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STUDY OF PESO - PHOTORESISTIVE CHARACTERISTICS OF $TlInSe_2$ SINGLE CRYSTALS

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Abstract:

Objective. *The aim of the study is to establish the patterns and mechanisms of the effect of various impurities on the tensoresistive characteristics of $TlInSe_2$ single crystals, as well as in solid solutions based on it. When studying the piezophotoresistive characteristics of $TlInSe_2$ single crystals, it will determine the nature of the effect of uniaxial elastic deformation on the electrical, photoelectric and tensoresistive properties of $TlInSe_2$ crystals.*

Methods. *Crystals synthesized by fusing the components in accordance with stoichiometry in evacuated (~ 10-4 mm Hg) and sealed quartz ampoules were used for the study. Highly pure elements thallium (Tl - 000), indium (In - 000) and selenium (Se - high purity grade - 17 - 4) were used as initial components for the synthesis. Single crystals were grown by the improved Bridgman method, the crystallization front velocity varied from 0.5 to 0.9 mm / h.*

The samples required for the study were prepared by cleaving the grown single crystals from an ingot by the simplest indentation with a sharp knife, the blade thickness of which is ≤ 0.01 mm, along two mutually perpendicular planes of natural cleavage. The samples were in the form of rectangular parallelepipeds with dimensions of 10x10x0.25 mm. Thus obtained "needle" crystals, without any additional processing were ready for welding contacts.

Two methods were used to solder mechanically reliable ohmic contacts on these blanks:

a) fusion of indium in a stream of inert gas, followed by brazing of copper (or nickel) wires ($\varnothing$ = 0.01 mm).

b) direct spot welding of the corresponding wires with a capacitor discharge to the ends of the blanks heated in a flow of an inert gas. The second method turned out to be more efficient and reliable (especially for moderate temperatures).

Plates of steel 45 with a thickness of 0.5 - 1.0 mm and a length of 20 - 80 mm served as calibration beams for the glued sensors. The surface of the substrate according to the processing class was at least 7.

Findings. *The nature of the influence of uniaxial elastic deformation on the electrical, photoelectric and tensoresistive properties of $TlInSe_2$ crystals is studied. It has been established that in the case of uniaxial compression along the [001] direction, the dark and photoconductivity of the samples increase, and on the contrary, decrease in case of stretching. Moreover, the maxima of the spectral photosensitivity due to direct optical transitions at the point G at K = 0, do not change. The long-wavelength limit of intrinsic photoconductivity decreases with compression and increases with stretching which indicates the multi-valley nature of the band electronic spectrum of $TlInSe_2$ crystals.*

Conclusions. *Under uniaxial compression of $TlInSe_2$ crystals in the [001] direction, their band gap increases. As a result, the Fermi level shifts and the concentration of electrons at r-centers increases, which leads to an expansion of the linear sections of the lux - ampere characteristic.*

It was found that the absolute value of the photocurrent in positively deformed samples increases, and in negatively deformed ones it decreases in comparison with the un deformed *sample. In this case, the maximum of the photocurrent does not change, which indicates that the width of direct transitions in $TlInSe_2$ crystals remains unchanged at different types of deformation.*

Keywords: *elastic deformation, electrical characteristics, photoelectric characteristics, strain resistance properties, compression, tension, multi-valley, band electronic spectrum.*

Introduction. In recent years the interest from researchers and practitioners to various semiconductor converters including strain gauges has sharply increased, the main advantages of which are high sensitivity and small size. The requirements of modern science and technology are steadily growing which necessitates the search for materials with various properties that meet these requirements, since in most cases new compounds exhibit special qualities and thereby contribute to the solution of important technical problems that arise. Consequently, at present, along with the improvement of the properties of existing materials, the search for new semiconductor materials, including ternary and more complex compounds, the study of their various characteristics is one of the key tasks of semiconductor physics. However, the development and application of semiconductor converters, which include strain gauges, are impossible without a clear understanding of the features of the behavior of semiconductor crystals under certain specific conditions.

$TlInSe_2$ crystals - structural analogs of $A^{III}B^{IV}$ crystals [1-3] - have shown themselves to be promising for tensometry. The interest in these compounds from a scientific point of view is due to the specific structure of its crystal lattice. The unit cell of these crystals contains two independent structural units, which provide different coordination, valence state and character of chemical bonds for the constituent opposite cations of the same group. These features are the reason for the sharp anisotropy of the physical properties of this type of semiconductor.

In $TlInSe_2$ crystals, a strong piezoresistive effect was found in the direction of the crystallographic axis [001], which was explained on the basis of a four-ellipsoidal model of the band structure, with the most probable locations of the extrema at points G, N, and T of the Brillouin zone [4, 5].

In [6 - 9], using the pseudo-potential method, the band structures of crystals of the $TlInSe_2$ type were theoretically calculated, as well as the effect of spin-orbit interaction, pressure and temperature on them. It was shown [9, 10] that under hydrostatic compression, as well as under uniaxial tension along the tetragonal axis [001] of crystals of the $TlInSe_2$ type, as well as under compression perpendicular to this axis, their forbidden bands narrow, and under uniaxial compression along [001] the indirect gap corresponding to the $T_3 \rightarrow D$ transition will increase.

It was predicted in [4] that under uniaxial compression (tension) the energy gap corresponding to the direct transition (K = 0 at the point G) will remain unchanged, and the indirect transition gap increases upon compression of the crystal along the [001] crystallographic axis, which contradicts to the conclusions of [6-9] that when the crystal is compressed in the [001] direction, the indirect energy gap of the $TlInSe_2$ crystal narrows.

To determine the nature of the shift of the band gap of the p-$TlInSe_2$ crystal, as well as the details of the band structure of crystals under uniaxial deformation, the electro physical and photoelectric properties (specific electrical conductivity, spectral distribution of photoconductivity and the lux-ampere characteristic) of specimens under compression and tension along the [001] direction to various degrees of deformation.

Although the results of the above works testify in favor of the multi-valley character of the electronic structure of $TlInSe_2$ crystals, however, there is still no complete clarity in this issue. There are conflicting results and interpretations of the results obtained. Therefore, in this work, to test the multi-valley mechanism of the tensoresistive effect in p-$TlInSe_2$ crystals, we investigated the spectral distributions of the photocurrent I_s and the tensosensitivity K along the [001] direction in a wide spectral region of the incident light in under formed, positively, and negatively deformed states.

Samples for research and experimental technique. Crystals synthesized by fusing the components in accordance with stoichiometry in evacuated (~ 10-4 mm Hg) and sealed quartz ampoules were used for the study. Highly pure elements thallium (Tl - 000), indium (In - 000) and selenium (Se - high purity grade - 17 - 4) were used as initial components for the synthesis. Single crystals were grown by the improved Bridgman method, the crystallization front velocity varied from 0.5 to 0.9 mm / h.

The samples required for the study were prepared by cleaving the grown single crystals from an ingot by the simplest indentation with a sharp knife, the blade thickness of which is ≤ 0.01 mm, along two mutually perpendicular planes of natural cleavage. The samples were in the form of rectangular parallelepipeds with dimensions of 10x10x0.25 mm. Thus obtained "needle" crystals, without any additional processing were ready for welding contacts.

Two methods were used to solder mechanically reliable ohmic contacts on these blanks:

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Plates of steel 45 with a thickness of 0.5 - 1.0 mm and a length of 20 - 80 mm served as calibration beams for the glued sensors. The surface of the substrate according to the processing class was at least 7.

These substrates were treated in toluene before the sublayer was applied for the purpose of degreasing and then washed in ethyl alcohol. On the substrates cleaned in this way (on the required surface area), an underlayer of epoxy - cresol varnish (EP - 96) was applied with a brush, which is a solution of E - 40 epoxy resin modified with moninic acid, with the addition of buceonolized «RB» resole and K – 421 – 02. The sublayer thickness corresponded to 10 - 15 μk. In the process of applying the subcoat, a uniform coating thickness was provided.

After holding for an hour at room temperature, the substrate was transferred to an oven for high-temperature polymerization. A slow increase in temperature to 453 K and holding the substrates at this temperature for 1 hour ensured complete polymerization and excluded the appearance of air bubbles. On the substrate prepared in this way, on top of the sublayer, a second layer of varnish was applied, slightly exceeding the dimensions of the strain gauge.

$TlInSe_2$ samples with welded leads were placed on a varnish layer and slightly pressed down. In this case, the surface of the samples is completely varnished. Simultaneously, the samples were assigned the required positions in the plane of the substrate. For tight contact of the sensor body, as well as to maintain the specified orientation of the sensor relative to the substrate, the device was covered with a thin fluoroplastic tape 1.5 mm wide. The device was dried at a temperature of 291 - 296 K for 1 hour, followed by annealing at 463 ± 3 K for about 1.5 hours.

After drying, if necessary, the fluoroplastic tape was easily separated from the finished sensor. The specified drying mode turned out to be the most optimal, and the devices showed the maximum sensitivity.

Research results and their discussion. Under uniaxial elastic deformation of $TlInSe_2$ crystals, the value of the dark conductivity in the [001] $\sigma_T^{[001]}$ direction changes. Under compression (positive deformation) of the samples, the value of dark conductivity increases, and under tension (negatively deformed state), it decreases (Fig. 1). As noted in [10], during the deformation of the samples, the values of photoconductivity change strongly, but the position of the spectral maximum of intrinsic photoconductivity does not change, but the long-wavelength boundary of intrinsic photoconductivity is significantly displaced.

The observed experimental results on the shift of the long-wavelength boundary of intrinsic photoconductivity can be explained on the basis of the presence in the band structure of crystals of direct (corresponding to the transition at K = 0 at the point G) and indirect (corresponding to transitions at K≠0) optical transitions. Such a band structure is shown schematically in Fig. 2.

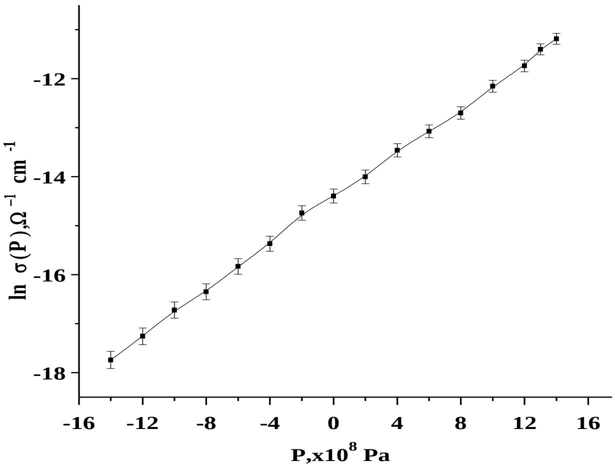

Fig. 1. Change in electrical conductivity from hydrostatic pressure

Transition 1 in the diagram corresponds to the case where the wave vector $\vec{K}$ is conserved during the interaction of an electron with photons, i.e. the transition of an electron from the valence band of E_v to the conduction band of E_c occurs $\Delta\vec{K}$ = 0 (in the diagram of the dependence, $E(\vec{K})$ the maximum of the valence band and the minimum of the conduction band are on the same vertical), such transitions are called direct. If during the interaction of electrons with photons the wave vector of the electron is not conserved (Fig. 2, transitions 2, 3, 4), i.e. the transition of an electron from the valence band to the conduction band occurs $\Delta\vec{K}$ =0 (in the $E(\vec{K})$ diagram, the minimum of the conduction band and the maximum of the valence band lie at different values $\vec{K}$), then such transitions are called indirect.

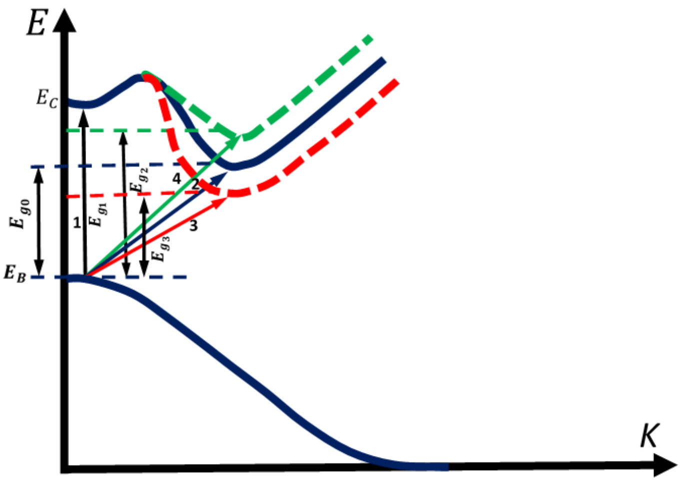

Fig. 2. Diagram of direct (1) and indirect (2, 3, 4) optical inter band transitions for $TlInSe_2$ crystals: 2 - in an under formed sample, 3 - under compression deformation, and 4 - under tensile deformation

The invariance of the maximum in the spectral dependence of the photoconductivity of the crystal under compression and tension deformations indicates that the energy of direct optical transitions remains unchanged during deformation. The shift of the long-wavelength boundary of intrinsic photoconductivity indicates a change in the energy of indirect optical transitions during deformation of the semiconductor. Under uniaxial compression of the p-$TlInSe_2$ crystal in the direction of the [001] crystallographic axis, the energy of the indirect optical transition decreases (transition 3 in Fig. 2), and when the crystal is stretched in this direction, on the contrary, the energy of the indirect optical transition increases (transition 4 in Fig.2). The dependence of the change in the width of the indirect transition gap E.g. on the degree of deformation is given in Table 1. These experimental results are in good agreement with the conclusions of the band model proposed in [4], which testifies to the multi valley character of the band electron spectrum of crystals of the p-$TlInSe_2$ type.

It should be noted that the values of the barometric coefficients of the displacement of the photoconductivity boundary determined from the observed change in the electrical conductivity of these crystals under hydrostatic - all-round - compression ($G = -1.75 \cdot 10^{-10}$ eV / Pa) and from the shift of the long-wavelength boundary of the intrinsic photoconductivity spectra under unilateral elastic compression along the crystallographic axis [001], which are close, they indicate that the displacement of the zone boundary under both uniaxial elastic compression and hydrostatic compression is mainly the result of crystal compression along the [001] direction.

Table 1

Shift of the long-wavelength boundary of intrinsic photoconductivity of an undoped $TlInSe_2$ crystal under uniaxial compression along [001]

Pressure P, Pa	Long-wavelength boundary of intrinsic photoconductivity $E_{\Gamma}(P)$, eV	Barometric displacement coefficient $G^{(001)} = \frac{\partial E_{\Gamma}(P)}{\partial P}$, eV/Pa
0	0,976±0,005	
$1{,}64 \cdot 10^7$	0,970±0,005	$-3{,}46 \cdot 10^{-10}$
$3{,}28 \cdot 10^7$	0,963±0,005	$-3{,}90 \cdot 10^{-10}$
$4{,}92 \cdot 10^7$	0,958±0,005	$-3{,}7 \cdot 10^{-10}$
$6{,}56 \cdot 10^7$	0,953±0,005	$-3{,}5 \cdot 10^{-10}$
$8{,}2 \cdot 10^7$	0,943±0,005	$-3{,}98 \cdot 10^{-10}$
$17{,}28 \cdot 10^7$	0,899±0,005	$-4{,}45 \cdot 10^{-10}$

To confirm the multivalley nature of the electronic bands, the spectral distributions of the photocurrent Is and the tensosensitivity coefficient K_{ε} of $TlInSe_2$ crystals along the [001] direction in the spectral region 0.8 - 3.2 eV were investigated for underformed, positively deformed, and negatively deformed states. The results of these studies showed that, as in the case of photoconductivity, the absolute value of the photocurrent in positively deformed samples increases, and in negatively deformed samples it decreases in comparison with an underformed sample. In this case, the maximum of the photocurrent does not change and is at a value of 1.20 eV, which confirms the results [10] on the invariability of the energy of direct optical transitions in $TlInSe_2$ crystals under various types of deformation (Fig. 3).

Fig. 3 also shows that, in contrast to the results of [10], under deformations, changes in the spectral distribution of the photocurrent are observed not only in the long-wavelength, but also in the short-wavelength region of the spectrum compared to the maximum of the photocurrent. This result indicates the presence of indirect band transitions in $TlInSe_2$ crystals not only in the low-energy, but also in the high-energy spectral region compared to the maximum of the photocurrent.

Figure 4 shows the spectral distributions of the photo stimulated tensosensitivity coefficient of deformed crystals exposed to light in the spectral range of 0.8 - 3.2 eV. In the distribution of the spectrum of a crystal positively deformed in the direction along the [001] direction, there is an intense peak with a maximum at 1.20 eV, and a comparatively less intense peak with a maximum in the region of 1.6 - 1.70 eV. The maximum of the intense peak in the distribution spectrum of the photo stimulated tensosensitivity corresponds to the maxima of the photoconductivity and photocurrent of the crystal (see Fig. 3 and Fig. 4, a). With negative crystal deformation, the value of the maximum of the intense peak of the photo stimulated tensosensitivity at 1.20 eV does not change, but its intensity slightly decreases and a new peak appears with a maximum at 2.10 eV (Fig.4, b).

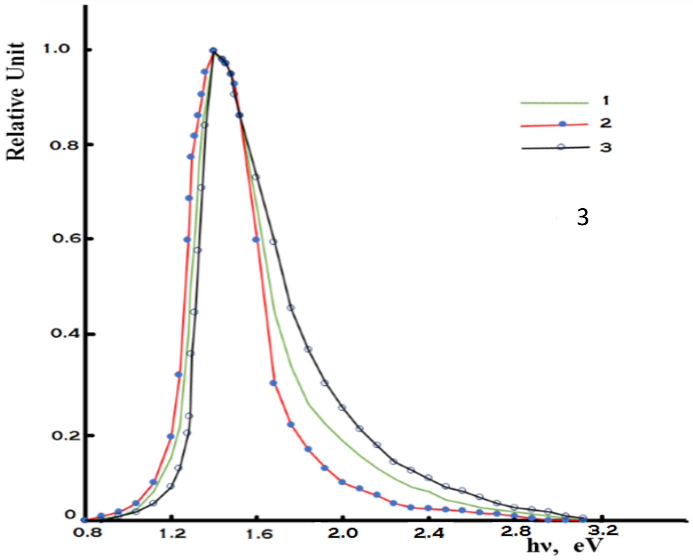

Fig. 3. Spectral distribution of the photocurrent of an underformed (1), positively deformed (2), and negatively deformed (3) p-$TlInSe_2$ sample

The values of the energy maxima and minima of the spectral distribution of the photo stimulated tensosensitivity of deformed $TlInSe_2$ samples, determined by us by illumination with light of different energies, strictly correspond to the values of the energies of inter band transitions given in [15], in which the spectra of X-ray photoelectrons were experimentally determined and their energy levels were theoretically calculated using the unrestricted Hartree - Fock method (Table 2).

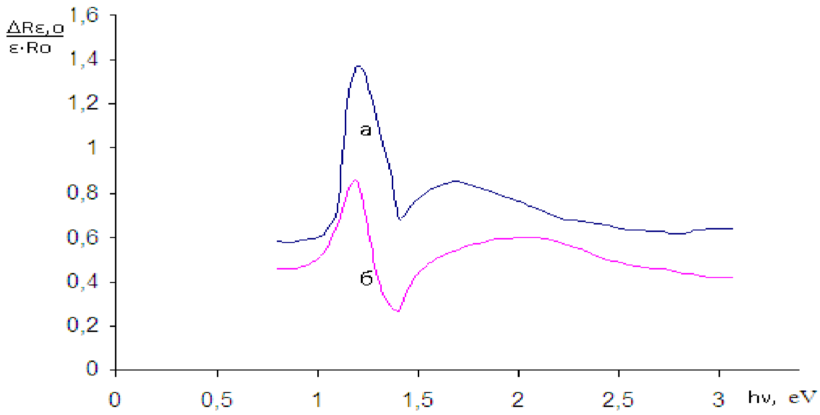

Fig. 4. Spectral distribution of tensosensitivity positively (a) and negatively (b) of the deformed p -$TlInSe_2$ crystal

Table 2

Theoretically calculated in the $TlIn_4Se_{16}$ cluster and experimentally determined in $TlInSe_2$ crystals band-to-band electronic transitions

Theoretically calculated in the $TlIn_4Se_{16}$ cluster, eV [by 15]	-	1,2	1,4	1,8	2,1	2,3	2,5	2,9
Experimentally determined in a crystal $TlInSe_2$, eV	1,08	1,2	1,43	1,7	2,13	2,3	2,5	2,8

If we take into account that the maximum of the photo stimulated tensosensitivity at 1.20 eV is due to direct interband transitions, then it can be argued that all the other maxima of the photo stimulated tensosensitivity observed in experiments in $TlInSe_2$ crystals are associated with indirect interband electronic transitions caused from different points of the Brillouin zone.

These changes are more clearly manifested in the difference between the normalized spectral distributions of the coefficients of the photo stimulated tensosensitivity of positively and negatively deformed crystals (Fig. 5, curve 3). It can be seen from the figure that at negative crystal deformation (compression deformation), the fraction of electronic transitions with an energy of 1.4 eV is dominant, but the fraction of tensosensitivity in the region of all other transitions is significantly reduced. In contrast, with positive crystal deformation (tensile strain), the fraction of transitions with an energy of 1.4 eV is the smallest, and the fraction of the tensosensitivity for all other transitions noticeably increases. Based

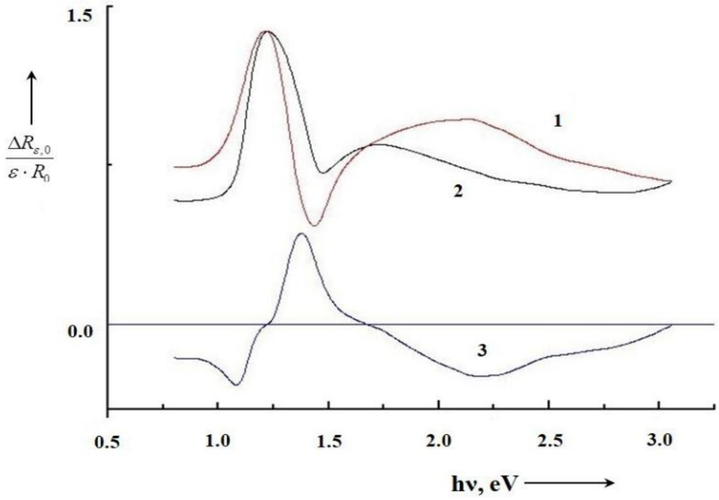

Fig. 5. Normalized spectral distributions of the photo stimulated tensosensitivity of negatively deformed (1), positively deformed (2) $TlInSe_2$ crystals and the difference 2 and 1 (3).

Therefore, we concluded that the tensosensitivity mechanism of $TlInSe_2$ crystals is indeed due to the mechanism of charge flow from one valley to another, due to a change in the energy positions of the valleys upon deformation.

The above results, as well as the results obtained in [13, 14], show that semiconductor converters can be created from $TlInSe_2$ single crystals, in which the resistance changes depending on the applied (mechanical) voltage, is substantially linear over the entire voltage range, and strongly depends on temperature. Although an amplifier is required with $TlInSe_2$ strain gauges, the linearity is very high and the temperature effect can be easily compensated. In addition, these strain gauges have certain advantages:

- have small dimensions and weight;
- are low-inertia, which makes it possible to use strain gauges both for static and dynamic measurements;
- have linear characteristics for all physical parameters;
- allows for remote measurement and at many points, by the method of multi-point strain gauging;
- the method of installing them on the investigated part does not require complex devices and does not distort the deformation field of the investigated part.

Conclusion. It was found that the tensosensitivity of $TlInSe_2$ crystals is due to the multi-valley mechanism, i.e. overflow of charges from one valley to another, due to changes in the energy positions of the valleys during deformation. The presence of a strong piezo - photoresistive effect in $TlInSe_2$ crystals makes it possible to create on their basis highly sensitive sensors of displacement, force, pressure, acceleration, and torque sensors. Such sensors make it possible to create powerful low-resistance strain gauges with a small (miniature) surface of connection with the sample, which reduces leakage currents at high temperatures and gives a higher isolation voltage between the sensitive crystal and the base (substrate). It is possible to significantly increase the sensitivity of $TlInSe_2$-based sensors to measured values using heating and optical illumination.

It is shown that under compression and tension deformations in the [001] direction, a change in the spectral distribution of the photocurrent is observed not only in the long-wavelength, but also in the short-wavelength region of the spectrum as compared with the maximum of the photocurrent. This result indicates that in $TlInSe_2$ crystals there are indirect band transitions not only in the low-energy, as compared to the direct band transition, but also in the high-energy region of the spectrum. It was found that the band gap increases under uniaxial compression, and decreases under tension.

At positive and negative deformations, the maxima in the spectral distribution of the tensosensitivity coefficient of $TlInSe_2$ crystals caused by electronic transitions from different valleys are redistributed, which indicates that the mechanism of the tensosensitivity of $TlInSe_2$ crystals is caused by the flow of charges from one valley to another due to changes in the energy positions of the valleys upon deformation.

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