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DEFORMATION MODULATION OF ELECTRONIC STATES IN LAYERED CRYSTALS

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DEFORMATION MODULATION OF ELECTRONIC STATES IN LAYERED CRYSTALS

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Abstract. The paper analyzes the changes in electronic states of a layered crystal caused by shear deformation of layers under action of the force consisting of both constant and harmonic components. It is shown that such deformation can change the component of lattice spacing by the magnitude of one layer thickness. In the frame of adiabatic approximation of the perturbation theory it is calculated the electronic states, whose degeneracy is removed by deformation perturbation, as well as the time moments of realization of degeneracy removal.

Keywords: layered crystals, shear deformation, modulation of electronic states

ДЕФОРМАЦІЙНЕ МОДУЛЮВАННЯ ЕЛЕКТРОННИХ СТАНІВ У ШАРУВАТИХ КРИСТАЛАХ

Й. М. Стахіра

Анотація. Аналізуються зміни структури електронних станів шаруватого кристала, зумовлені деформацією зсуву шарів, яка здійснюється сумою сталої і гармонічної складових сили. Показано, що така деформація кристала може змінювати складову періоду ґратки на товщину

шару. В адиабатичному наближенні теорії збурень встановлені енергетичні стани, виродження яких знімається деформаційним збуренням, і моменти часу його реалізації.

Ключові слова: шаруваті кристали, деформація зсуву, модуляція електронних станів

ДЕФОРМАЦИОННОЕ МОДУЛИРОВАНИЕ ЭЛЕКТРОННЫХ СОСТОЯНИЙ В СЛОИСТЫХ КРИСТАЛЛАХ

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Аннотация. Анализируются изменения структуры электронных состояний слоистого кристалла, обусловленные деформацией сдвига слоев, которая совершается сумой постоянной и гармонической составляющих силы. Показано, что такая деформация кристалла может изменять составляющую периода решетки на толщину слоя. В адиабатическом приближении теории возмущений установлены энергетические состояния, вырождения которых снимается деформационным возмущением, и моменты времени его реализации.

Ключевые слова: слоистые кристаллы, деформация сдвига, модуляция электронных состояний

The presence of layers in the crystal structure of layered of crystals as their structural elements causes certain features of electronic properties of these materials. They are clearly displayed in the electron transfer phenomena and the structure of interband and phonon absorption spectra. The role of layers, as structural elements bringing forth the adequate spatial dependence of a potential, in forming the peculiarities of electronic properties depends on the structure of the layers and interaction between them. A separate layer in these crystals can have complex multilevel structure which is formed by the intense interatomic interaction [1, 2] which leads to a certain isolation of the layer. This property is manifested in the residual interaction between the adjacent layers and conservation of their structure in a wide range of external influences. It is clearly displayed during the actions of external deformation forces on the layered crystals [3-6]. The characteristic changes in the set of electronic states of layered crystals, arising from deformation of these materials, are resulted from the changes in the interaction between the layers due to approach and relative shift of the layers. This work is devoted to analysis of the changes, caused by deformation, in the structure of electronic layers and their dynamics which are realized in the form of relative shift of the adjacent layers. It is a continuation of papers [7, 8] where a mechanism has been proposed for the formation of spectra of the photoconductivity

component which is proportional to the product of light intensity and mechanical stress.

Unlike the deformation of common solids, the deformation of a layered crystal can be realized by significant approaching and relative shear the layers without essential changes in their structure. Due to such deformation the lattice constant and interaction between the adjacent layers undergo changes what results in adequate changes in the structure of electronic states. For estimation of these changes we consider the structural deformation for the case of a simplified model of layered crystal whose lattice parameter in the xy plane of the layer is equal to d_x i d_y , and in the direction z normal to the layer plane – d_z^0 which coincides with the thickness of the layer. The crystal is exposed to the deformation force which is a sum of the constant F_c and variable harmonic $F_s \cdot \sin \Omega t$ components and which leads to a relative displacement between the adjacent layers by the appropriate distance $l = l_c + l_s \cdot \sin \Omega t$ without substantial changes in the structure of layers and distance between them. Deformation of the crystal by this force does not change the periodicity of the lattice in the xy plane but leads to changing the lattice period in the direction normal to its planes.

Within the model under consideration the lattice constant in the direction normal to the layer plane is determined by the number n of shifted

layers per total distance $n \cdot l$ which is equal to the least number of period d_{xy} . (d_{xy} is the lattice constant in the layer plane where the shift occurs). Accordingly, the number n is determined by the equality $N \cdot d_{xy} = n \cdot l$, where N is the smallest integer for which n is an integer at given values of l and d_{xy} . In the case of multiplicity of the period d_{xy} to the shift value, which is reached for the shifts $l \leq (d_{xy} / 2)$, it is realized the small values of n and correspondingly the short periods $d_z = n \cdot d_z^0$ of a deformed crystal.

Due to conservation of the layer structure during the process of shear deformation the lattice period d_z can be changed only discretely by the minimum thickness of one layer d_z^0 . This fact gives rise to complex non-harmonic dependence of period d_z on the shift which is changing continuously.

Dependence of the lattice parameter d_z of the deformed crystal on the shift depends substantially on the ratio of their quantities to the period d_{xy} . The main requirement of this relationship - the period d_{xy} should be multiple to the shift l . For a given shift direction in the layer plane the effective displacements are selected discrete values of shifts which results in the corresponding changes of the period d_z of a deformed crystal.

The value of period d_z of the crystal deformed by a shift can be changed to integer p using an additional shift. Changing the period d_z by p units of layer thicknesses is achieved by the increments

$$\Delta l_p^+ = -\frac{p \cdot l_c}{n + p}, \quad (1)$$

$$\Delta l_p^- = \frac{p \cdot l_c}{n - p}, \quad (2)$$

which gives rise to the increase of period (1) or its decrease (2).

This property leads to the change in number n and period d_z of a deformed crystal over the time by harmonic component of the deforming force that creates the required increments of shifts.

Time changes of the period d_z of deformation perturbation, $d_z = n \cdot d_z^0$, which directly coincide with the time dependence of the corresponding multiplicity n , are caused by the harmonic

component of shift. Relatively to the time start, $\Omega \cdot t_0 = 0$, the value of a given fixed displacement l_c , to which the multiplicity n corresponds, is changed by the harmonic component $l_s \cdot \sin \Omega t$ of the shift to the extreme values $l_c \pm l_s$. During the process of increasing the harmonic component of shift, it is realized such its values which are equal to the increment of shift Δl_p^- what decreases the multiplicity of n by p units. Accordingly, a decrease of the harmonic shift with time is accompanied by its values equal to the increment Δl_p^+ what increases the multiplicity by p .

Deformation of a layered crystal by the force, whose harmonic component has the period much longer than the time of elasticity relaxation of a crystal, results in creating the shifts between the layers which are harmonic functions of time being in-phase to the deformed force. If at the initial time $t = 0$ the shift between layers created by the constant component of force is equal to l_c , its increase, caused by growth of the force harmonic component, occurs within the harmonic phase range $0 - \pi$, whereas within the range $\pi - 2\pi$ a decrease takes place. The time moments, at which the change of period d_z , is realized, are determined by the following equations

$$-\frac{p \cdot l_c}{n - p} = l_s \cdot \sin \varphi_p^-, \quad (3)$$

$$\frac{p \cdot l_c}{n + p} = l_s \cdot \sin \varphi_p^+. \quad (4)$$

Variation of the deformation perturbation period, which is performed by means of sudden increments of its value by integer number of thickness, is realized only in the time moments when the shift takes the values with respect to which the period d_{xy} is multiple of their magnitude. Figure 1 shows the time changes of the deformation perturbation period for the specific case of shifts: $l_c = 0.25$, $l_s = 0.125$, $d_{xy} = 1$.

Discrete time change of the period of the deformation perturbations $d_z = n \cdot d_z^0$ this is caused by continuous in time the harmonic shift of layers, gives rise to certain time intervals within the period of harmonics, in which the period d_z remains constant over the time. The value of

period d_z , which is caused by the shift l_c remains unchanged in the time intervals where the value of harmonic shift is insufficient to change the multiplicity by unity.

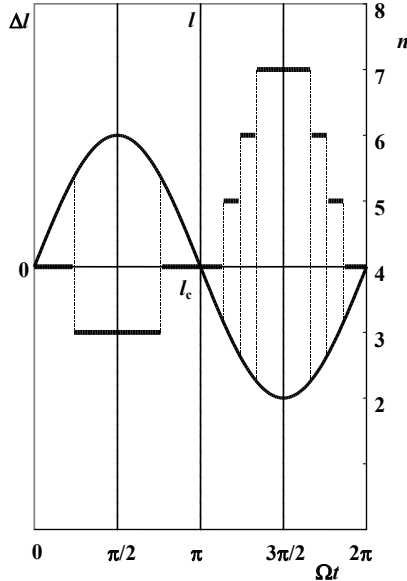


Fig.1. Schematic depiction of time dependence of the deformed crystal period.

In relation to the phase origin, $\varphi = 0$, it refers the intervals of phase $0 - \varphi_1$, $\pi - \varphi_1$, $\pi - \varphi_2$, $2\pi - \varphi_2$, for which

$$\varphi_1 = \arcsin\left(\frac{l_c}{l_s} \cdot \frac{1}{n-1}\right), \quad (5)$$

$$\varphi_2 = \arcsin\left(\frac{l_c}{l_s} \cdot \frac{1}{n+1}\right). \quad (6)$$

Thus, the time dependence of the multiplicity period d_{xy} to the value of total shift l , given at the initial fixed time, is realized by an additional shift whose magnitude is limited by the amplitude of the harmonic component shift. As a result of harmonic dependence of the total shift on time it is realized the time intervals during which the multiplicity n varies in opposite directions. At that the change of multiplicity is always opposite to the change of harmonic shift.

Deformation of a layered crystal leads to some changes in the crystal structure, due to which the appropriate changes of coordinate crystal dependence of the crystal field potential is realized. In the case of deformation, which results in a rela-

tive shift of the adjacent layers, there takes place a displacement by relatively large distances of the macroscopic fragments of crystal structure what can lead to significant changes in the electronic states. Such deformation of a layered crystal is usually achieved by small external forces and can be considered as a cause for appearance of the deformation perturbation of the potential.

Deformation potential perturbation is formed by the relative shift between the layers and significant changes in the interaction between them. The shift between the layers leads to changes of the distances between the identical structural elements of the adjacent layers and thus to change of the interaction between them and its dependence on the structure of the layer. The changes by relative shift of the weak interaction between the layers in the condition of conservation of their structure, which leave the deformed crystal in a layered state, gives rise to changes of the potential which are considered as a perturbation, leading to potential changes being considered as a perturbation. If a deforming force has low frequency harmonic component, which provides a quasi-stationary regime of the process, the period of such perturbation can depend on the shift between the adjacent layers and be of a complex discrete function of time.

The relative shift between the adjacent layers occurs simultaneously with the harmonic phase and changes of the perturbation period are realized only at the discrete values of the shift which arise in the corresponding time moments during the period of the harmonic. This time-dependent perturbation allows one to consider its impact on the electronic states in the adiabatic approximation of the perturbation theory. In the frame of this approximation the harmonic component of the perturbation is considered as slow one against which the slow (compared with the harmonic period) time intervals of changing the perturbation periodicity are fast components of the time dependence of the perturbation.

The deformation of a crystal by the total harmonic force leads to a shift of the layers which is realized in phase with the phase of harmonic component of the force. Synphasing to this harmonic of the time dependence of deformation perturbation period disappears due to the complex non-linear dependence of perturbation on the shift

magnitude. Changing the perturbation period by means of sudden increments of its value is realized only in the time moments at which the shifts acquire the magnitudes with respect to which the shift period d_{xy} is multiple to their magnitude.

In particular, the perturbation period created by constant component of the force $d_{zc} = \frac{d_{xy}}{l_c} \cdot d_z^0 = n_c \cdot d_z^0$ decreases by the harmonic component due to increasing the total shift in the time intervals corresponding to the change of phase from $\Omega \cdot t_0 = 0$ to $\Omega \cdot t_1 = \pi$ and increases within the time intervals that correspond to the difference in the phase of harmonics from π to 2π . As regards to the time origin $t_0 = 0$, at which the shift is equal to l_c , changing the period of deformation perturbation occurs in the phase opposite to that of harmonic.

Thus, the shear deformation of layers gives rise to forming a set of short time Bragg's planes which coincide with the planes of layers, and their energy location and time of origin are determined by the value $q = \frac{2\pi}{d_z}$. As a consequence of

the Bloch's wave diffraction on these planes there occur in the corresponding time moments the discontinuities in the energy dependence of electronic states $E(\mathbf{k})$ for all wave vectors \mathbf{k} , whose k_z component is $q = \frac{2\pi}{d_z}$. Commensurability q and k_z

components of wave vectors of the Brillouin zone allows realization of such gaps in the energy $E(\mathbf{k})$ in the whole range of energies of the conduction band. For a given, fixed in time, period of perturbation d_z , the discontinuities in energy dependences occur simultaneously at all energies, when the wave vectors \mathbf{k} have k_z component satisfying

the condition $k_z = \frac{2\pi}{d_z} \cdot m$ ($m = 1, 2, 3 \dots$). As a

result of the gaps in the energy dependences on the wave vector, $E(\mathbf{k})$, in the vicinity of these energies the density of states changes dramatically leading to the corresponding changes in the electronic properties of the crystal. The energy bands, which appear due to the Bragg's diffraction, are forming in the time moments that correspond in a one-to-one manner to the values of q and can be identified by its magnitude.

The processes of removing the degeneracy of energies $E(\mathbf{k})$ by deformation perturbation give rise to appropriate changes in the structure of electron states of a deformed crystal. Due to changes in the coordinate dependence of potential and its symmetry at the expense of layer shift the degeneracy is removed for the energies which depend on wave vectors and have the k_z component equal to the perturbation period q_i . Removing degeneracy of the energy states leads to the gaps in the energy dependence on wave vector for those vectors that have k_z component equal to q_i , and appropriate changes of electronic properties. Dynamics of the degeneracy removal and its impact on the structure of electronic states entirely coincides with the results of Bragg's diffraction of the Bloch's waves.

The shear deformation of layers maintains a layered structure and basic physical parameters of the crystal except for changing the component of lattice period in the direction perpendicular to the plane of layers which occurs discretely by the thickness of single layer. This model allows us to compare the changes in the structure of electron states that occur in the crystals, whose d_z components of the lattice constant differ by integer number. Under condition of constancy of the deformed crystal volume such deformation leads to the multiple changes in the volume of unit cells of the reciprocal lattice and the corresponding changes in the number of allowable values of wave vectors. Each fixed period of a deformed crystal corresponds to the exclusive spatial distribution of structural units and the appropriate interaction between them, depending on the structure of the layers. Among the different kinds of changes in the structure of electronic states induced by layer shift the gap of the energy functional dependence on wave vector and time of its realization remains the same for the considered model of a deformed crystal.

Structural changes in the energy of electronic states due to layer shift by controlled deforming forces can be experimentally established by observing the time dependence for one period of harmonic absorption coefficient of the monochromatic light, which acts as a monitor. In particular, this opens an opportunity to estimate the spectral composition of illuminating light on the basis of the time dependence of its absorption measured

by a radio-technical method without using the dispersion elements. In [7, 8] such changes have been estimated upon the time spectral dependence of the piezophotocurrent phase shift with respect to the phase of a deforming force.

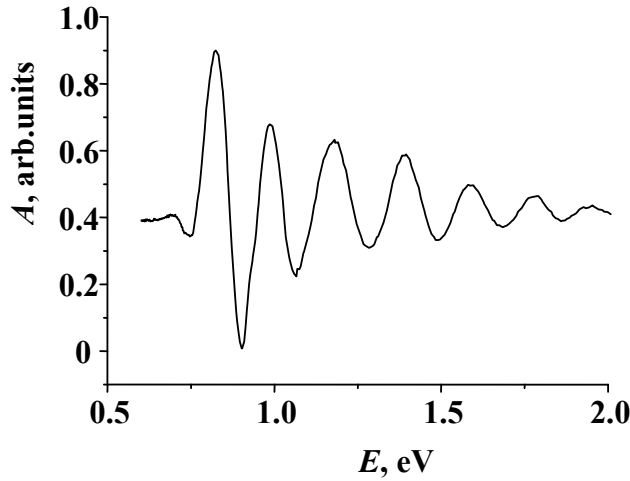


Fig.2. Spectrum of piezophotocurrentivity of In_4Se_3 crystal.

From the view point of practical application of the presented results of special attention is a spectral distribution of time moments of removing the degeneracy of energy of electronic states. With the aim of estimation of practical use of this effect it has been carried out experimental investigation of the dependence of the time moments of removing the degeneracy on the energy of electronic states. The experiments have been performed on crystals of the In_4Se_3 semiconductor compound. In_4Se_3 crystal belongs to the rhombic system in which weakly interacting layers have a complex structure. Unit cell of the crystal contains 28 atoms between which there are bonds of different types. In_4Se_3 single crystals were grown by the Czochralski method, and the samples under study presented natural cleaved plates. Deformation of the samples was carried out by the force which had a low frequency harmonic component. The force was applied perpendicularly to the plane of the layers at a small angle to their surface. We measured the spectra of a piezophotocurrentivity component of conductivity which is proportional to the product of light intensity and mechanical stress. The spectrum of this component in the range of photon energy which corresponds to the interband transitions in the In_4Se_3 compound is

shown in Fig. 2. Fig. 3 presents the spectral dependence of the phase shift of the piezophotocurrentivity component relatively to the phase of deforming force measured by a phase meter. It should be noted that there are not physical causes for occurrence of the phase differences in the process under consideration. An imaginary phase shift results from the sudden change in the moments of removing the degeneracy of the form of harmonics which occurs at the corresponding time moments.

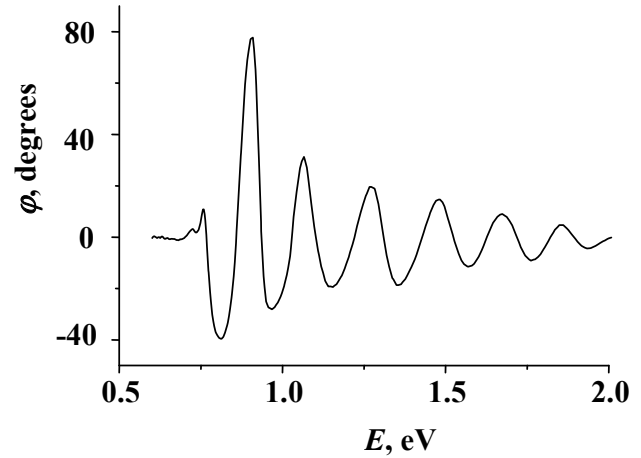


Fig.3. Spectrum of the phase shift of the piezophotocurrentivity component relatively to the phase of deforming force of In_4Se_3 crystal.

High sensitivity of the phase values to the energy of electronic states, $d\phi/dE(k)$, allows us to conclude that the deformation changes in the structure of electronic states of layered crystals, which are caused by a shift of the layers, can be effectively used for development of electronic analyzers of spectral composition of light and systems of object recognition.

Summing up the obtained results we come to conclusion that shear deformation by means of the deforming force with a harmonic component results in removing the degeneracy of energies of the electronic states whose wave vectors have the

component equal to the reverse period, $q = \frac{2\pi}{d_z}$.

Removal of the degeneracy occurs under action of the potential deformation perturbation subsequently in time during the period of the harmonic in the time moments for which the equality

$$k_z = q = \frac{2\pi}{d_z^0} \cdot \frac{l_c + l_s \cdot \sin \Omega t_p}{d_{xy}}, \quad (7)$$

is satisfied.

One-to-one correspondence of the time moments t_p to the energies, whose degeneracy are removed, allows one to identify the energies of electronics states upon the time moments of the degeneracy removal. One of the experimental methods of measuring the time moments of removing the degeneracy of the given energy state is study of the time dependence of the monochromatic light absorption. A sudden change in the coefficient of interband absorption of the monochromatic light of the known frequency, which arises due to a sudden change of the interband electronic state density, can be registered by using the common radio-technical methods. Time changes of the absorption coefficient can be controlled by the direct measurements as well as by changing the physical quantities which depend on this absorption.

Experimentally the changes of electronic states structure, caused by the shearing deformation of layers, can be elucidated via time dependency of the monochromatic radiation absorbance monitored during the harmonic period. In [7.8] such dependencies were estimated by spectral variation of shift of the piezophotoconductivity phase with respect to the phase of deformation force. Below we present the experimental results the component of the piezophotoconductivity spectra of In_4Se_3 layered crystal, proportional to the product of a monochromatic light intensity and a value of mechanical stress. Fig.4 illustrates the spectral dependencies of synchronized and phase-shifted components of the piezophotoconductivity current of In_4Se_3 layered crystal. Fig.5 presents the spectra of piezophotoconductivity and phase shift of In_4Se_3 crystal.

The main results on the shearing investigation deformation allow us to draw the following conclusions. The shearing deformation consisting of both constant and harmonic components leads to the changes in constituent of lattice parameter d_z^0 of a layered crystal. The value d_z is varied proportionally to the number of layers as, where n is an integer number determined by the equation $d_{xy} = l \cdot n$. Minimal values of d_z correspond to such values of shifts l , being divisible to d_{xy} .

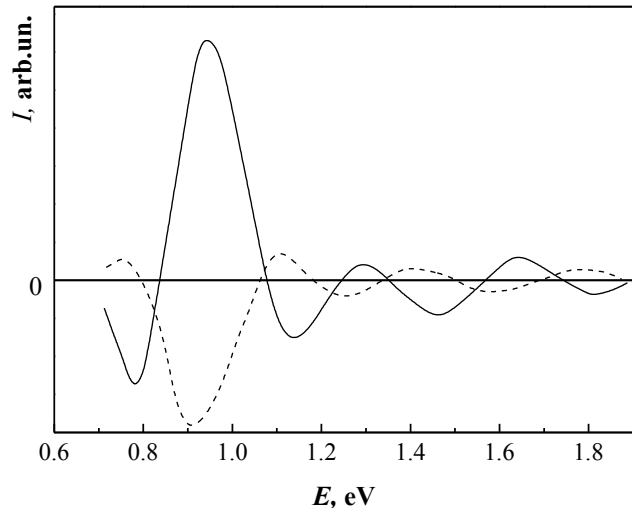


Fig.4. Spectral dependencies of in-phase (solid line) and phase-shifted (dash line) components of piezo-photoconductivity current of In_4Se_3 crystal.

The variation of shifts l during the harmonics period leads to an appearance of a certain number of new values of d_z , which are realized in the corresponding time moments t_p . During the shearing deformation the value d_z changes discretely at the specific time moments. At that, the time dependency of the deformation perturbed potential is a sum of the slow component, dependent on frequency shift, and the short time component caused by a discrete shift. Those perturbations result in partial removing degeneracy of the electronic states whose k_z component of the wave vector is determined by (7). Degeneracy removal is realized at the time moments corresponding to that energy.

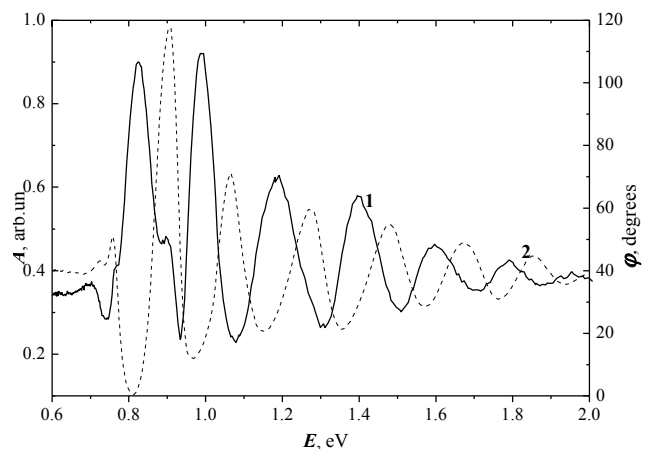


Fig.5. Spectra of piezophotoconductivity and phase shift between current and deformation force of In_4Se_3 crystal (1 – amplitude, 2 – phase shift).

One-to-one correspondence of the energies of the electronic states energies to the time moments of their degeneration removal allows determining the energy values by means of radio-technical measurements of the corresponding time moments. Such dependencies can be a basis for development of radio- technical analyzers of spectral composition of light without using dispersion elements [9].

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Summary

The paper substantiates the intended creation of the discrete periods of crystalline lattice of layered crystals due the relative displacement of layers. The shear deformation of the layers retains unchanged the layered structure and basic physical parameters of the crystal, except for the change of the component of the period of crystalline lattice in the direction normal to the plane of the layers, which occurs discretely by the thickness of one layer. Deformation of a layered crystal gives rise to certain changes in the crystalline structure due which the corresponding changes in the coordinate dependence of the crystalline field potential are realized. In the case of deformation, which leads to the relative displacement of adjacent layers, the displacement takes place at relatively large distances of the macroscopic fragments of the crystalline structure what can lead to significant changes in the electronic states. It is shown that the main changes caused by the shift of the layers in the structure of electronic states are due to the processes of degeneration of the energies of these states. The dynamics of the degeneration of electronic states is established what makes it possible to identify them upon the time moments of removing the degeneracy. It is proved that structural changes of the energies of electronic states due to the shift of the layers can be experimentally established upon the time dependence during the harmonic period of the absorption coefficient of the monochromatic light, which plays the role of a monitor. This, in particular, opens the possibility of evaluating the spectral composition of the illuminating light upon the time dependence of its absorption by means of radiotechnical methods without using dispersing elements.

Keywords: layered crystals, shear deformation, modulation of electronic states

ДЕФОРМАЦІЙНЕ МОДУЛЮВАННЯ ЕЛЕКТРОННИХ СТАНІВ У ШАРУВАТИХ КРИСТАЛАХ

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Реферат

Обґрунтовано планомірне створення дискретних періодів кристалічної ґратки шаруватих кристалів відносним зсувом шарів. Деформація зсуву шарів зберігає шарувату структуру і основні фізичні параметри кристала за винятком зміни компоненти періоду кристалічної ґратки у напрямку, нормальному до площини шарів, яка відбувається дискретно на товщину одного шару. Деформація шаруватого кристала призводить до певних змін кристалічної структури, внаслідок яких реалізуються відповідні зміни координатної залежності потенціалу кристалічного поля. У випадку деформації, яка призводить до відносного зсуву сусідніх шарів, відбувається переміщення на відносно значні відстані макроскопічних фрагментів кристалічної структури, що може привести до суттєвих змін електронних станів. Показано, що основні зміни, спричинені зсувом шарів, у структурі електронних станів зумовлені процесами зняття виродження енергій цих станів. Встановлено динаміку зняття виродження електронних станів, що дає змогу ідентифікувати їх за моментами часу зняття виродження. З'ясовано, що експериментально структурні зміни енергій електронних станів, які зумовлені зсувом шарів контрольованими деформуючими силами, можуть бути встановлені за часовою залежністю протягом періоду гармоніки коефіцієнта поглинання монохроматичного світла, яке відіграє роль монітора. Це, зокрема, відкриває можливість оцінки спектрального складу опромінюючого світла за часовою залежністю його поглинання радіотехнічним способом без використання диспергуючих елементів.

Ключові слова: шаруваті кристали, деформація зсуву, модуляція електронних станів