

Oscillations of the photocurrent in a magnetic field during intense photoexcitation of doped germanium

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(Submitted July 18, 1977)
Zh. Eksp. Teor. Fiz. 73, 2337-2346 (December 1977)

It is shown experimentally that the low mobility of electron-hole drops in germanium doped with impurities in amounts $N > 10^{15} \text{ cm}^{-3}$ may result in occupation of a considerable proportion of a crystal by the electron-hole condensate. This gives rise to percolation in the drop system: the photocurrent then rises strongly and begins to oscillate as a function of the magnetic field. The oscillations are due to the fact that when the Landau levels of electrons in a drop cross the Fermi level, oscillations of the equilibrium density of the condensate are generated and this affects the drop size the percolation conditions.

PACS numbers: 71.35.+z, 72.40.+w

According to the current ideas, a two-phase system comprising an electron-hole condensate and a rarefied "vapor" forms in germanium when the rate of interband photoexcitation is sufficiently high and the temperature is sufficiently low.^[1,2] The condensate is a metallic Fermi system with a degenerate spectrum of electrons and holes.

In a magnetic field H the carrier spectrum is of the form $\varepsilon = \varepsilon_r(H) + p^2/2m^*$ ($r = 0, 1, 2, \dots$), where the momentum p and mass m^* represent one-dimensional motion along H . The density of states has maxima near the bottom edges of the Landau bands $\varepsilon = \varepsilon_r$. Therefore, when H is varied so that the condition

$$\varepsilon_r = \varepsilon_F \quad (1)$$

(ε_F is the Fermi energy) is satisfied successively for $r = 1, 2, 3, \dots$, all the parameters of the condensate exhibit oscillations. These oscillations have been observed in germanium by optical methods.^[3-7] We shall study the manifestation of these oscillations in the electrical properties of germanium. A similar attempt has been made earlier.^[8] However, as found subsequently, the oscillations observed on that occasion^[8] were of completely different physical origin.^[9,10]

When the electrical properties are investigated, the shape of the regions in a crystal occupied by the condensate becomes very important. Under steady-state conditions in pure undeformed germanium the condensate is present in the form of drops of $r = 2-5 \mu$ radius.^[2] The drop radius is governed by the dynamic equilibrium in the gas-liquid system and this equilibrium is affected significantly by recombination processes inside a drop. In a pure semiinfinite crystal the possibility of creating a high density of drops is restricted by the repulsion forces between the drops, which cause them to scatter all over the crystal.^[11,12] The nature of these forces is not well understood; they may be associated with the phonon wind, which appears as a result of recombination processes inside the drops.^[13] It would seem possible to force the coalescence of drops by preparing a small crystal. However, we shall show that this method is ineffective under steady-state excitation conditions evidently because of the fast recombination on the surface of a crystal.

The situation is somewhat different in doped germanium. The presence of impurities hinders greatly the diffusion of drops.^[14,15] Therefore, as reported below, coalescence of drops and the appearance of macroscopic regions (possibly multiply connected) filled with the condensate can be achieved even in a bulk doped germanium crystal by a suitable increase in the pumping rate. Oscillations of the photocurrent in germanium subjected to a surface magnetic field are observed under these conditions (see below).

EXPERIMENTAL METHOD

A germanium plate with welded gold wires (line electrodes) was placed at the center of a superconducting solenoid immersed directly in liquid helium in such a way that the surface with the electrodes was perpendicular to the magnetic field H . The plate could be rotated about this position through a small angle, which altered the direction of H relative to the crystallographic axes (Fig. 1). This was used for tuning purposes. In all the experiments the field was directed along the $[100]$ axis. One of the electrode pairs (either the upper pair 1 and 2 or the lower pair 3 and 4) was subjected to a static voltage and the photocurrent passing through the electrodes was recorded by a X-Y plotter as a function of the reciprocal magnetic field H^{-1} . In this way only two of four contacts were used simultaneously.

Light reached a Dewar through a window in its upper cover; this light was focused into a spot 2-3 mm in diameter which covered the electrodes 1 and 2. The light

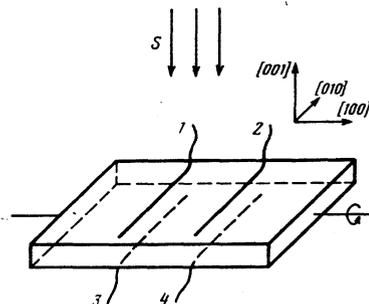


FIG. 1. Positions of contacts on a germanium sample; here, S is the light flux incident on the sample.

source was either a He-Ne gas laser (0.63μ) or a cw GaAs laser (0.85μ): the change in the wavelength did not affect the results. The maximum power which could be directed to the surface of the sample was about 50 mW.

Most of the experiments were carried out in superfluid helium. The experiments at higher temperatures were conducted with the helium pumping system shut off so as to ensure that boiling did not interfere with the optical excitation of the sample. The temperature of the sample was monitored separately.

Our experiments were carried out on *p*-type germanium doped with gallium in concentrations N_{Ga} from 1×10^{15} to $15 \times 10^{15} \text{ cm}^{-3}$, and on *n*-type germanium, doped with antimony in concentrations N_{Sb} from 2×10^{15} to $6 \times 10^{15} \text{ cm}^{-3}$. Moreover, several control experiments were carried out on pure germanium ($N_A + N_D \approx 10^{12} \text{ cm}^{-3}$). In most cases the samples had the dimensions $4 \times 4 \times 0.3 \text{ mm}$. A comparison of the photocurrents flowing through the contacts on the illuminated and dark sides of the samples with these impurity concentrations indicated that the thickness 0.3 mm was considered to be greater than the photocarrier diffusion length l ; the ratio J_{34}/J_{12} was less than 0.1. (This comparison was carried out at low excitation rates so that the value of l was governed by photocarriers rather than by drops.) In this connection we should recall that similar experiments gave $J_{34}/J_{12} \approx 1$ for less heavily doped samples.^[10]

Two series of experiments were carried out on samples of different dimensions. The possibility of filling a pure germanium sample with the condensate without significant overheating was checked and the ideas on the coalescence of drops in doped germanium were tested by experiments on thin (about 0.1 mm thick) samples. On the other hand, large samples ($8 \times 8 \times 1.5 \text{ mm}$) were used in the determination of the temperature dependences. An increase of the surface area by a factor of 10 improved the thermal contact between a sample and helium. Moreover, the greater thickness of these samples prevented photocarriers from reaching the lower surface. Hence, the lower pair of contacts could be used as a thermometer which was calibrated first in the absence of illumination. It was found that overheating of such a large sample in superfluid helium was about 0.1°K at the highest rate of excitation, whereas in normal helium the overheating was 0.5°K .

A simple test showed that there was practically no temperature gradient across a sample. Focusing of the light beam into a relatively small (about 0.5 mm in diameter) spot and moving of this spot across the upper surface of the sample demonstrated that the temperature measured with such a thermometer did not change even when the distance between the thermometer and the point of excitation was varied by a factor exceeding 3. This was understandable because the thermal resistance was concentrated at the crystal-helium interface.

The temperature of the helium bath was found in each case (it is given in the caption of each figure). Estimates deduced from the thermometric experiments described above, subject to allowance for the dimensions

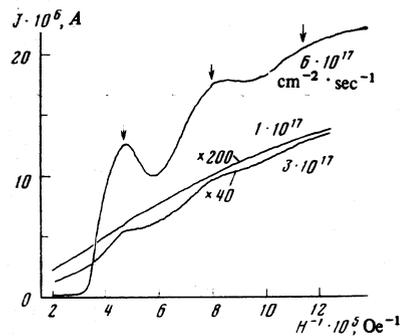


FIG. 2. Dependences of the photocurrent on the reciprocal magnetic field applied to a gallium-doped germanium sample with $N_{Ga} = 6 \times 10^{15} \text{ cm}^{-3}$; $T = 1.6^\circ \text{K}$, $E = 2 \text{ V/cm}$. The rate of excitation (the number of photons incident on a unit surface area per second) is given alongside each curve.

of the sample and the rate of excitation, indicated that the temperature rise in the lattice in these experiments did not exceed 0.1°K . Strictly speaking, these estimates were inapplicable to the electron temperature in the region adjoining the excitation zone. Since electron-electron collisions in these regions were very frequent because of the high carrier density, the transfer of energy from a single hot electron to the lattice took place (at least partly) through the electron system. This could result in a difference between the electron and lattice temperatures. However, our results on the sensitivity of the observed oscillations of the photocurrent to the temperature of the sample demonstrated (as shown later) that any difference between the electron and lattice temperatures was very slight.

RESULTS

1. Figure 2 shows examples of records of the photocurrent in a sample with a gallium concentration $N_{Ga} = 6 \times 10^{15} \text{ cm}^{-3}$ obtained for different interband excitation rates. It is clear from these records that an increase in the excitation rate produced oscillations. Although the third maximum could not be observed clearly, one could nevertheless say that the maxima were equidistant and oscillations periodic in terms of the reciprocal magnetic field. The positions of the maxima were independent of the excitation rate, temperature, and other parameters, which could be varied experimentally. When the temperature was increased, the oscillation amplitude decreased and the oscillations disappeared completely when the lattice temperature reached 2.8°K .

These oscillations were exhibited also by samples with lower impurity concentrations right down to $N_{Ga} = (1-1.5) \times 10^{15} \text{ cm}^{-3}$ (samples with $N_{Ga} = 1 \times 10^{15} \text{ cm}^{-3}$ exhibited the magnetoimpurity oscillations^[9] as well as the long-period—though very weak—oscillations described here). The oscillations were not observed for samples with high impurity concentrations such as $N_{Ga} = 15 \times 10^{15} \text{ cm}^{-3}$.

Figure 3 shows the records of oscillations obtained for samples with different impurity concentrations. Separation between the first and second maxima (period P) was approximately the same when expressed in terms of the reciprocal field: it ranged from 3.3×10^{-5} to 3.5

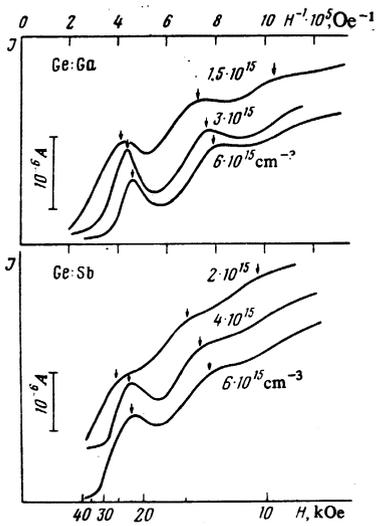


FIG. 3. Oscillations of the photocurrent in *n*- and *p*-type germanium samples with different impurity concentrations (given alongside the curves); $T=1.6^\circ\text{K}$, $E=2\text{ V/cm}$. For convenience, the curves are displaced by arbitrary amounts relative to one another along the vertical axis. For each sample the rate of excitation is selected to maximize the oscillation amplitude (see text).

$\times 10^{-5}\text{ Oe}^{-1}$. The differences were random and not correlated with the impurity concentration. Therefore, we assumed that the oscillation period P was the same for all the samples and equal to

$$P=3.4 \cdot 10^{-5}\text{ Oe}^{-1} \quad (2)$$

We also observed (Fig. 3) a slight shift of the oscillation phase, i.e., a simultaneous shift of both maxima, when the impurity concentration was altered. This shift is reflected in the results shown in Fig. 4.

2. We shall now return to the plots in Fig. 2, showing another noteworthy feature, which is a steep rise of the current. A comparison of the curves indicated that in fields below 30 kOe a sixfold increase in the rate of excitation increased the photocurrent by a factor of 200–300. When the rate of excitation was kept constant at $S=6 \times 10^{17}\text{ cm}^{-2} \cdot \text{sec}^{-1}$, a reduction in the field from 30 to

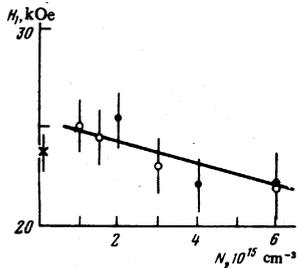


FIG. 4. Dependence of the position of the first photocurrent oscillation extremum on the impurity concentration N : \circ Ga; \bullet Sb; \times) position of the corresponding extremum in measurements of the optical characteristics of pure germanium. [3–5]

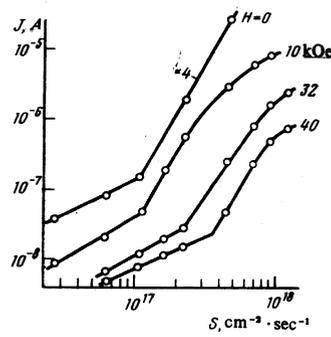


FIG. 5. Dependences of the photocurrent on the rate of excitation in various magnetic fields H ; $N_{\text{Ga}}=6 \times 10^{15}\text{ cm}^{-3}$, $T=1.6^\circ\text{K}$. The curve $H=0$ was recorded in an electric field $E=0.5\text{ V/cm}$ and the other curves in $E=2\text{ V/cm}$.

25 kOe also increased the photocurrent by almost two orders of magnitude. This increase was revealed more clearly by plotting the dependences $J(S)$ for fixed values of the field (Fig. 5).

A steep rise of the current on increase of the rate of excitation was first observed by Asnin *et al.* [16,17] and these observations were essentially the starting point of intensive investigations of the exciton condensation. Somewhat later an abrupt rise of the current was also reported by Gurnee *et al.* [18] In contrast to these earlier investigations, [16–18] our experiments were carried out in a magnetic field and on germanium samples with much higher impurity concentrations.

Our experimental results on the steep rise of the current can be summarized as follows.

a) Under steady-state excitation conditions in the investigated dopant concentration range an increase in this concentration reduced the critical excitation rate S_{cr} from which the current rose steeply. According to Fig. 6, this rate decreased by almost an order of magnitude when the impurity concentration was increased from $N=1.5 \times 10^{15}\text{ cm}^{-3}$ to $N=15 \times 10^{15}\text{ cm}^{-3}$. In the case of samples with $N=1 \times 10^{15}\text{ cm}^{-3}$ we were unable to reach the critical rate of excitation.

b) A further increase in the rate of excitation S made the rise of the current less abrupt so that the whole curve plotted on a logarithmic scale assumed the form of a step (current discontinuity). The shape of the curve was practically the same for samples whose surface areas differed by a factor exceeding 10. This demonstrated that the appearance of the flat fat part in the dependence $J(S)$ at high values of S was clearly not due to the lattice heating.

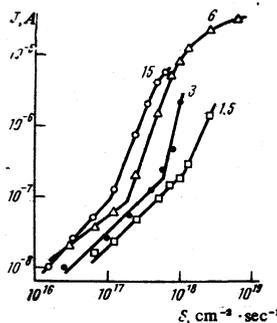


FIG. 6. Dependences of the photocurrent in a field $H=21\text{ kOe}$ on the rate of excitation S of germanium samples with different gallium concentrations; $T=1.6^\circ\text{K}$. The values of N_{Ga} are given (in units of 10^{15} cm^{-3}) alongside the curves.

c) The photocurrent curves depended strongly on the applied magnetic field (Fig. 5). An increase in this field increased the value of S_{cr} and made the step wider. In fields greater than 40 kOe the curve was no longer a step.

d) There was a correlation between the rise of the current and oscillations. The oscillation amplitude was highest at those values of S at which the photocurrent rose most rapidly: $S \approx (2-3)S_{cr}$.

DISCUSSION

All these experimental observations can be explained most logically and consistently and in a unified manner by adopting the following model.

The presence of impurities hinders the diffusion of electron-hole drops and increases their concentration in the surface layer. At some critical excitation rate S_{cr} a system of drops exhibits percolation, i.e., coalescence of drops produces conducting channels which pass entirely through the condensate; the photocurrent then rises steeply. The value of S_{cr} is a function of the magnetic field and this function has monotonic and oscillatory components. The magnetic field dependence $S_{cr}(H)$ is responsible for the features of the $J(H)$ curves observed for fixed values of $S \geq S_{cr}$.

We shall now discuss in turn all the assumptions of our model.

We have mentioned earlier experiments^[14,15] confirming a reduction in the drop mobility in doped germanium. This reduction occurs probably because the thickness of the phase boundary is comparable with the impurity dimensions and the motion of a drop gives rise to a macroscopic resistance force F , proportional to the cross-sectional area of the drop, its velocity v , and impurity concentration N : $F \propto r^2 v N$.

The interaction of this boundary with an impurity may occur because in the region occupied by the condensate the impurities are ionized since the free-carrier density in the condensate in germanium is approximately the same as the critical density for the Mott transition (see, for example, Alexander and Holcomb^[19]) and outside the condensate the impurities are mainly neutral.

The effectiveness of this reduction in the mobility is demonstrated by a comparison of the following quantities. In the case of pure germanium the fractional volume of the crystal occupied by the condensate is $k = N_d(4\pi r^3/3) \approx 0.01$, where N_d is the drop concentration.^[2,11] However, percolation in a system of conducting spheres located at random sites occurs for $k_{cr} \approx 0.35$.^[20] Even when we allow for the fact that the impurity concentration $N \lesssim 10^{16} \text{ cm}^{-3}$ does not reduce significantly the carrier lifetime in the drops,^[2,15] such a very high value of k near the surface at the excitation rates used in our experiments is possible only if k decreases steeply in the interior of the sample. The simplest assumption to make is that the number of particles bound into drops decreases exponentially with depth: $k = k_0 \exp(-x/L)$. This assumption is partly supported by the theoretical calculations and experimental results^[13]

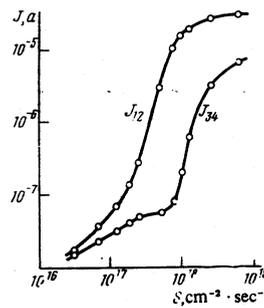


FIG. 7. Dependences of the photocurrent on the illuminated and dark sides of a sample on the excitation rate. The sample was $d = 0.1 \text{ mm}$ thick; $N_{Ga} = 6 \times 10^{15} \text{ cm}^{-3}$, $T = 1.6 \text{ }^\circ\text{K}$, $H = 21 \text{ kOe}$, $E = 3 \text{ V/cm}$.

relating to pure germanium. The total volume of the condensate per unit excited surface area $V = k_0 L$ is proportional to the excitation rate S . Therefore, if we assume that the condition $k_0 = k_{cr}$ is obeyed at $S = S_{cr}$, the results in Fig. 6 can be used to find the dependence of L on N : in the investigated range $10^{15} \text{ cm}^{-3} < N < 10^{16} \text{ cm}^{-3}$ we may assume that $L \propto N^{-1}$.

This hypothesis of a strongly inhomogeneous distribution of the condensate with depth was checked by an experiment on a thin sample. The results (Fig. 7) indicated that percolation between the two lower contacts did indeed occur when the excitation rate was high.

It would seem that at sufficiently high excitation rates the J_{12} and J_{34} curves should coincide. However, the J_{34} curve shows the tendency to saturation at lower currents than J_{12} . One of the possible explanations was that the resistances of the contact regions were different. This could arise as follows: because of rapid recombination, the condensate could not exist in the immediate vicinity of macroscopic defects such as the surface of a sample or the contact with the metal. The resultant gap would be small because of the low drop mobility. Nevertheless, it would give rise to some additional resistance which would naturally decrease strongly on illumination of the contact.

In view of the low mobility of drops in doped germanium, the surface recombination is in a sense a secondary effect. The situation is different in pure germanium, where the repulsion forces give rise to a strong flux of drops traveling toward the surface. In the case of a small sample, this should affect the average concentration of drops in the sample. In an experiment on a sample of the same dimensions as that used to obtain the curves shown in Fig. 7, but made of pure germanium, we were unable to induce a steep rise of the current or oscillations.

We shall now consider the influence of a magnetic field. This field can generally alter the shape and size of the drops. (It is unlikely that a magnetic field affects the drop concentration, so that we shall not discuss this possibility.) Flattening of drops in a magnetic field has been observed only for large drops in deformed germanium^[21,22] and clearly an increase in the number of impurities will tend to enhance such flattening.^[22] It is not clear whether the same effect occurs also in undeformed germanium in the case of small drops. However, according to the percolation theory^[23] a change in

the shape of the conducting regions without a change in their volume has practically no effect on the critical fractional volume k_{cr} occupied by the condensate. Therefore, if there is a change in the drop shape in the field, it should not affect the percolation process.

The drop size is closely related to the condensate density $n(H)$. If we ignore the phonon wind, the rate of arrival of excitons in a drop $G \propto r^2$ is independent of n , but the rate of recombination is $R \propto r^3 n(A + Bn + Cn^2)$, where the coefficients A , B , and C are governed by the cross sections of the impurity, radiative, and Auger recombination channels, and the expression in parentheses is the reciprocal of the carrier lifetime in a drop:

$$\tau = (A + Bn + Cn^2)^{-1}. \quad (3)$$

If, for example, the main recombination channel is the Auger process,^[2] then only the term Cn^2 is important in Eq. (3) and it follows from $G = R$ that the drop radius is then

$$r \propto n^{-3}. \quad (4)$$

Thus, it follows from the above discussion that the observed oscillations may be due to the field dependence of the equilibrium value of the condensate density $n(H)$. The alternative is the Shubnikov-de Haas effect in the condensate, i. e., oscillations of the transverse magnetoresistance under conditions such that the photocurrent between the contacts flows through the regions occupied by the condensate. The main argument in support of this hypothesis is that the oscillation amplitude is always maximal in the initial part of the photocurrent growth, when the percolation process just begins; further increase of the excitation rate S reduces the oscillation amplitude and this occurs irrespective of the impurity concentration N , i. e., irrespective of the absolute value of S (this means that a reduction in the oscillation amplitude cannot easily be explained by the overheating of the electron system). Moreover, the oscillation amplitude is generally too high for the Shubnikov-de Haas effect and also it does not vary sufficiently regularly with the serial number of the oscillations. However, if we consider oscillations of the radius r , we find that because of the dependence $S_{cr}(H)$ (Fig. 5), the optimal conditions for the observation of the oscillations can be realized only in a certain range of fields H . This explains the observation why sometimes the recorded relative amplitude of the first maximum is greater than in Figs. 2 and 3 and the subsequent maxima are not observed at all.

Oscillations of the condensate density have been considered on several occasions.^[24,25] Direct measurements^[5] have yielded the amplitude of the oscillations of the density $n(H)$ and also the amplitude of the oscillations of the carrier lifetime $\tau(H)$ in a drop. If the relative amplitude is taken to be the change of a given quantity in a field $H = 18$ kOe, we find from the graphs given by Betzler *et al.*^[5] that

$$A_r = \left. \frac{n(H) - n(0)}{n(0)} \right|_{18 \text{ kOe}} \approx 0.05, \quad A_\tau = \left. \frac{\tau(H) - \tau(0)}{\tau(0)} \right|_{18 \text{ kOe}} \approx -0.11.$$

The relationship $A_r \approx -2A_\tau$ confirms that the main recombination channel is the Auger process [see Eq. (3)], so that on the basis of Eq. (4) we can expect the corresponding amplitude of the oscillations of the drop radius to be $A_r \approx -0.15$. (This corresponds to a 40% reduction in the drop volume and in the functional volume k .) Since the conductivity in a system of conducting spheres at random sites depends exponentially on their radius^[20]

$$\sigma \propto \exp(-r_{cr}/r), \quad r_{cr} = (3k_{cr}/4\pi N_d)^{1/3}, \quad (5)$$

it follows that

$$A_\sigma = \Delta\sigma/\sigma \approx (r/r_{cr})A_r, \quad (6)$$

and in the range $r \approx r_{cr}$ the relative change in the conductivity is $A_\sigma \approx A_r$.

In the investigated range of magnetic fields the density $n(H)$ oscillates because the condition (1) is satisfied by electrons. Since in a field $H \parallel [100]$ all the electron valleys are equivalent, the condition (1) is satisfied simultaneously by all the valleys. It follows from Eq. (1) and from the measured period (2) that the density of electron-hole pairs in the condensate is $n_0 = 2.4 \times 10^{17} \text{ cm}^{-3}$ in good agreement with the generally adopted value.^[2] However, it should be stressed that two assumptions are made in deducing n_0 from Eq. (2). First, it is assumed that the anisotropy of the electron spectrum of the condensate is the same as that of the spectrum of germanium itself, i. e., that the ratio of the cyclotron and density-of-states masses m_c/m remains constant. Second, since only the first oscillations are observed experimentally, it is assumed that $A_n \ll 1$, so that the measured value is $n_0 \approx n(0)$. These comments apply also to other determinations of the value of n_0 from the oscillation period.^[3-6]

The fact that in the impurity concentration range $N < 10^{16} \text{ cm}^{-3}$ the value of n_0 is independent of N is also in agreement with the results of other experiments.^[2,26] However, we cannot propose any convincing explanation of the slight changes in the oscillation phase which occur when N is varied (Figs. 3 and 4). We shall confine ourselves to general comments. There are two logical possibilities. Either the impurities do not affect the value of $n(0)$ but influence the dependence $n(H)$ and, therefore, the value of ε_F in Eq. (1), or the maxima of the dependences $J(H^{-1})$ occur at fields somewhat different from those at which the condition (1) is satisfied; for example, the shift may be due to the fact that changes in n alter the relative cross sections of the various recombination channels and this may affect the carrier mobility in the condensate for the phonon wind intensity. Then, naturally, such a shift may depend on N .

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Translated by A. Tybulewicz