

An investigation of the effect of a magnetic field on the chemical potential of electrons in bismuth and in a GaAs-Al_xGa_{1-x}As heterojunction

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The effect of a magnetic field on the chemical potential of the electrons in bismuth and in a GaAs-Al_xGa_{1-x}As heterojunction has been investigated by measuring the contact potential difference. No chemical-potential oscillations due to oscillations of the density of states were observed for bismuth, and it is shown that they may be absent because of the compensating effect of magnetostriction. Sawtooth oscillations of the chemical potential were observed on studying a GaAs-Al_xGa_{1-x}As heterojunction, the decreasing sections of the oscillations coinciding with the plateau regions of the quantum Hall effect. The results point to an oscillatory dependence of the electron concentration on the magnetic field strength and allows one to explain the quantum Hall effect without invoking the localization concept.

We reported earlier¹ on a search for oscillations of the chemical potential μ of electrons in beryllium, associated with oscillations of the density of states in a high magnetic field. Kaganov *et al.*² were the first to draw attention to the possibility of such oscillations. It turned out, as a result of the measurements, that the chemical potential of the electrons in beryllium does not oscillate. To put it more precisely, the amplitude of oscillations of the chemical potential did not exceed the noise level ($1.5\text{--}2\ \mu\text{eV}$), which is an order of magnitude smaller than the estimated² value of $|\mu_{\text{osc}}|$. This result was attributed to the fact that the oscillations in the chemical potential, due to the oscillations of the density of states, are cancelled by chemical-potential oscillations due to magnetostrictive oscillations of the volume. Such cancellation is possible if the electron contribution to the bulk modulus of the metal dominates.

It was of interest to study the effect of a magnetic field on the chemical potential of the electrons in bismuth, which has $\sim 10^4$ -fold fewer current carriers than beryllium. The small number of current carriers in bismuth should, on the one hand, lead to a large magnitude of the oscillations of the chemical potential, associated with oscillations of the density of states, and on the other hand should make the electron contribution to the bulk modulus negligibly small. According to Brignall and Shoenberg,³ the amplitude of the oscillations of the magnetization of bismuth in a field $H = 7\ \text{kOe}$ at $T = 4.2\ \text{K}$ amounts to $|M_{\text{osc}}| \approx 0.07\ \text{G}$, which should lead to oscillations of the chemical potential with an amplitude $|\mu_{\text{osc}}| = 2/3 |M_{\text{osc}}| H / N \approx 300\ \mu\text{eV}$ ($N = 6 \times 10^{17}\ \text{cm}^{-3}$ is the total number of electrons and holes in bismuth⁴).

In the present work, in addition to the investigation of the chemical potential of electrons in bismuth, we determined the effect of a magnetic field on the chemical potential of electrons in a GaAs-Al_xGa_{1-x}As heterojunction. In the latter case the electron occupy a two-dimensional layer with surface density $N_s \approx 5 \times 10^{11}\ \text{cm}^{-2}$, which corresponds to a volume concentration $N \approx 5 \times 10^{17}\ \text{cm}^{-3}$, typical of semimetals. The principal difference consists in the change in the dimensionality of the space occupied by electrons.¹⁾

THE EXPERIMENTS

The effects of a magnetic field on the chemical potential of electrons can be determined from the change in the contact potential difference between the plates of a measuring capacitor consisting of the specimen being studied and a polycrystalline electrode. In general, the contact potential difference differs from the difference of the chemical potentials by the magnitude of the potential difference of the doubly charged layers present on the surface of metals.⁸ However, the influence of a magnetic field on this correction can, evidently, be neglected.⁹

We shall consider the limits imposed on the parameters of the external circuit connected to the measuring capacitor, and on the rate of change of the magnetic field. Suppose that the measuring capacitor is formed by bringing together metals *A* and *B* with chemical potentials μ_A and μ_B ($\mu_A > \mu_B$), while the external circuit has infinite resistance (Fig. 1a). It is clear that there is no electric field in this case, since the electrons of metal *A* are "unaware" of the existence of empty

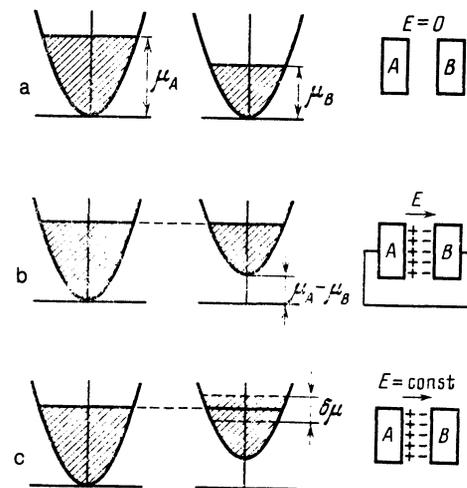


FIG. 1. A schematic explanation of the effect of a resistance in the external circuit on the electric field in the measuring capacitor.

places in metal *B*. On shorting the measuring capacitor by a wire (Fig. 1b), a flow of electrons from metal *A* to the unoccupied states of metal *B* takes place until the electrochemical potentials $\mu + e\varphi$ of both metals are equalized. As a result, a potential difference $\Delta\varphi = (\mu_A - \mu_B)/e$ arises between the plates of the measuring capacitor. If the wire is removed again (Fig. 1c) and some external action changes the chemical potential of one of the metals (*B*), then the difference in potential will not vary, since a change of the capacitor voltage for a constant capacitance is possible only if the capacitor charge is changed. If the external circuit has resistance *R*, the voltage on the measuring capacitor will change with a time constant $\tau_1 = RC$ (*C* is the capacitance of the measuring capacitor) and rapid changes in chemical potential with characteristic period $\tau_2 \ll \tau_1$ will not be discernible. An upper bound of the allowable rate of change of magnetic field follows from this. It should be noted that the experiment recommended by Kulik and Gogadze¹⁰ for observing oscillations of chemical potential contradicts this reasoning.

Two methods for measuring oscillations of chemical potential were used in this work. The first method was the same as that already described.¹ It is in fact based on the determination of the charge-transfer current of the measuring capacitor from the voltage drop across the resistor *R* ($\sim 10^{12} \Omega$) connecting its plates. The voltage drop was measured by a voltmeter-electrometer with a dynamic capacitor at the output. The transfer characteristic of the measuring device was resonant and was described by the expression

$$A\ddot{y} + B\dot{y} + Cy = \dot{x}, \quad (1)$$

where *x* is the potential of the specimen studied and *y* is the output voltage. The coefficients which enter in Eq. (1) were determined from the amplitude-frequency and phase-frequency characteristics of the measuring device:

$$k(\omega) = B^{-1} [1 + (C/B\omega - A\omega/B)^2]^{-1/2}, \quad (2)$$

$$\varphi(\omega) = \arctg(C/B\omega - A\omega/B),$$

and also from the reaction of the measuring device to a linearly varying input voltage applied to the specimen studied. The initial value of the specimen potential was deduced by numerical integration of the plots of the output voltage in conformity with Eq. (1). The accuracy in this reconstruction is illustrated in Fig. 2, which shows the reaction of the measuring device to a linearly increasing voltage and the reconstructed signal.

The second method for measuring oscillations of chemical potential is based on the direct determination of the charge on the measuring capacitor. The measuring capacitor was connected for this purpose to an electrometer operating in the current-integrating mode. The output signal from the electrometer is in this case proportional to the charge on the measuring capacitor and therefore to the potential difference between its plates.

The advantage of the first method lies in its high sensitivity to changes of chemical potential; the merit of the second is the greater accuracy, since it requires practically no additional analysis of the electrometer output traces. The first method was therefore used in searching for oscillations in the chemical potential of beryllium and bismuth, while the

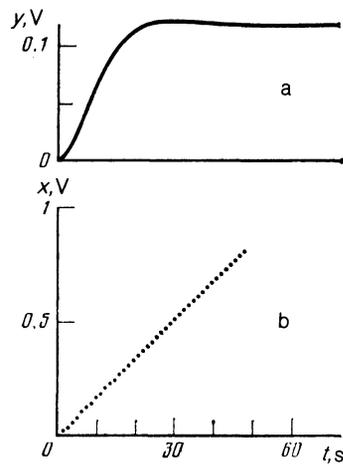


FIG. 2. The reaction of the measuring system to a linear growth of voltage (a) and the reconstruction of the input signal (b).

second was used for a more detailed study of oscillations in the chemical potential of a GaAs- $\text{Al}_x\text{Ga}_{1-x}\text{As}$ heterojunction.

The designs of the measuring capacitors rated 5–8 pF were similar to those used before (Fig. 2b of Ref. 1). The working gap of the measuring capacitor was parallel to the field in the study of bismuth and perpendicular in the study of heterojunctions.

The bismuth specimen was a $7 \times 5 \times 1$ mm plate with the normal along the trigonal axis. The magnetic field was directed along the trigonal axis. The magnetic field was directed parallel to the bisectrix. The resistivity ratio was $\rho_{300\text{K}} / \rho_{4.2\text{K}} = 72.4$.

The heterojunction specimens were similar in structure to the specimens on which the quantum Hall effect was studied. They were grown by the method of molecular epitaxy on a single-crystal semi-insulating GaAs substrate in the following sequence: undoped GaAs, 0.8 μm ; undoped $\text{Al}_x\text{Ga}_{1-x}\text{As}$, 100 \AA ; $\text{Al}_x\text{Ga}_{1-x}\text{As}$, doped with Si to a concentration of $5 \times 10^{17} - 10^{18} \text{ cm}^{-3}$, 500 \AA ; undoped GaAs, 200 \AA . The carrier mobility was $\sim 10^5 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, the specimen dimensions 10×10 mm. All measurements were carried out at $T = 4.2$ K.

RESULTS

Bismuth

As when studying beryllium,¹ oscillations of magnetoresistance were used as a reference in searching for oscillations of the chemical potential. To show up the oscillatory component better, the monotonic component of the magnetoresistance of bismuth was compensated by the magnetoresistance signal of a polycrystalline beryllium specimen. Such a trace of the oscillations is shown in Fig. 3a; the amplitude of the oscillations in a field of 7.5 kOe amounted to about 10% of the total magnetoresistance signal of bismuth.

In the present work an attempt was made to find an oscillation of the chemical potential of bismuth corresponding to the oscillation in magnetoresistance in a field of 7.5 kOe. Since the transfer characteristic of the measuring device was resonant, it would be advantageous to sweep the

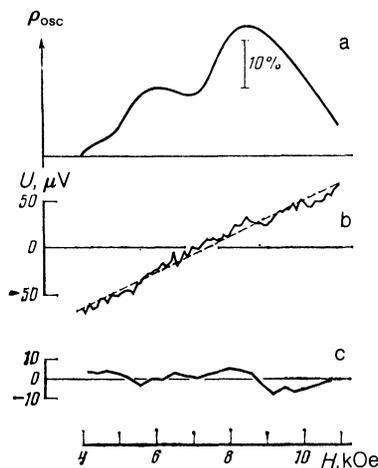


FIG. 3. The search for oscillations of the chemical potential of bismuth: a) oscillations of the magnetoresistance of bismuth; b) the output voltage of the measuring system, averaged over 24 traces in a decreasing magnetic field; c) the output voltage of the measuring system averaged over 48 traces in a decreasing and increasing magnetic field.

magnetic field at a rate corresponding to the maximum transfer coefficient of the measuring system:

$$|\dot{H}| = H^2 \Delta(1/H) \omega_p / 2\pi, \quad (3)$$

where $\Delta(1/H)$ is the oscillation period of the chemical potential, and ω_p is the resonance frequency of the measuring system. It was technically difficult to satisfy Eq. (3) over a wide range of fields, so that we confined ourselves to sweeping the field at a constant rate, satisfying Eq. (3) only at $H = 7.5$ kOe: $|\dot{H}| = \text{const} \approx 80 \text{ Oe} \cdot \text{s}^{-1}$. The magnetic field dependence of the voltage at the output of the measuring system is shown in Fig. 3b obtained by averaging 24 traces in a decreasing magnetic field. It is a straight line straddled by a noise track $\pm 10 \mu\text{V}$ in width. The slope of the line is produced by the induction signal arising from the large rate of change in the magnetic field. A similar relation is shown in Fig. 3c, obtained by averaging 48 traces made alternately in increasing and decreasing magnetic field. An additional smoothing of the curve over three adjacent points was carried out to reduce the high-frequency noise.

It follows from Fig. 3c that oscillations of the chemical potential of bismuth in a field of 7.5 kOe do not exceed $5 \mu\text{eV}$, which is nearly two orders of magnitude less than the expected value.

GaAs-Al_xGa_{1-x}As heterojunction

Unlike the negative results obtained on beryllium and bismuth, we found a strong effect of a magnetic field on the chemical potential of the two-dimensional electrons in a GaAs-Al_xGa_{1-x}As heterojunction. The traces of the voltage at the output of the measuring system were taken by sweeping the magnetic field in the range 15–50 kOe at the constant rates $|\dot{H}| = 14.4, 57.5, 86.3$ and $115 \text{ Oe} \cdot \text{s}^{-1}$. Examples of such traces are shown in Fig. 4. The traces were repeated several times for each value of H and averaged. The main error on reconstructing the specimen potential according to Eq. (1) was due to the drift of the electrometer zero. Within the limits of this error, the reconstructed specimen

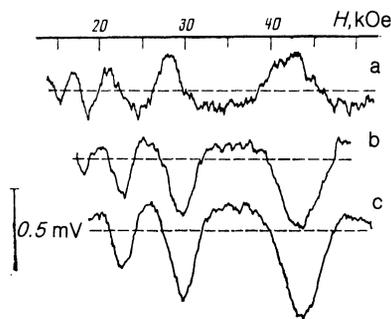


FIG. 4. Traces of the output voltage of the measuring system produced when studying a heterojunction. The rates of changing the magnetic field were a) $\dot{H} = -57.5 \text{ Oe} \cdot \text{s}^{-1}$; b) $\dot{H} = 86.3 \text{ Oe} \cdot \text{s}^{-1}$; c) $\dot{H} = 115 \text{ Oe} \cdot \text{s}^{-1}$. The dashed horizontal lines indicate the zero values of the signals.

potential was dependent neither on the rate nor on the direction of the change in magnetic field. The magnetic field dependence of the change in chemical potential of the heterojunction obtained in such a way is shown in Fig. 5a.

It is interesting to compare the change in chemical potential of the heterojunction in a magnetic field with the results of measurements of magnetoresistance ρ_{xx} and Hall effect ρ_{xy} (Fig. 5). The $\rho_{xx}(H)$ and $\rho_{xy}(H)$ plots have shapes typical of the quantum Hall effect¹¹: a succession of plateaus on the $\rho_{xy}(H)$ curve and deep minima in the $\rho_{xx}(H)$ curve corresponding to them. The magnetic field dependence of the chemical potential of the heterojunction has a sawtooth appearance, with the smooth fall in potential taking place in those intervals where $\rho_{xy}(H) = \text{const}$.

Similar magnetic field dependences of the chemical potential of the heterojunction were also obtained by determining directly the charge on the measuring capacitor. An example of the trace of the electrometer output voltage in this case is shown in Fig. 6a.

DISCUSSION

Bismuth

As when studying beryllium,¹ we could not observe oscillations of the chemical potential of bismuth. Moreover, the margin in sensitivity (the ratio of the expected oscilla-

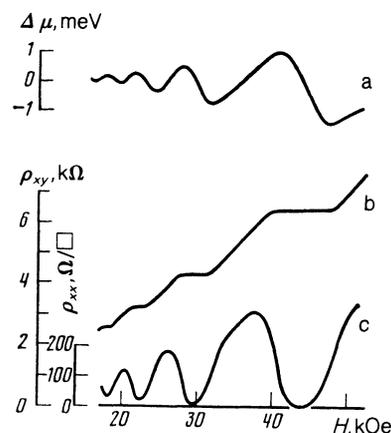


FIG. 5. Comparison of results of measurements of a) the chemical potential, b) the Hall effect and c) the magnetoresistance of a heterojunction. The field dependence of the chemical potential was obtained by numerical analysis of the traces of Fig. 4 (the first measurement method).

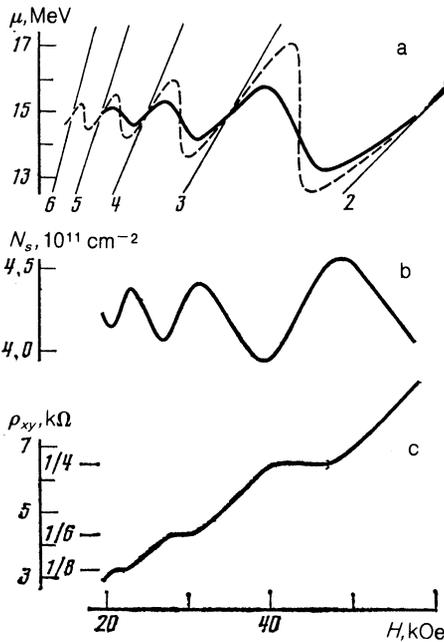


FIG. 6. a) Comparison of the experimental (solid curve) and theoretical (dashed curve) magnetic field dependences of the chemical potential of a heterojunction. The experimental $\mu(H)$ dependence was obtained by integration of the charge transfer current of the measuring capacitor (the second measurement method). The theoretical $\mu(H)$ dependence was calculated on the assumption that $N_s = \text{const}$. The Landau levels $\varepsilon_n = \hbar\omega_c (n + 1/2)$ are shown by the sloping straight lines. b) The magnetic field dependence of the electron concentration, calculated from the measured $\mu(H)$ dependence. c) The Hall resistivity calculated from measurements of the $\mu(H)$ dependence. The horizontal strokes indicate the positions of the plateaus in units of h/e^2 .

tion amplitude to the noise) for bismuth was almost an order of magnitude greater (≈ 60).

At first sight, this result contradicts existing views on the effect of a magnetic field on the spectrum of electrons and holes in bismuth.⁴ In fact, according to Vecchi *et al.*¹², for example, a magnetic field redistributes the electrons over the valleys and changes their overall number and the Fermi energy; this explains the singularities of the reflection spectra of bismuth in quantizing magnetic fields. In addition, since the condition for electroneutrality requires that

$$\sum_{i=1}^3 \Delta N^{(i)} = \Delta P \quad (4)$$

($\Delta N^{(i)}$ is the change in the number of electrons in the i th valley, ΔP is the change in the number of holes), oscillations in the number of electrons should lead to oscillations in the number of holes, as is also confirmed by experiment.⁴

We note, however, that when considering bismuth, the Fermi energies of the carriers are measured either from the edges of the energy band or from the energy of the level with quantum number $j = (n + 1/2) \pm s = 0$. The values of the Fermi energies for different groups of carriers in bismuth therefore do not coincide, and these energies are not related to the chemical potential, defined as $\mu = \partial F / \partial N$ (F is the free energy). It can also be noted that thanks to the condition of electroneutrality [Eq. (4)], the shift of chemical potential drops out of the calculations of the change of free energy in a magnetic field [see Eq. (8) of Ref. 13]. The absence of

oscillations of the chemical potential of bismuth thus does not on the whole contradict results on oscillations of the number of carriers.

As in the case of beryllium, it was natural to try to attribute the absence of oscillations of the chemical potential of bismuth to the compensating effect of magnetostriction, the more so because bismuth has the largest magnetostrictions of all the diamagnetic materials.¹⁴ However, the carrier concentration in bismuth is so low that the electron contribution to its bulk modulus can be neglected (according to estimates, $B_{el}/B_{\text{tot}} \sim 10^{-7}$). The mechanism of compensation of the oscillations of the chemical potential proposed earlier¹ for metals with a high electron concentration, therefore does not operate in bismuth.

Nevertheless, the possibility of a magnetostrictive compensation of oscillations of the chemical potential in bismuth remains. In fact, the existing experimental results^{15,16} and the deformation-potential approximation¹⁷ indicate that the magnetostrictive deformation u_{ik} behaves similarly to the change in the number of light carriers:

$$u_{ik} = K_{ik} \delta N. \quad (5)$$

Since¹⁸

$$\varepsilon_F \propto N^{2/3}, \quad u_{ik} = -\partial \ln F / \partial \sigma_{ik} \mathbf{M}_{\text{osc}} \mathbf{H}$$

(F is the magnetization oscillation frequency), the magnetostrictive change of chemical potential

$$\delta \mu = \frac{2}{3} \varepsilon_F \frac{\delta N}{N} = -\frac{2}{3} \frac{\varepsilon_F}{K_{ik}} \frac{\partial \ln F}{\partial \sigma_{ik}} \frac{\mathbf{M}_{\text{osc}} \mathbf{H}}{N}$$

will compensate the oscillations of the chemical potential associated with oscillations of the density of states,

$$\mu_{\text{osc}} = -\frac{2}{3} \mathbf{M}_{\text{osc}} \mathbf{H} / N,$$

under the condition

$$K_{ik} = -\varepsilon_F \partial \ln F / \partial \sigma_{ik}. \quad (6)$$

According to the model of McClure and Choi,¹⁹ the Fermi energy of bismuth is $\varepsilon_F \approx 35$ meV, while the measured magnetostriction in the direction of the bisectrix¹⁵ gave $\partial \ln F / \partial \sigma_{22} = (4.7 \pm 0.8) \times 10^{-10} \text{ cm}^2 \cdot \text{dyne}^{-1}$. Oscillations of the chemical potential should therefore be absent if $K_{22} = -(2.6 \pm 0.4) \times 10^{-23} \text{ cm}^3$. This value agrees surprisingly well with the value determined from the parameters of the deformation potential of bismuth for just this magnetic field direction, $K_{22} = -3.04 \times 10^{-23} \text{ cm}^3$ (Ref. 16). Bismuth is thus one more metal in which oscillations of the chemical potential are absent because of the compensating effect of magnetostriction.

It should be noted that our results of looking for oscillations of the chemical potential of beryllium and bismuth disagrees with the results of other works²⁰⁻²³ in which the finding of oscillations of the chemical potential of a number of metals was reported. We note, however, that in all these studies the method recommended by Kulik and Gogadze¹⁰ was used to record the voltage on the measuring capacitor in so rapidly a varying magnetic field that the time for one oscillation was appreciably shorter than the discharge time of the capacitor. We criticized the idea of this method above. In addition, Caplin and Shoenberg have already rejected their earlier results and consider that the large-amplitude

oscillations which they observed are a consequence of the Hall effect and eddy currents.²⁴ Parasitic oscillations of the signal could also arise as a result of magnetostriction amplified by the piezo-effect in the solid dielectric of the measuring capacitor. According to Shoenberg,²⁵ there has not been a really convincing experimental confirmation of oscillations of chemical potential.

The GaAs-Al_xGa_{1-x}As heterojunction

A study of the influence of a magnetic field on the chemical potential of electrons in a GaAs-Al_xGa_{1-x}As heterojunction is especially interesting for explaining the quantum Hall effect, which manifests itself in plateaus arising in the field variations of Hall voltage, corresponding to the quantized values $\rho_{xy} = h/ie^2$ ($i = 1, 2, \dots$), at a longitudinal resistivity $\rho_{xx} = 0$.¹¹ The general explanation of the quantum Hall effect is that the electrons occupy a whole number of Landau levels and their chemical potential lies either in the energy gap or in the mobility gap.

One of the first explanations of the quantum Hall effect in heterojunctions²⁶ was based on the existence of an electron reservoir outside the two-dimensional electron layer. Since the degeneracy of each Landau level is $N = eH/ch$, to occupy i levels we need $N_s = ieH/ch$ electrons. Then $\sigma_{xy} = N_s ec/H = ie^2/h$, while $\sigma_{xx} = 0$, which gives $\rho_{xy} = h/ie^2$ and $\rho_{xx} = 0$. The electron reservoir must offer a possibility of changing N_s over sufficiently wide limits, in order to achieve observable widths of the ρ_{xy} plateaus. According to Baraff and Tsui,²⁶ donor impurities near the two-dimensional electron layer could be such a reservoir, but doubts later arose about the reality of this estimate²⁷ and the idea of a reservoir was abandoned.

The idea of localized states was set forth to replace it and was recently developed intensely. According to this idea, the conduction is produced by electrons with energy lying near the centers of the Landau levels, while all the remaining electrons ($\approx 95\%$) lying on the wings of the Landau levels are localized. Thanks to a peculiar compensation, the current carried by the delocalized states then turns out to be equal to the current from all the states of the filled Landau levels.²⁸ In this case, a finite Hall resistance plateau width is also possible for a constant electron concentration; it is important that the chemical potential of the electrons should lie in the range of localized states. Although the idea of localization in a random potential gives a qualitative understanding of the quantum Hall effect, there is however so far no quantitative theory, while localization has no complete experimental confirmation.²⁹

It was of interest to compare the variation of chemical potential of a heterojunction as measured in the present work with the theoretical, calculated for $N_s = \text{const}$. The density of the two-dimensional electron gas can be represented in the form³⁰

$$N_s = 2 \left(\frac{2}{\pi} \right)^{1/2} \frac{eH}{ch\Gamma} \sum_n \int_0^\infty \exp \left[-2 \left(\frac{\varepsilon - \varepsilon_n}{\Gamma} \right)^2 \right] \times \left[1 + \exp \frac{\varepsilon - \mu}{k_B T} \right]^{-1} d\varepsilon. \quad (7)$$

This expression is obtained on the assumption that the spin splitting can be neglected, while the broadening of the Landau levels $\Gamma_n = \hbar\omega_c (n + 1/2)$ is described by a Gaussian distribution with the parameter Γ independent of magnetic field.

The theoretical $\mu(H)$ dependence was determined by numerical solution of Eq. (7). The effective mass was taken as³¹ $m^* = 0.068 m$, the electron density $N_s = 4.23 \times 10^{11} \text{ cm}^{-2}$ was calculated from the period of the conductivity oscillations $\sigma_{xx} = \rho_{xx} / (\rho_{xx}^2 + \rho_{xy}^2)$, the broadening $\Gamma = 0.15 \text{ meV}$ of the Landau levels was estimated from the carrier mobility.

The comparison of the experimental and theoretical $\mu(H)$ dependence is shown in Fig. 6a. Since only the increase in chemical potential $\Delta\mu(H)$ was determined in the experiment, the position of the measured dependence on the energy scale was chosen in such a way that the chemical potential scale should intersect the Landau levels at field values corresponding to the maxima of the conductivity σ_{xx} . It can be seen from Fig. 6a that the falling sections of the experimental $\mu(H)$ curve are appreciably broader and less steep than those calculated on the assumption that $N_s = \text{const}$. It should be noted that even a ten-fold increase in the width of the Landau levels did not improve the agreement: the theoretical curve for $H > 25 \text{ kOe}$ has the same steep falling sections and only the amplitude of the $\mu(H)$ oscillations decreased somewhat.

The assumption of the constancy of the electron concentration thus does not agree with the results of measurements of the magnetic field dependence of the chemical potential of the two-dimensional electron gas in a heterojunction. It was therefore natural to use the measured $\mu(H)$ dependence to determine the effect of a magnetic field on the density N_s of a two-dimensional gas. Equation (7) was used to calculate $N_s(H)$ with the parameters $m^* = 0.068 m$ and $\Gamma = 0.15 \text{ meV}$. The $N_s(H)$ dependence obtained in this way (Fig. 6b) has an oscillatory character; the change of electron concentration for $H > 30 \text{ kOe}$ exceeds 10%. If the Hall resistivity $\rho_{xy}(H) = H/ecN_s(H)$ is calculated starting from the $N_s(H)$ dependence, then a relation typical of quantum Hall effect (Fig. 6c) is obtained and agrees excellently with that measured directly (Fig. 5b).

The experimental results of the present work thus reawake the idea of an electron reservoir and enable the quantum Hall effect to be explained without invoking localization.³² We note, however, that the accuracy in measuring $\mu(H)$ does not allow the existence of localized states to be ruled out completely, and the question of the possible influence of localization on the accuracy of the reproducibility of the quantum values $\rho_{xy} = h/ie^2$ is not removed by our experiments.

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¹¹The effect of a magnetic field on the chemical potential of two-dimensional electrons on an Si surface has been studied by several authors.⁵⁻⁷

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