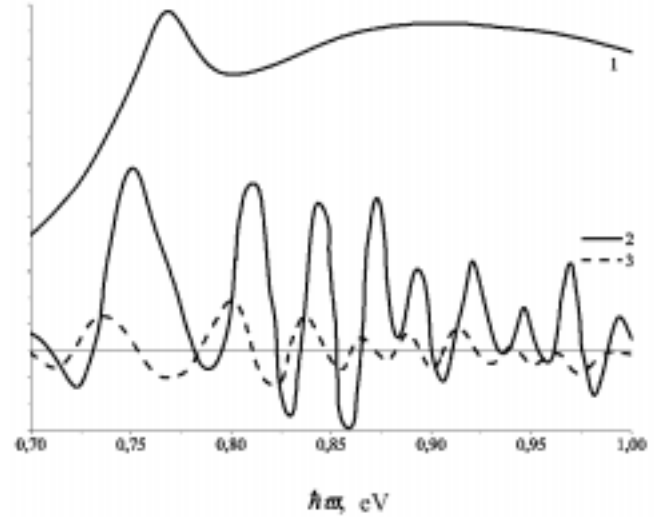


Fig. 2. Spectra of the photoconductance (1) and of the in-phase (2) and  $\pi/2$ -shifted (3) piezophotoconductances of the thin 0.1-mm specimen of  $p\text{-In}_4\text{Se}_3$  at 200 K

where  $\tau_p$  is the current carrier thermalization time,  $\Omega_0$  is the frequency of specimen free oscillations, and  $g_p$  is the peak value of the concentration increment owing to the action of the pressure  $p = p_0 \sin \Omega t$ .

It should be noted that the fast damping of free oscillations of the concentration is mainly brought about by the damping rate of the specimen free oscillations which may depend on the conditions of the specimen fixation. At the same time, the absence of a substantial phase shift of the forced oscillations of the concentration with respect to the phase of the pressure oscillations is caused by a small value of the thermalization time and a small value of the modulation frequency in comparison with the free oscillation frequency of the specimen.

The availability of light-excited nonequilibrium current carriers in the semiconductor, which undergoes the action of the alternating mechanical strain, can essentially vary the concentration dynamics as compared to the previous case. Even in the case of illuminating the specimen by a stationary flux of light within the spectral range that corresponds to interband absorption, the generation-recombination processes do not remain constant in time. This phenomenon stems from the modulating influence of the mechanical strain upon the energy gaps between the levels, between which the corresponding transitions are executed. Accordingly, the right-hand side of Eq. (2) should be appended by the terms describing acceleration or, on the contrary, deceleration of the nonequilibrium current carrier generation,  $g(t)$ , and recombination,  $R(t)$ , rates.

We notice that recombination processes cannot be a determining factor concerning the interpretation of the main experimental result, the phase shift between the mechanical pressure and the piezophotocurrent, because the lifetime of nonequilibrium carriers is much smaller than the period of pressure variation. In this connection, we confine ourselves further to the simplest case of linear recombination taking into account that, in this case, the nonequilibrium concentration is in excess with respect to the concentration created by pressure. Therefore, for the recombination rate, we have

$$R(t) = \frac{1}{\tau_0}(n - n_0 - g_p \sin \Omega t), \quad (3)$$

where  $\tau_0$  is the lifetime of nonequilibrium current carriers.

Taking these speculations into consideration, we rewrite Eq. (2) in the form

$$\frac{d^2 n}{dt^2} + \frac{1}{\tau} \frac{dn}{dt} + \Omega_0^2 n = \frac{d}{dt} g(t) - \Omega^2 g_p \sin \Omega t + \frac{\Omega}{\tau_0} g_p \cos \Omega t, \quad (4)$$

where  $\tau = \frac{\tau_0 \tau_P}{\tau_0 + \tau_P}$  is the effective relaxation time of piezophotoc conductivity.

To specify the view of the right-hand side of Eq. (4), let us consider the features of the generation of nonequilibrium carriers under the examined conditions in more details.

#### 4. Time-dependence of Nonequilibrium Current Carrier Generation Under the Conditions of Nonstationary Deformation

Let us estimate the time-dependence of the generation rate of nonequilibrium current carriers in a thin semiconductor monopolar specimen which undergoes the action of an unidirectional pressure. It is supposed that the dimensions of the specimen allow the diffusive processes of nonequilibrium current carriers to be neglected. According to this, we should pay attention to the fact that the spectral dependence of the generation rate in the quasi-stationary regime, similarly to the stationary regime case, is defined by the spectrum of the absorption coefficient. To estimate the dynamics of the latter, we carry out the corresponding calculation for a semiconductor, where the mechanical strain of extremely low frequency is available and the other above-discussed restrictions are ensured.

It is evident that the estimation of the absorption coefficient dynamics demands the information concerning the probability of interband transitions and the electron density of states under non-stationary

conditions. With this purpose in view, we analyze the Schrödinger equation for an electron under the specified conditions, putting the main emphasis on the processes which determine the variations in the energy structure of electron states. We write down the corresponding equation in the form

$$i\hbar \frac{\partial \psi(r, t)}{\partial t} = [\mathcal{H}_0 + W(r) \sin \Omega t] \psi(r, t), \quad (5)$$

where  $\mathcal{H}_0$  is the Hamiltonian of the undeformed crystal and  $W(r)$  is the peak value of the perturbing potential induced by a mechanical strain. We note that the value of  $W(r)$  under the condition of stationary squeezing (or stretching) is a sum of two terms, one of which is brought about by the variation of the crystal lattice constant and the other by the variation of the potential, both being proportional to the pressure.

We note that, prior to the pressure exertion upon the crystal, the wave function  $\psi(r, t)$  possesses the Bloch structure and corresponds to the energy  $E_0(k)$ . Since the main result of this calculation is the dependence of the generation rate at frequency  $\Omega$  on time, we confine ourselves in what follows to the estimation of the temporal dependences of the wave function and the dynamics of the probability of interband transitions.

In this connection, we make a certain simplification concerning the perturbation magnitude. It can be reasonable by two following essential circumstances. First, the time periodicity of the Hamiltonian allows the quasi-energy  $E(k)$  to be regarded as a quantity which characterizes electron states. Second, extremely low frequencies of the mechanical strain allow the solutions of Eq. (5) to be considered in the adiabatic approximation. Therefore, the wave function may be approximated as the product  $\psi(r, t) = \psi(r)\psi(t)$ . In this case, after simple calculations, we obtain the following expression for the wave function:

$$\psi(r, t) = \psi(r) \exp \left\{ -\frac{i}{\hbar} \left( E_0 t - \frac{W(k)}{\Omega} \cos(\Omega t) \right) \right\}. \quad (6)$$

Here,  $W(k)$  is the amplitude displacement of the state  $k$ , which corresponds to the energy  $E_0(k)$ . As for the latter quantity, its value is determined by specific conditions. As is known, the perturbing term in Eq. (5) can stimulate, under the stationary pressure, the energy levels to be shifted, the effective mass of current carriers to be modified, or the degeneration to be cancelled.

Let us proceed to the calculation of the interband transition probability in the linear approximation in the vector potential of the light wave [7]. Using the expression for the light perturbation operator, we obtain,

after simple calculations, the following expression for the matrix element:

$$W_{cv} = W_r \sum_{n=-\infty}^{\infty} i^n J_n(\eta) \frac{e^{\frac{i}{\hbar}(E_c - E_v - \hbar\omega - n\hbar\Omega)t} - 1}{\frac{i}{\hbar}(E_c - E_v - \hbar\omega - n\hbar\Omega)}. \quad (7)$$

Here,  $W_r$  is a multiplier calculated on the coordinate part of the matrix element with regard for the crystal deformation,  $J_n(\eta)$  is the Bessel function of the  $n$ -th order, and  $\eta = \frac{W_v(k) - W_c(k)}{\hbar\Omega}$ .

The probability of the interband transition will be calculated in view of the interest in the component of the absorption coefficient which oscillates in time with the modulation frequency  $\Omega$ . Omitting simple calculations, we give an expression for the probability of such a transition in unit time, where only those components are separated, whose oscillation frequency does not exceed  $\Omega$ :

$$P_{cv} = \frac{4}{\hbar} |W_r|^2 \left\{ J_0^2(\eta) \frac{\sin \Delta\omega t}{\Delta\omega} + J_1^2(\eta) \frac{2\Delta\omega}{\Delta\omega^2 - \Omega^2} [\sin(\Delta\omega - \Omega)t + \sin(\Delta\omega + \Omega)t] - J_0(\eta)J_1(\eta) \frac{2}{\Delta\omega(\Delta\omega^2 - \Omega^2)} \times \left[ \Omega^2 + (\Delta\omega^2 - \Omega^2) \cos \Delta\omega t \right] \cos \Omega t \right\} + P'_{cv}. \quad (8)$$

Here,  $P'_{cv}$  stands for the components of the probability which oscillate with frequencies more than  $\Omega$ .

Let us write down the next simplification of expression (8) taking into account that, according to the adopted conditions, the inequality  $\hbar\omega \gg \Omega$  always holds true. In this case, we obtain

$$P_{cv} = \frac{4\pi}{\hbar} |W_r|^2 \left\{ J_0^2(\eta) - 2J_0(\eta)J_1(\eta) \cos \Omega t \right\} \times \delta(E_v - E_c + \hbar\omega) + P'_{cv}. \quad (9)$$

We note that the phase of the probability  $P_{cv}$  (9) is shifted by  $\pi/2$  with respect to that of the modulating factor.

Now, let us return to the estimation of a variation of the density of states induced by a mechanical strain. It is evident that if the dynamic deformation is of extremely low frequency, the density of electron states available to transitions is equal to the sum of the stationary density of states and its increment at the present moment. Confining ourselves to the linear approximation of the

increment of the density of states under the examined conditions, let us turn to the accompanying reference system for calculating the number of transitions per unit time. In this case, the expression for the number of transitions per unit time looks like

$$\delta n = \frac{1}{\pi^2 \hbar} \int_{V_k} |W_r|^2 \left\{ J_0^2(\eta) - 2J_0(\eta)J_1(\eta) \cos \Omega t + J_0^2(\eta) \varepsilon \sin \Omega t \right\} \times \delta(E_v - E_c + \hbar\omega) d\tau_k + \delta n'. \quad (10)$$

Here,  $\varepsilon$  is the relative deformation of the crystal and  $\delta n'$  denotes terms with oscillation frequencies higher than  $\Omega$ . The conduction band is assumed empty.

To make expression (10) more specific, a further information concerning the types of transitions and the density of states is needed. Leaving those questions beyond the scope of this work, we can conclude that, in the case of the varying deformation in a semiconductor, the absorption coefficient has the following form:

$$\alpha(\hbar\omega) = \alpha_0(\hbar\omega) + \alpha_1(\hbar\omega) \sin \Omega t + \alpha_2(\hbar\omega) \cos \Omega t. \quad (11)$$

Analyzing this expression, we can ascertain that, in the case of the varying deformation in a semiconductor, the absorption coefficient can have a component, the phase of which is shifted with respect to that of the modulating pressure. We note that the presence of such a shift essentially depends on the argument of the Bessel function. This argument does not equal to zero in the case, where the pressure-induced energy displacements of the states, between which the optical transitions take place, are different.

## 5. Piezophotoconductance Spectra

Let us pass to the estimation of the spectral dependence of the piezophotocurrent component, by remaining within the scope of the concentration mechanism. For this purpose, we specify Eq. (4), using the results of the previous section. Taking into account that the absorption coefficient is expressed by formula (11), the generation rate of nonequilibrium current carriers can be written as

$$g(t) = g_0 + g_1 \sin \Omega t + g_2 \cos \Omega t, \quad (12)$$

where the parameters  $g_0$ ,  $g_1$ , and  $g_2$  include the components of the absorption coefficient, quantum yield, and light intensity, respectively, relevant to piezophotoconductivity.

Equation (4), taking into account expression (12), reads

$$\frac{d^2 n}{dt^2} + \frac{1}{\tau} \frac{dn}{dt} + \Omega_0^2 n = \Omega g_c \sin(\Omega t + \varphi), \quad (13)$$

where

$$g_c = \Omega \sqrt{\left(g_1 + \frac{1}{\tau} g_p\right)^2 + \left(g_2 + \Omega g_p\right)^2},$$

$$\text{tg} \varphi = -\frac{g_1 \tau + g_p}{\tau(g_2 + \Omega g_p)}, \quad (14)$$

Analogously to the previous case, Eq. (13) describes the forced oscillations of the concentration under the action of a driving factor. The phase of oscillations is shifted by  $\varphi$  with respect to the pressure phase. The spectral dependence of the angle  $\varphi$  is shown in Fig. 3.

The solution of Eq. (13) is

$$n = N \sin(\Omega t + \varphi + \delta), \quad (15)$$

where

$$N = \frac{\Omega \tau \sqrt{\left(g_1 + \frac{1}{\tau} g_p\right)^2 + \left(g_2 + \Omega g_p\right)^2}}{\sqrt{(\Omega_0^2 - \Omega^2) \tau^2 + \Omega^2}},$$

$$\text{tg} \delta = \frac{\Omega}{\tau(\Omega_0^2 - \Omega^2)}. \quad (16)$$

These results allow one to find the expression for the orthogonal components of the piezophotocurrent. Taking into account the limitations accepted before, which concern the frequency and the relaxation time, the expressions for the corresponding components of piezophotoconductance are

$$\sigma_s = \sigma_0 J_0^2(\eta) \sin \Omega t,$$

$$\sigma_a = \sigma_0 J_0(\eta) J_1(\eta) \cos \Omega t, \quad (17)$$

Here,  $\sigma_a$  and  $\sigma_s$  correspond to the in-phase and  $\pi/2$ -shifted components of the piezophotoconductance, respectively.

In Fig. 3, the spectra of the components  $\sigma_a$  and  $\sigma_s$  are presented within the interval of the light frequencies which correspond to interband absorption. As is seen from these dependences and as follows from expressions (17), both the components have oscillation behavior. We note that the angle  $\delta$  is close to zero, because  $\Omega \ll \Omega_0$ .

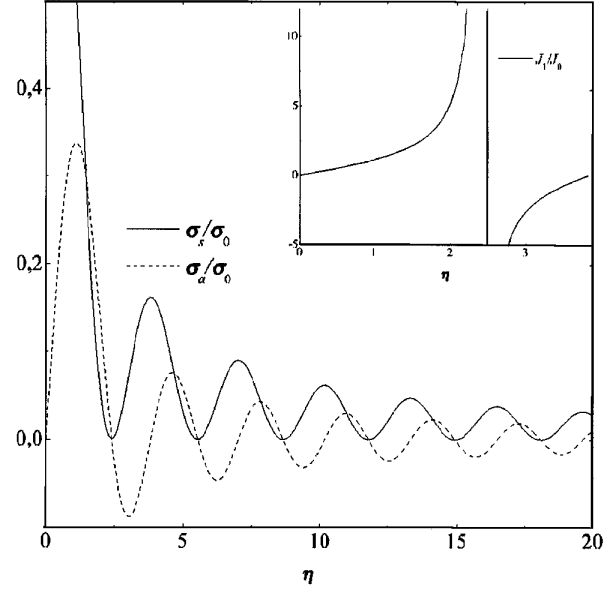


Fig. 3. Spectra of the  $\sigma_a$  and  $\sigma_s$  components and the spectral dependence of the angle  $\varphi$  (in the inset)

## 6. Concluding Remarks

In conclusion, we consider some features of the results presented above. First of all, we point out the obvious fact that the necessary condition for such a component of conductivity in semiconductors as piezophotoconductivity to exist is a combined action of a mechanical strain and light on those electron states which take part in optical transitions.

Among the presented results of the studies of piezophotoconductance spectra of  $\text{In}_4\text{Se}_3$  crystals, qualitatively new are the availability of the component, whose phase is shifted with respect to that of the modulating pressure, and the oscillation character of the spectra themselves. The phase shift of the piezophotocurrent depends on the energy of a light quantum and does not depend on the photoconductivity relaxation time and the modulation frequency within the range of extremely low frequencies.

This result unambiguously proves that the retardation effects in the piezophotocurrent dynamics are mainly brought about by the mechanism of generation of nonequilibrium current carriers under the combined action of light and a mechanical strain. This conclusion agrees with the results of calculations of the piezophotocurrent dynamics.

At last, we point out that the oscillation character of the piezophotoconductance spectra can be qualitatively

explained by the time- and energy-modulations of energy gaps, within the limits of which the optical transitions occur at the given energy of a light quantum. In fact, as the experimental researches and corresponding estimations of the piezophotocurrent spectra evidence for, the oscillation character of the spectra is not indicative, because it may depend on conditions of the experiment. Nevertheless, the presented results testify to that the phenomenon of piezophotoconductivity can be an efficient way for researching the fine structure of the energy states in semiconductors. In this regard, the quantity  $\eta$  and its spectral dependence are of importance.

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#### КОНЦЕНТРАЦІЙНИЙ МЕХАНІЗМ П'ЄЗОФОТОПРОВІДНОСТІ

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#### Резюме

Наведено результати дослідження п'єзофотопровідності — складової електропровідності, яка пропорційна добутку інтенсивності світла та механічного напруження в напівпровіднику. Експериментальні дослідження проведено на кристалах шаруватої структури  $\text{In}_4\text{Se}_3$ . Виявлено осциляційний характер спектра п'єзофотопровідності для частоти світла, що відповідає міжзонному поглинанню. Встановлено, що має місце зсув за фазою п'єзофотопровідності відносно фази модулюючого чинника, який може становити  $\pi/2$ . Проведено аналіз спектрів п'єзофотопровідності на основі квантово-механічних розрахунків імовірності переходів в умовах спільної дії світла та змінного у часі механічного напруження. Показано, що осциляційний характер спектрів п'єзофотопровідності та ефекти запізнення в їх динаміці зумовлені особливостями генерації нерівноважних носіїв струму, тоді як чутливість п'єзофотопровідності залежить від часу життя останніх.