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Light flux effect on the electron gas dimension

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Abstract : The suggested spatial-energy model for electron spectrum of layered InSe semiconductor explains the magnetoresistance behaviour in the case of optical excitation, physical nature of rapid and slow photoconduction relaxations and absence of two-dimensional excitons (not surface).

Keywords Layered semiconductor, magnetoresistance, photoconduction, two-dimensional gas, exciton.

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1. Spatial-energy model

The specific crystal structure of the layered semiconductor InSe is due to the difference of the character of the chemical bond in different crystallographic direction-ion-covalent bond within a layer and Van-der-Waals bond between layers. A layer consists of atom sublayers in succession Se-In-In-Se which are placed along *C*-axis and perpendicular to sheet plan.

In energy scheme (Figure 1), weak Van-der-Waals bond between the packets Se-In-In-Se may be described by the system of potential barriers between the packets. There are 3 packets in the unit cell of γ-polytype InSe and barrier heights within the cell can be different. Since, the widths of the wells are rather low ($C = 2.496$ nm (Madelung 1983)) and the potential barriers are high enough, the carrier spectrum is two-dimensional and size quantized. The electron spectrum consists of E_i levels placed in the potential well which are limited by potential barrier.

Two-dimensional (2D) character of conductivity in InSe in the temperature range T = 4.3-20 K (helium temperatures) manifested in the peculiarities of magnetoresistance (MR) at $I \perp C$ (Brandt *et al* 1987). Due to these data, the Fermi surface is a cylinder with a generatrix parallel to *C* axis (Figure 2). That is why the angular dependence of oscillation period is determined only by the magnetic field component parallel to *C* axis. If the angle between the magnetic field vector and *C* axis increases one can observe the decrease of the amplitude of quantum oscillations and their complete disappearance at $\alpha_{max} \approx 70°$.

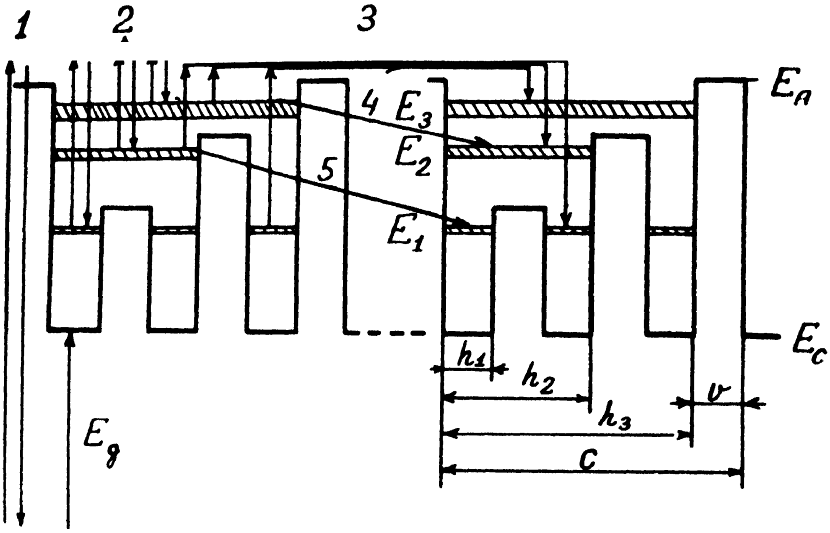

Figure 1. A diagram of electron transitions in InSe. E_c – a bottom of the conduction band; E_A – a value of the potential barrier towards C axis; E_i – energy of dimensionally quantized levels in potential holes in width h_i ; V – width of the Van-der-Waals slot ; c – a lattice parameter : 1, 2 – transitions of carriers to the excited state with the light turned off – corresponds to curve AB in Figure 3; 3 – types of transitions when the temperature of sample is higher than 50 K ; 4, 5 – tunneling through the potential barrier when equilibrium state is established – corresponds to curve BC in Figure 3.

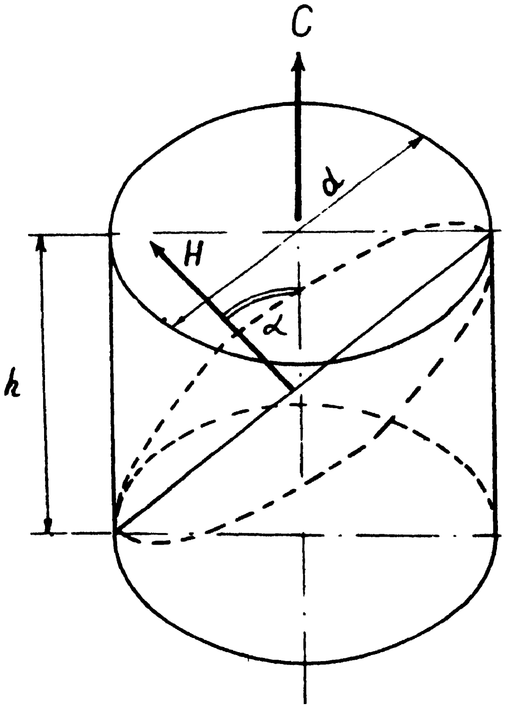

Figure 2. Cylindrical Fermi surface with diameter d limited by thickness h of sublayer Se-In-In-Se. α - angle of deviation of the vector of magnetic field H from the crystallographic axis C.

The height of the cylinder generatrix of the Fermi surface in the coordinate space is limited by the layer thickness. At the electron orbit comes in to contact with the layer boundary resulting in scattering and thus disturbing the electron precession and resulting in disappearance of quantum oscillations of MR. The quantum oscillation·frequency F is determined by the value of the Fermi surface section by the plane normal to the magnetic field vector (Figure 2). Since in coordinate space the Fermi cylindrical surface diameter d at $H \parallel C$ equals

$$d = [\, \pi\,(0.956\times10^{8}F\,)^{-1}]^{1/2} \tag{1}$$

it is possible to calculate thickness of the elementary layer

$$h = d\,\mathrm{ctg}\,\alpha_{\max} \tag{2}$$

In our case (Dmitriev *et al* 1989) we have observed two frequencies of MR oscillations with $F = 4.1\text{-}5.1\ T$ and $F = 10.9\ T$ which corresponds to $h \approx 2.04$ nm and $h = 1.28$ nm. An analysis of the cyclotron resonance data (Kress-Rogers *et al* 1983) shows that at low concentrations of current carriers at $T = 4.3$ K we have $h \approx 0.46$ nm. If we suppose that the packets Se-In-In-Se and Van-der-Waals gaps are of the same thickness, then the values of h are near to those calculated from the data of X-ray analysis with $C = 2.4946$ nm (Madelung 1983) as shown in Table 1.

Table 1. Values of h_i calculated from quantum phenomena and X-ray studies.

	Quantum phenomena (Brandt *et al* 1987, Dmitriev, *et al* 1989) h'_i nm	X-ray studies (Madelung 1983) h''_i nm	h''_i %
h_1	0.46	0,415	10,8
h_2	1,28	1,24	2,6
h_3	2,04	2,09	1,9

Due to the theory (Altshuler *et al* 1981), 2D character of electron gas must manifest itself in a form of negative magnetoresistance (NMR) at $I \perp C$, $H \perp C$. In the weak magnetic field we have $\delta\rho \sim \ln\,(H^{-1})$ and in strong one - $\delta\rho \sim \ln H$. In Figure 3 the dependences $\delta\rho$ ($\ln H$) at different temperatures is shown. There are two linear sections in weak and strong magnetic fields. This confirms experimentally the prediction of theory.

From Figure 1, it follows that electron gas is two-dimensional if its energy does not exceed the magnitude of potential barrier E_A. Otherwise there is no limitations to electron movement along C-axis and the electron gas will be three-dimensional. Our data on specific resistance and MR within temperature range 4.2-300 K are evidence of three dimension of electron gas in InSe above $T = 80$ K. Data of cyclotron resonance (Kress-Rogers 1983) indicate that three-dimensional properties of carriers becomes dominant at $T > 50$ K.

Continuous metal films or precipitates of Pb in the interlayer sections, do not form at low concentrations of Pb. The dependence $\delta\rho$ (T, H) do not differ from those for pure InSe

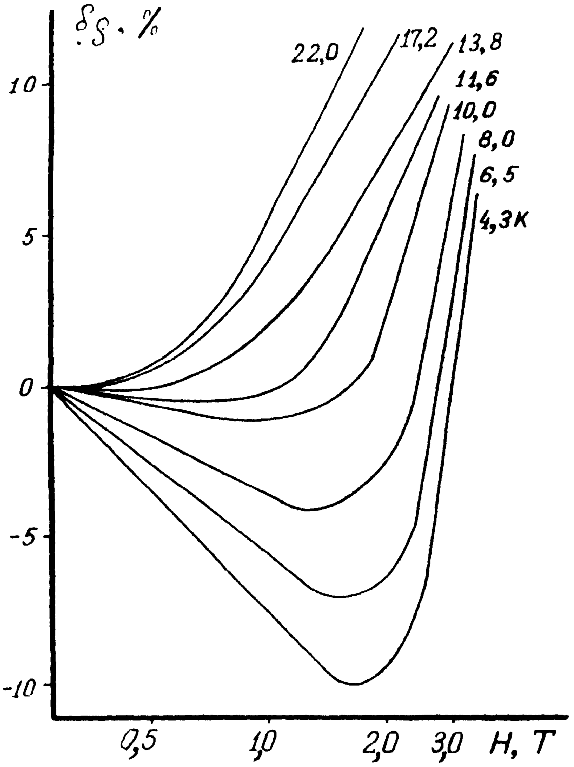

Figure 3. Magnetoresistance of InSe dependence on ln *H* at different temperatures, *H* ∥ *C*

(Figure 3). The increase of Pb content results in qualitative change of the mentioned dependences. If the temperature decreases below 7 K can observe change in $\partial\rho/\partial T$ sign from

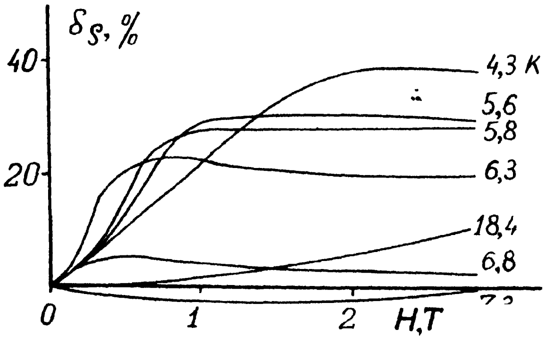

Figure 4. Magnetoresistance of InSe intercalated by Pb (~ 10^{19} cm^{-3}) at different temperatures, *H* ∥ *C*.

negative to positive (Dmitriev *et al* 1989) and transition from NMR to positive MR (Figure 4). An analogous change of MR sign is registered both for *H* ∥ *C* and *H* ⊥ *C*. Such a change may be explained supposing that precipitates of Pb passes at $T < 7$ K to superconducting state. The availability of such superconducting impregnations results in the shunting of a large number of interlayer barrier. This promotes transition to three-dimensional carrier transport and positive MR.

Microprobe analysis and electron microscopic studies (Dmitriev *et al* 1989) have shown that lead intercalate forms precipitates which do not exceed 1 μm in size in the layer plane. It is experimentally shown in (Dmitriev *et al* 1988, 1989), that a spectrum of current carriers in InSe is 2D at $T > 80$ K. Lead intercalation results in a three-dimensional spectrum at $T < 7.2$ K. This work shows that it is also possible to regulate gas dimensions by illumination.

2. Relaxation processes in two-dimensional quantized system

Illumination with energy $E = 1.95$ eV on the surface InSe layers (Brandt *et al* 1988) results in excitation of carries-transitions of type 1, 2, 3 in Figure 1. This excitation energy exceeds the value of potential barrier E_A, therefore gas of carriers by the illuminated surface becomes three-dimensional. MR is determined by the shunting effect of 3D gas of the sample surface and 2D gas of its volume, that defines the shape of curve 2 in Figure 5. While switching off light nonequilibrium carriers whose relaxation is impeded by potential barrier (transition of type 4.5) at low temperatures appear in the surface layers.

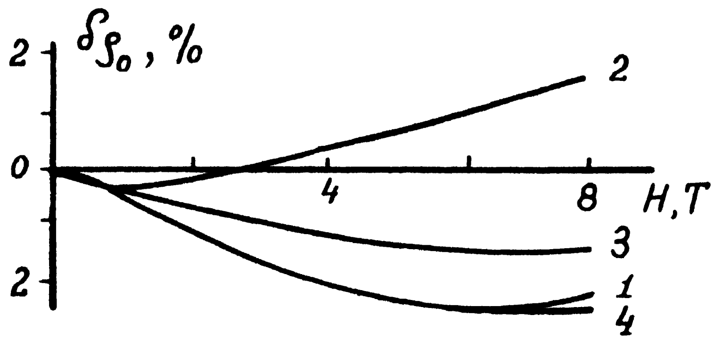

Figure 5. Magnetoresistance of the InSe sample at 4.2 K, $H \perp E$. 1 – prior to exposure to light ; 2 – during it ; 3 – after exposure to light ; 4 – after heating to > 50 K. Reproduced by data (Brandt *et al* 1988).

Potential barrier E_A impede the nonequilibrium state establishment, that promotes spatial distribution of nonequilibrium current carriers and determines residual photoconduction (curve 3 in Figure 5 and part of BC in Figure 6). Heating of the sample to $T > 50$ K increases electron energy to values sufficient to tunnel through barriers E_A, transition of type 3 in Figure 1. It favoures establishment of the initial equilibrium state (curve 4 in Figure 5).

The frequency of quantum MR oscillations is independent of illumination. It indicates that concentration of equilibrium carriers n_o in the volume of single crystal InSe in light remained practically unchanged : $n_0 >> \Delta n$.

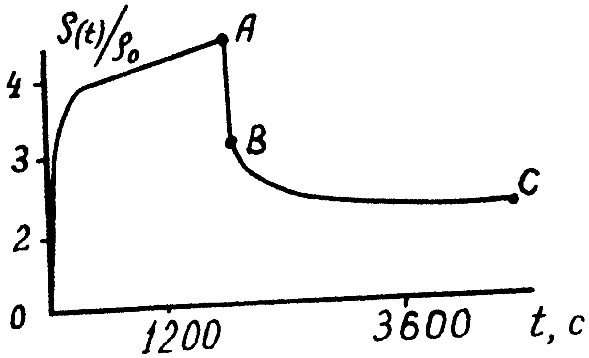

Figure 6. Time-dependent photoconduction at 4.2 K. Points: *O* – the light switched on ; *A* – the light switched off. Reproduced by data (Brandt *et al* 1989).

Relaxation processes are caused by existence of two groups of nonequilibrium carriers. The first group consists of electron with energy $E > E_A$, which are light generated near the sample surface. At first after cutting-off light, rapid relaxation takes place (curve AB on Figure 6), which is connected with transitions 1, 2, 3 Figure 1. Then the second group of nonequilibrium carriers with energies within $E_3 < E < E_A$ relaxes slowly which causes transitions 4 and 5 in Figure 1 (curve BC on Figure 6). Large relaxation times of the second group are connected with tunneling and flow through and over potential barriers. It causes residual photoconduction due to spatial separation of those carriers (Brandt *et al* 1988) by potential barriers (Dmitriev *et al* 1989).

3. A Problem of two-dimensional exciton

It should be noticed that 2D spectrum is between the limits of the conduction band bottom E_c and E_A, the potential barrier height. Beyond that band the spectrum is three-dimensional. For energies of electrons larger than E_A, it is evidently confirmed by the analysis of experimental data. Numerous unsuccessful efforts to reveal 2D excitons in layered semiconductors generally and particularly in InSe (Gnatenko and Zhirko 1987) are experimental proof of the three-dimensionality of spectrum at $E < E_c$.

4. Conclusions

Then the electron gas in the layered semiconductors InSe is two-dimensional only in the carriers energy band limited by the height of potential barrier E_A from E_c to $(E_c + E_A)$. Its dimensionality can be controlled both by intercalation with superconducting metal atoms or by temperature variation (Dmitriev *et al* 1988, 1989) and by illumination. The suggested spatial-energy model of the band spectrum completely explains photoconduction phenomenon in the layered InSe crystals (Brandt *et al* 1988) as well as accounts for the absence of experimental proof of the existence two-dimensional exciton states in InSe (Gnatenko and Zhirco 1987).

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