Quantum Hall effect in layered InSe crystals

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Quantization of the Hall resistance ρ_{xy} is observed in investigations of the galvanomagnetic properties of bulk crystals of InSe at temperatures $1.3\ \mathrm{K} \leqslant T \leqslant 4.2\ \mathrm{K}$ and in fields $H \leqslant 80$ kOe. The values of ρ_{xy} on the plateau were close to the values h/ie^2 ($i = 1,2,...$) which are characteristic of the quantum Hall effect. Quantization of the Hall resistance in InSe is connected with the presence of plane defects in the interior of the crystal. At low temperatures, electrons are localized at these defects, thus forming two-dimensional electron gas regions. In fields $H \gtrsim 30$ kOe, a doubling in the quantum oscillation frequency is observed, due to the lifting of the spin degeneracy of the Landau levels. It is demonstrated that the two-dimensional electrons in InSe belong to the lowest minimum of the conductivity band, this minimum being located at the center of the Brillouin zone.

INTRODUCTION

The presence of a two-dimensional gas of current carriers in the sample is necessary for observation of the quantum Hall effect (QHE), which manifests itself as a plateau in the dependence of the Hall resistance on the magnetic field H. At present, the QHE has been observed and is being studied in detail on the inversion layers of a two-dimensional gas of carriers which are located either near a semiconductor-dielectric boundary, as in the pioneering Ref. 1, or near a heterojunction (see, for example, Ref. 2), or near the boundary of a bicrystal.[3,4]

In the present paper, we describe an observation of the quantum Hall effect, and discuss the results of investigations into it, in single crystals of the layered semiconductor InSe, in which the nature of the two-dimensional carrier gas regions is essentially different. The presence of two-dimensional electron gas regions in the bulk of InSe samples was first communicated in Refs. 5–7. Using the cyclotron resonance and Shubnikov–de Haas oscillations, the authors of these papers demonstrated that the energy separation between the Landau levels was determined only by the component of the magnetic field vector **H** parallel to the C axis of the crystals (directed perpendicular to the layers) and suggested that the existence of two-dimensional electron gas regions in indium selenide was connected with the presence in these crystals of extended plane defects, due to the disordered stacking of the layers.

A recent investigation which two of us took part in,[8] and which studied Shubnikov–de Haas oscillations in InSe, demonstrated that in some samples the magnetoresistance oscillations were characterized by deep minima with an activation dependence of the resistance at the minima on temperature. These results indicated that a quantum Hall effect regime could be realized in the bulk of single crystals of InSe. Results of the investigation of the quantum Hall effect in indium selenide are given below.

METHODOLOGY AND RESULTS OF THE INVESTIGATIONS

For the investigations, we used samples of InSe prepared from monocrystalline ingots that were grown by the Bridgman method. The surface perpendicular to the planes of the layers was prepared by means of a chemical cutting, and the surface containing the layer plane was prepared by simple splitting. In chemical cutting, we used a $HCl:CrO_3$ solution. The samples were in the form of rectangular plates $10 \times 2\ \mathrm{mm}^2$ in area (plane of the layer) and with a depth of 0.3–1 mm. Indium contacts applied to the freshly-cut surface enabled us to measure both the voltage $V_\parallel$ along the current I_0 going through the sample and the Hall voltage V_H (four-point scheme). The measurements were carried out with a variable current of frequency 20 Hz, whose magnitude was at most 1 μA. The magnetic field was directed along the normal to the plane of the layers of the sample, which was located in the center of a superconducting solenoid with a maximum value for the magnetic field of 80 kOe. The signal, which was proportional to $V_\parallel$ or V_H, was recorded on a two-coordinate recorder as a function of the magnetic field. Most of the InSe samples had a positive magnetoresistance in fields greater than a few kilooersteds; as the temperature was lowered, the resistance of the samples increased. On some of the samples, we were able to record magnetoresistance oscillations, which were periodic in H^{-1}. The depth of these oscillations increased significantly if the sample had undergone a preliminary vacuum annealing at a temperature of 400–450 °C, and at the same time the overall resistance of the sample decreased.

In some samples, the dependence of the magnetoresistance on temperature measured in this geometry in the temperature range 1.3–4.2 K was weak or completely absent—that is, the conductivity was metallic in character. The characteristic resistances of such samples was less by one to three orders than the resistance of those samples in which the conductivity decreased with decreasing temperature. We observed quantum oscillations, whose amplitude increased sharply with decreasing temperature, in the magnetoresistance of such samples. The position of the extrema of the oscillations did not change after many cycles of cooling and heating between liquid helium temperature and room temperature. Calibration experiments in an oblique field, like those we had carried out earlier,[8] demonstrated that the position of the extrema of the oscillations was determined only by the component of the **H** vector parallel to the C axis. Samples with the properties listed above were the objects of investigation for the present paper.

Figure 1 shows the dependence of the tractor voltage

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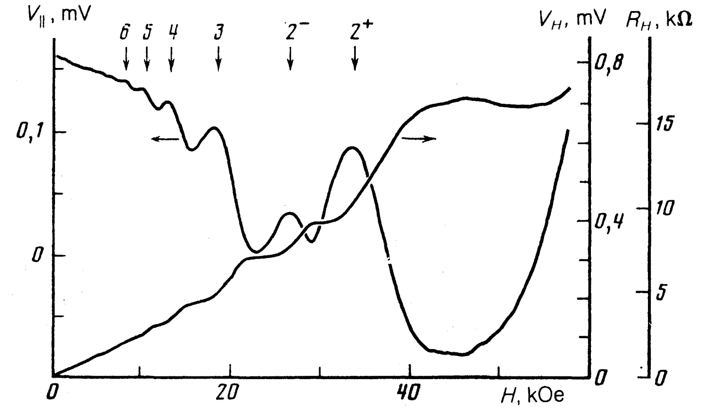

FIG. 1. Dependence of the Hall voltage V_H, the tractor voltage $V_{\parallel}$, and the Hall resistance on the magnetic field for sample No. 1 at $T = 1.3$ K. The numbers indicate the number of the Landau level whose intersection with the Fermi level leads to the appearance of a corresponding maximum in the magnetoresistance; '+' and '−' are the spin orientations.

and of the Hall voltage on the magnetic field, these dependences being recorded in one of the samples at a temperature $T = 1.3$ K. The sign of the Hall signal corresponds to electron conduction. The horizontal plateaus that disappear when the temperature is raised to 4.2 K are easily visible in Fig. 1. The absence of a significant inclination in these plateaus indicates that intermixing of the tractor voltage in the Hall voltage does not occur. The range of values of H that correspond to the plateaus has the minima of the dependence $V_{\parallel}(H)$ located in it. In magnetic fields $H \geqslant 40$ kOe we observe a change in the sign of $V_{\parallel}$ which is connected, in our opinion, with the mixing of the Hall component into the tractor voltage. This mixing can arise because of the complex character of the lines of flux that go through the two-dimensional electron gas region, and because of imperfections of the sample geometry. In sufficiently strong magnetic fields, when $\rho_{xy} \gtrsim 10\rho_{xx}$, these factors can change the value of $V_{\parallel}$ and also its sign. They should not, however, have a significant influence on the position of the minima of the dependence $V_{\parallel}(H)$.

The extrema of the quantum magnetoresistance oscillations at small magnetic fields are disposed periodically, as a function of H^{-1}. In the strong magnetic field region (with $H \geqslant 30$ kOe in Fig. 1) there arise, as a rule, additional extrema. This effect is clearly demonstrated in Fig. 2, where we give data for a sample in which the dependence of the resistance on the field contains a large number of extrema. The fact that the appearance of additional extrema leads to a doubling of the quantum oscillation frequency (Fig. 3) proves to be important for interpreting the results.

DISCUSSION OF RESULTS

Energy spectrum of two-dimensional electrons in InSe; spin splitting

In a two-dimensional electron gas in a transverse magnetic field such that the components of the magnetoresistance tensor obey the condition $\rho_{xy} > \rho_{xx}$, the minima of the Shubnikov–de Haas oscillations are observed when the Fermi level is located between two neighboring Landau levels (in the mobility gap). At $T = 0$ this means that electrons with a concentration N_{2D} completely occupy all the n Landau levels that lie below the Fermi level, that is

$$n\nu eH/ch = N_{2D}. \qquad (1)$$

Here ν is the multiplicity of the degeneracy of the Landau levels, eH/ch is the capacity of one nondegenerate level. With a fixed value of N_{2D} the values of H at which condition (1) is satisfied are disposed periodically in H^{-1} with a period

$$P = \nu e/chN_{2D}. \qquad (2)$$

In the region of sufficiently large values of H, we can have partial or complete lifting of the degeneracy of the lev-

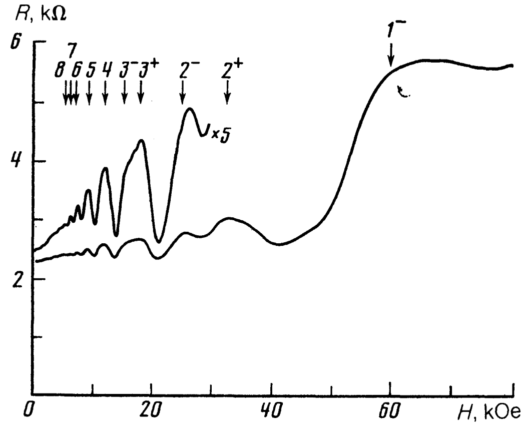

FIG. 2. Dependence of the resistance R on the magnetic field for sample No. 2 at $T = 1.3$ K.

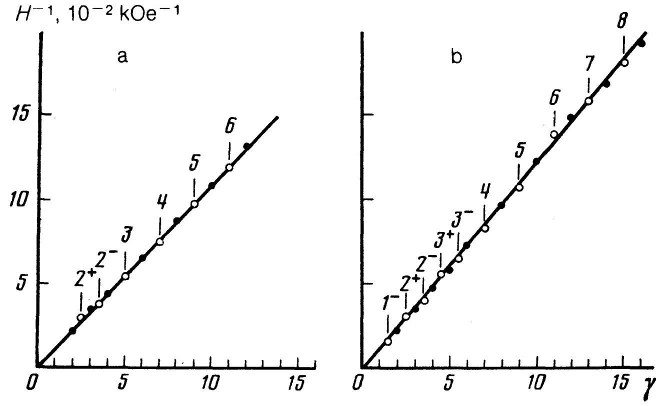

FIG. 3. Dependence of the positions of the oscillation extremes in the scale of H^{-1} on the occupation factor: ●—minima, ○—maxima, a—sample No. 1, b—sample No. 2.

els, which leads to a decrease in ν and, consequently, in P. As can be seen from Fig. 3, in our experimental situation lifting of the degeneracy decreases the value of ν by a factor of two, which we naturally ascribe to the splitting of the Landau levels. A decrease by a factor of two in the period of the quantum oscillations when the spin degeneracy is lifted is characteristic of systems of two-dimensional electrons with a fixed N_{2D} concentration.[9] Since the position of the resonance fields in this case is described entirely in terms of occupation of the Landau levels (1), and the size of the energy gas in the spectrum does not influence the position of the extrema, in order to determine the values of the effective g-factors (g^*) of electrons in systems with a fixed concentration N_{2D}, we need additional experiments in an oblique field.[10]

We cannot, unfortunately, carry out direct measurements of the g-factor analogously to Ref. 10 because of the rapid decay of the quantum oscillations in weak fields. Our data only allow us to estimate the upper bound of the quantity g^*. The fact is that if the magnitude of the spin splitting $\mu_B g^* H$ ($\mu_B = e\hbar/2m_0c$ is the Bohr magneton) is greater than half the energy $\hbar\Omega$ ($\Omega = eH/m_c c$ is the cyclotron frequency, m_c is the cyclotron mass of the electrons) that separates the Landau levels, then the values of the occupation factor $\gamma = N_{2D}(eH/ch)^{-1}$ at which minima of ρ_{xx} are observed prove to be odd in weak fields, in which close-lying levels are not split. The factor γ is even at the minima of ρ_{xx} if the inverse inequality $\mu_B g^* H < \hbar\Omega/2$ is satisfied. According to the data given in Fig. 3, the abscissae of the points that correspond to magnetoresistance minima in the weak field region correspond to even values of γ in our case. Thus, spin splitting of the Landau levels in the two-dimensional electron system of indium selenide is less than $\hbar\Omega/2$, that is $g^* < M_0/m_c \approx 7.7$. For the calculation, we used the value $m_c = 0.13m_0$, which was determined in Ref. 4.

Attempts to estimate g^* for InSe have also been carried out earlier.[7,11] Kress-Rogers *et al.*,[7] observing that lifting of spin degeneracy results in doubling of the frequency of the oscillations in ρ_{xx}, proposed that the spin splitting of the Landau levels was approximately equal to $\hbar\Omega/2$. In Ref. 11, where doubling in the oscillation frequency was not observed, the value $g^* = 4.6$ was determined from the size of the spin splitting of the maxima of the oscillations. This procedure is valid if the system of two-dimensional carriers has a fixed Fermi energy, and does not have a fixed concentration. At the same time, even in a system with a fixed concentration N_{2D}, a monotonic mobility dependence $\mu(H)$, and also nonsymmetric spreading of the Landau levels, may lead to the result that when the spin degeneracy is lifted the splitting of the maxima in ρ_{xx} will not amount to half the period measured in weak fields. Here, however, the size of the splitting does not give information about the value of g^*. We emphasize once more that in all the experiments we carried out (see Fig. 3) the maxima of the quantum oscillations on the H^{-1} scale lie practically in the middle between minima, and lifting the degeneracy makes the separation between maxima equal to half the period measured in weak fields.

Concentration and Hall resistance of the two-dimensional electron gas in InSe

Equation (2) for the period of the quantum oscillations enables us to determine uniquely the concentration N_{2D} of two-dimensional electrons if we assume that complete lifting of the degeneracy of the Landau levels occurs under spin splitting in fields $H \gtrsim 30$ kOe. The assertion that there is no valley degeneracy for electrons in InSe (that is, that the absolute minimum of the conduction band is located at the point $k = 0$ of the Brillouin zone) follows from the results of investigations into cyclotron resonance[5] and, as will be demonstrated below, is in complete agreement with our investigations of the quantum Hall effect. Taking $\nu = 1$ in the strong field region, we obtain the value $N_{2D} = 2.2 \times 10^{11}$ cm^{-2} for a sample which corresponds to the data shown in Fig. 1. The value of N_{2D} determined in a similar fashion for various samples lay in the range $(1.8\text{–}2.3)\cdot 10^{11}$ cm^{-2}.

Having the value of N_{2D} for an actual sample available, and using the results of measuring the Hall effects in weak fields where V_H depends linearly on H (see, for instance, Fig. 1 with $H < 5$ kOe), it would be possible to estimate the real value of the current I_{2D} that flows through the region with two-dimensional electronic conductivity. The fact is that the regions of two-dimensional metallic conductivity under investigation are separated from the contacts by a medium with a significantly smaller conductivity (this follows from a comparison of the resistance of samples that contain or do not contain macroscopic regions of two-dimensional conductivity). Therefore, the value of I_{2D} is determined by the resistance of the high resistance region of the sample and proves to be less than the complete current. At the same time, the V_H is chiefly formed in the two-dimensional region. This follows from the fact that the plateaus observed in the $V_H(H)$ dependence are practically horizontal. Using the data of Fig. 1 and substituting the value of N_{2D} into the formula

$$V_H = \frac{H}{ec}\frac{I_{2D}}{N_{2D}}$$

we can estimate the value of I_{2D} and the portion of the current that goes through the two-dimensional region in relation to the total current I_0: $I_0/I_{2D} \approx 2.3$.

Knowing this quantity, we can determine the quantity $R_H = V_H/I_{2D}$ which relates to the two-dimensional electron gas (R_H in Fig. 1), and we can compare the results obtained for R_H in the plateau region with the known values h/ie^2 ($i = 1,2,3,...$) that characterize the value of the Hall resistance on the plateau in the quantum Hall effect regime. The table gives the quantitites $R_H^* = V_H/I_0$, $R_H = V_H/I_{2D}$ and the values of h/ie^2. We know the values of i for each plateau, since we know the occupation factor (see Fig. 3). There is a satisfactory agreement of the values of R_H obtained here with those which correspond to an integer quantum Hall effect. The systematic excess of the experimental values of R_H over the calculated ones which occurs in strong magnetic fields is connected, apparently, with the redistribution of the current in the sample on account of the magneto-

TABLE I.

	$i=2$	$i=3$	$i=4$	$i=6$	$i=8$	$i=10$
R_H^*, kΩ	7.1	4.0	3.1	1.92	1.37	1.05
R_H, kΩ	16.5	9.3	7.2	4.5	3.2	2.4
h/ie^2, kΩ	12.9	8.6	6.45	4.30	3.23	2.58

resistance, which leads to an increase in I_{2D}. This explains the superlinear dependence V_H (H) which is remarked in Fig. 1 (in fields that correspond to the right-hand plateau the deviation from a linear dependence is about 20%).

The data concerning the values of the Hall resistance quantities of a two-dimensional gas in InSe that are given in the table support the initial assertion that the lowest lying state of the conductance band of this crystal is located in the center of the Brillouin zone. In fact, since there remains only one occupied Landau level, which is spin-split, below the Fermi level in the field that corresponds to the rightmost plateau in Fig. 1, the quantity R_H for this plateau cannot be greater than $h/2e^2 \approx 13$ kΩ. If there should be in InSe a three-fold valley degeneracy, as was proposed in, e.g., Ref. 12, R_H would be smaller by another factor of three. The experimental value of R_H* for this plateau, which is equal to approximately 7 kΩ and bounds R_H below, excludes such a possibility.

Thus, we may consider it to have been demonstrated that the electrons out of which the two-dimensional conductance region in InSe is formed, belong to the minimum of the conductance band that is located at the center of the Brillouin zone. This means that the value of N_{2D} calculated using (2) from the oscillation period and the assumption that $\nu = 1$ in the region of large H was determined correctly.

The nature of the two-dimensional electron gas regions in InSe

The "electronic" sign of the Hall signal that is recorded in the experiment gives evidence of the fact that the system under investigation in InSe constitutes an enriched region with two-dimensional metallic conductivity. In single crystals, a similar system was discovered on a clean Ge surface[13,14] and on internal surfaces of germanium[15] and indium selenide,[3] which constitute the boundaries of bicrystals.

In our experiments, the concentration N_{2D} remains unchanged in the sample after several cycles of heating and cooling between 4.2 and 300 K, and also after resoldering of the contacts. It is difficult to understand this under the assumption that the two-dimensional electron gas region is formed on a free surface, and it may, apparently, give evidence of the connection between the quantum Hall effect we have observed and an "internal" surface that exists in bulk InSe crystals. Layer stacking defects, which are always present in $A^{III}B^{VI}$ semiconductors, and which arise in the form of polytype boundaries or under plastic flow, can act as such surfaces. Interstitial defects are located in the basal planes of the crystals, and are due to the peculiarity of their structure—the presence of a weak bond in the lattice. The presence of extended plane defects in indium selenide is connected with the anisotropy of the electrical properties of this semiconductor—this anisotropy cannot be described in terms of the width of the electron and hole bands that form its optical absorption edge.[16,17]

The connection of two-dimensional electron gas regions with interstitial defects was first announced in Refs. 5, 6, where the suggestion was made that the donor impurities that attach themselves to such interstitial defects at low temperatures and which trap electrons form two-dimensional electron gas regions. There are data which give evidence of the dislocation structure of the interstitial defects in layered semiconductors and of the capacity of the basal dislocations in these crystals to trap carriers. It has been demonstrated experimentally[18] that the basal dislocations that are introduced in GaSe (a structural analog of InSe) as a result of plastic deformation are centers of low-energy localization of excitons, and direct electron-microscope investigations[19] give evidence of the fact that in InSe the basal dislocations right up to 10^{10} cm^{-2} form plane interlayer defects. The fact that thermal annealing of InSe stimulates the creation of two-dimensional electron gas regions supports the assumption concerning their defect nature.

Thus, in indium selenide at low temperatures the electrons are located at plane defects, and thus create two-dimensional electron gas regions with an electron concentration $N_{2D} = (1.8\text{–}2.3)\cdot 10^{11}$ cm^{-2} and mobility $\mu = 10^4$ cm^2/V·s (Ref. 8). At 4.2 K, these regions are responsible for the quantum Hall effect in InSe.

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