Investigation of electron-hole drops in germanium in a heating microwave field. Nonstationarity of nucleation

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The condensation of excitons in germanium under conditions of heating of the free carriers in a microwave field was investigated. It is observed that only the excitons take part in the nucleation of the drops, and that the free electrons and holes hinder the formation of the nucleus by heating it. This effect is attributed to the nonisothermy of the nucleation process. It is shown that the produced drops with radii exceeding 10^{-5} cm trap excitons and free carriers at practically the same rate. An increase of the carrier temperature in the microwave field leads to an increase in the volume of the liquid phase if the carrier density is low. At an appreciable electron density, intense phonon fluxes are produced and carry the drops outside the optical-excitation region. The action of both factors—the fluxes of the phonons and of the free carriers, which appear when the electrons are heating—causes the condensation process to proceed jumpwise in a heating field and increases greatly the width of the hysteresis. The observed effects are used to measure the dependence of the rate of the condensation process on the degree of supersaturation. The nucleation was observed to be nonstationary when the supersaturation of the exciton gas was maintained for a short time interval.

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A hysteresis in the onset and vanishing of electron-hole drops (EHD) in germanium was observed in Ref. 1. These experiments have demonstrated convincingly the similarity between the condensation of excitons into EHD and the well known vapor-liquid transitions.[2] An investigation of the condensation of excitons in germanium makes it possible to study by relatively simple means the irregularities of a first-order phase transition; owing to the high degree of perfection and purity of the germanium crystals, one can expect the nucleation of the drops to be "homogeneous," i.e., the EHD nuclei result from fluctuations in the exciton gas, and are not produced on condensation centers such as impurities.

We report here the results of an investigation of condensation in a heating microwave field. It has turned out that application of a heating field to a system subject to hysteresis of the onset and vanishing of the condensed phase offers new possibilities of studying the kinetics of the nucleation, growth, and motion of the EHD. The point is that a heating microwave field acts in a complicated manner on a system of electrons, free carriers, and EHD. The heating of the electrons decreases the concentration of the excitons, while the density and temperature of the free carriers increase. This leads, firstly, to a change in the nucleation conditions, and secondly to a change in the flux of matter from the gas phase into the drops and consequently to a change of the drop size. Finally, owing to the heating of the free carriers, intense fluxes of nonequilibrium phonons are produced and can carry the drops out of the excitation region.

Our investigation made it possible to assess experimentally the role of the free electrons and holes during various stages of drop formation, and also in processes connected with the motion of the drop.

In Sec. 2 we report the methodological peculiarities of the experiments, while in Sec. 3 we discuss the influence of a heating field on the change of the density of the excitons and of the free carriers; the same section considers the kinetics of the relaxation of the exciton-electron plasma when the microwave field is rapidly turned on or off.

Section 4 deals with the influence of carrier heating on a system containing free carriers, excitons, and EHD, as well as the effect of EHD motion under the influence of the phonon fluxes emitted by the hot electrons.

Section 5 deals with the slowing down, observed by us, of the condensation process when the free-carrier density is increased; this effect is due to the nonisothermy of the nucleation. We describe also a unique effect of jumplike condensation which occurs when the EHD nuclei are dragged by a stream of nonequilibrium phonons.

The observed decrease of the condensation rate with increasing free-carrier density is used in Sec. 6 to devise a new method of measuring the rate of nucleation. The dependence of the rate of condensation on the degree of supersaturation is measured and it is observed that this process is nonstationary when the degree of supersaturation is rapidly varied.

1. HYSTERESIS OF EXCITON CONDENSATION IN GERMANIUM

Condensation of excitons into EHD is a first-order phase transition of the gas-liquid type. At temperatures below critical (T_c) there is consequently a threshold exciton-gas density (n_{ex}) at which the drops are precipitated. If we disregard the kinetics of the transitions, then the condensation threshold corresponds to an exciton density equal to the saturated vapor density $n_{T\infty}$ over a flat surface of the liquid. However, the new phase in the gas is produced in the form of small droplets that are unstable at $n_{ex} \approx n_{T\infty}$, so that we must consider the kinetics of the nucleation of the new phase.

As shown by the classical theory of condensation,[2]

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the appearance of a liquid phase takes place at $n_{ex} > n_{T\infty}$; the presence of a threshold density n_{ex} is determined by the fact that the rate of the condensation process, i.e., the number of viable nucleation droplets produced in a supersaturated vapor per unit time and per unit volume, is an extremely strong function of the degree of supersaturation $n_{ex}/n_{T\infty}$:

$$\frac{dN}{dt} = A \exp\left[-\frac{\Lambda}{\ln^2(n_{ex}/n_{T\infty})}\right], \quad \Lambda = \frac{16\pi\sigma^3}{3n_0^2(kT)^3}, \tag{1}$$

where σ and n_0 are the surface tension and density of the liquid phase, and $\Lambda \gtrsim 10^2$. It is seen from (1) that when the degree of supersaturation $n_{ex}/n_{T\infty}$ is increased from 4 to 8, i.e., by a factor of two, the value of dN/dp increases by 5–10 orders. Obviously, the number of drops produced in the system near the condensation threshold during the time Δt in the volume V is determined by the relation

$$\Delta N = \frac{dN}{dt} V \Delta t$$

and, by virtue of (1), ΔN increases abruptly when the degree of supersaturation is increased. Thus, the experimentally observed threshold of the appearance of a liquid phase is determined by that degree of supersaturation at which the ratio of the condensation process becomes sufficient for an observable number of drops to appear in the system during the observation time:

$$N_{min} = (dN/dt)_{min} V \Delta t. \tag{2}$$

If the rate of generation of the nonequilibrium carriers G is increased sufficiently slowly, then condensation begins at $G_+ = \varkappa_0 n_{T\infty}/\tau_{ex}$ (τ_{ex} is the exciton lifetime), when the exciton concentration $n_{ex} = \varkappa_0 n_{T\infty}$ ($\varkappa_0 \approx 7$ at $T = 2$ K) is such that the condensation process reaches a rate of $\sim 10^5$ drops/cm^3 sec, corresponding to ~100 drops/sec in the investigated volume, which is the experimentally observable threshold of recombination radiation of the EHD. The produced nuclei grow to form macroscopic drops rapidly (within a time $\sim 10^{-4}$ sec shorter than the time of appearance of each succeeding nucleus, $\sim 10^{-2}$ sec). The radius of the drops is determined from the condition

$$n_{ex} = \varkappa_0 n_{T\infty} \approx R_m n_0 / 3 v_T \tau_0$$

(v_T is the thermal velocity of the excitons and τ_0 is the EHD lifetime), and the exciton density is stabilized by the process of appearance of new nuclei, so that the rate of the condensation process depends exceedingly strongly on the degree of supersaturation.

With increasing excitation level, at $G > G_+$, the number of drops of constant radius R_m in the sample increases. On the other hand, if the excitation level is decreased, starting with $G = G_{max} \gg G_+$, then the density of the excitons and accordingly the radius of the drops will decrease until the EHD radius decreases to a value

$$R_{min} \approx \frac{1}{n_0}\left(\frac{6\sigma\tau_0 v_T n_T}{kT}\right)^{1/2}$$

($R_{min} \sim 8 \cdot 10^{-6}$ cm for $T = 1.6$ K), when the droplets evaporate rapidly. This corresponds to a descending threshold $G_- \approx n_{ex}(R_{min})/\tau_{ex}$. Thus, hysteresis appears in the condensation of excitons and manifests itself distinctly in the dependence of the intensity of the EHD luminescence on the excitation level.[1,3-6] In contrast to the concepts presented above, however, it was observed in experiment[3,6,7] that the exciton density and the drop radius continue to increase at $G > G_+$.

The EHD nucleation process differs substantially from the usually considered condensation process in that the produced nucleus traps both excitons and free carriers—electrons and holes—but only excitons are evaporated mainly from the surface of the nucleus. This superheats the nucleus and decreases the condensation rate, leading to the nonisothermy of the condensation process.[8]

The density of the free carriers in a condensing exciton gas is usually quite appreciable, but this circumstance had been ignored previously. It will be shown below that the presence of free carriers leads to a substantial change in the condensation process in germanium, making it possible, on the one hand, to explain in natural fashion a number of hitherto not understood experimental results, and on the other provides a convenient method studying the nucleation process itself.

2. EXPERIMENTAL PROCEDURE

The experimental setup is shown in Fig. 1. The excitation source (He-Ne Laser) was capable of both volume ($\lambda = 1.52$ μm) and surface excitation of the germanium samples. In the main we investigated samples of pure n Ge ($N_d \approx 3 \cdot 10^{10}$ cm^{-3}), kindly supplied by E. E. Haller (Berkeley Laboratory, USA). We investigated the dependence of the intensity I of the EHD recombination radiaeion (709 meV line) and the intensity of the excitons I_{ex} (714 meV) on the generation rate G at various levels of microwave power applied to the sample.

The germanium sample in the form of a plate $4 \times 6 \times (0.2–0.5)$ mm^3 was placed in a short-circuited segment of an 8-mm waveguide and illuminated by unmodulated laser light through an opening in the narrow wall. A radiation modulator rotating at a frequency 120 Hz was placed in front of the entrance slit of the monochromator. The radiation receiver was a cooled Ge:Cu photoresistor, the signal from which was amplified, synchronously detected, and fed to the Y coordinate of the recorder. The signal to the X coordinate was proportional to the intensity of the exciting light. The excitation intensity was varied with a slowly rotating polaroid.

The experiments were performed in the temperature range of 1.6–4.2K. To ensure continuity of the illumination of the sample at $T = 1.6$–2.1 K, when the $I(G)$ hysteresis loops was plotted, provision was made in the construction of the glass cryostat to remove the nitrogen jacket (it is impossible to obtain these plots by passing light through boiling nitrogen).

The "ascending" branch of $I(G)$ was plotted with G smoothly increasing; to plot the "descending" branch it was convenient to apply to the sample additional illumination in the form of infrequently repeated pulses

from GaAs laser, of duration ~10^{-6} sec and frequency ~1 sec^{-1}. Illumination by such a light pulse establishes in the EHD system an absolutely stable state, in which the concentration of the excitons and the radius of the drops were the smallest possible.[5] The microwave generator had a maximum power output 200 mW; this power could be modulated with a mechanical attenuator or with a p-i-n switch (in the latter case it was possible to turn the microwave circuit on and off with frequency up to 2 MHz with a pulse rise time ~20 nsec). A homodyne microwave spectrometer was used for the microwave measurements; the absorption or dispersion signal was separated with a phase shifter in the reference arm of the microwave circuit. In some experiments, namely in investigations of cyclotron resonances, the waveguide with the sample was placed in a superconducting solenoid with maximum magnetic field intensity $H \sim 5$ kOe.

3. CHANGE OF EXCITON AND FREE-CARRIER DENSITIES IN A HEATING MICROWAVE FIELD. RELAXATION KINETICS

If the semiconductor is exposed to light that ensures a generation rate G, then the exciton density n_{ex} and the free-carrier density n are determined by the known kinetic equations

$$\frac{\partial n}{\partial t}=G-\alpha n^2-\frac{n}{\tau}, \quad \frac{\partial n_{ex}}{\partial t}=\alpha n^2-\frac{n_{ex}}{\tau_{ex}}, \tag{3}$$

where α is the coefficient of binding the free carriers into excitons, while τ and τ_{ex} are the lifetimes of the electrons and excitons.

In such semiconductors as germanium and silicon, at $T \leq 4.2$ K the free carriers are not in a state of thermal quasi-equilibrium with the excitons, therefore in Eqs. (3) the terms describing the thermal spike are omitted. Assuming that the lifetimes of the electrons and excitons are close, $\tau_{ex} \approx \tau$, we easily obtain the relations

$$\alpha n^2-n_{ex}/\tau=0, \quad n+n_{ex}=G\tau, \tag{4}$$

$$n_{ex}=G\tau-\frac{1}{2\alpha\tau}[(1+4G\alpha\tau^2)^{1/2}-1]. \tag{5}$$

The influence of the heating field, within the framework of (3)–(5), reduces to a change in the value of α; if we assume a binding cross section σ_b that depends on the carrier temperature T_e like T_e^{-2}, then $\alpha=\sigma_b v \propto T_e^{-3/2}$. In a strong microwave field, when impact ionization of the exciton is possible, the first relation of (4) takes the form

$$\alpha n^2-\gamma_i n n_{ex}-n_{ex}/\tau=0, \tag{6}$$

where γ_i is the coefficient of impact ionization of the excitons. Assuming, by analogy with the ionization of the shallow impurity that

$$\gamma_i \sim \sigma_0 v \exp(-\varepsilon_{ex}/kT_e),$$

where $\sigma_0 \sim \pi a_0^2 \sim 10^{-11}$ cm^2 (a_0 is the Bohr radius of the exciton), $v=(2kT_e/m^*)^{1/2}$, and ε_{ex} is the exciton binding energy, we find that

$$\gamma_i(T_e) \approx 10^{-5} T_e^{1/2} \exp(-\varepsilon_{ex}/kT_e). \tag{7}$$

Taking (4) and (6) into account we get

$$n_{ex}(T_e)=G\tau-\frac{1}{2\tau(\alpha+\gamma_i)}[(\gamma_i G\tau^2-1)+((1+\gamma_i G\tau^2)^2+4\alpha G\tau^2)^{1/2}]. \tag{8}$$

This expression describes the decrease of the exciton density when the carriers become heated; it is seen that in the case of impact ionization the exciton density decreases smoothly with increasing electron temperature.[9]

We shall be interested from now on in the density range $n \sim 10^{10}$–10^{12} cm^{-3} and the temperature region $T_e \lesssim 20$ K. The temperature of the hot carriers (assumed to have a Maxwellian distribution) is given in the case of small heating by the relation[10]

$$\frac{e^2E^2\nu_i}{m^*(\omega^2+\nu_i^2)}=\frac{T_e-T_0}{T_0}\frac{ms^2}{\nu_{ph}}, \tag{9}$$

where ω is the frequency of the microwave field, $\nu_i=\nu_{eh}+\nu_{ph}$ is the frequency of the collision of the carriers with one another and with the acoustic phonons ($\omega \gg \nu_i$). At $E \approx 20$ V/cm we have $T_0=2$ K and expression (9) yields $T_e \approx 10$ K; at the maximum microwave power $P_{microw} \approx 200$ mW we have $T_e=30$ K in the antinode of the electric field.

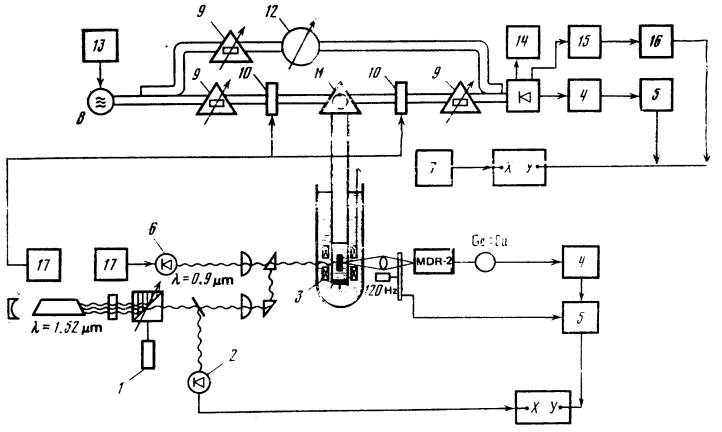

FIG. 1. Diagram of experimental setup: 1—attenuator of excitation intensity, 2—photodiode, 3—cryostat, 4—narrow-band amplifier, 5—synchronous detector, 6—pulsed GaAs laser, 7—solenoid supply block, 8—klystron, 9—microwave power attenuator, 10—microwave power switch using a p-i-n structure, 11—ferrite circulator, 12—phase shifter, 13—klystron power supply, 14—microwave power indicator, 15—broadband amplifier, 16—stroboscopic oscilloscope, 17—pulsed power supply.

Figure 2 shows a plot of $n_{ex}(T_e)$ calculated from Eq. (8). It is seen that at $T_e \lesssim 10$ K the exciton density changes insignificantly, and consequently the role of impact ionization is small. The same figure shows the experimental dependence of the extinction of the exciton radiation intensity in a microwave field, obtained at $T_0 = 4.2$ K. It is seen that these relations are in qualitative agreement. In Sec. 4 and 5 below, where we consider the influence of the microwave field on the shift of the condensation threshold, we shall see that the microwave field intensity of interest to us corresponds to small T_e and consequently to small changes of the exciton density ($T \lesssim 10$ K, $\delta n_{ex}/n_{ex} \lesssim 0.2$).

We proceed to a discussion of the kinetics of the relaxation of the exciton density and electron density when the heating field is rapidly turned on and off. To calculate the relaxation processes we solve the equation

$$\frac{\partial n_{ex}}{\partial t} = \alpha n^2 - \frac{n_{ex}}{\tau} - \gamma_i n n_{ex}$$

under the condition $n + n_{ex} = G\tau$. The solution is of the form

$$n_{ex}(t) = G\tau - \frac{(a-c)(De^{at/\tau}-1)}{2(\alpha+\gamma_i)\tau(De^{at/\tau}+1)},$$
$$a = [(1-\gamma_i G\tau^2)^2 + 4(\alpha+\gamma_i)G\tau^2]^{1/2}, \quad c = (1-\gamma_i G\tau^2)^{-1}, \qquad (10)$$
$$D = \frac{a+2(\alpha+\gamma_i)n_0\tau+c}{a-2(\alpha+\gamma_i)n_0\tau-c}.$$

The results of the calculation are shown in Fig. 3. It is seen that when the microwave field is turned on and at $T_e = 15$ K the exciton density decreases within the time ~0.5 μsec, with account taken of the impact ionization of the excitons.

If we disregard impact ionization, the exciton density decreases more slowly, with a time constant τ_{ex} ~5 μsec. When the field is turned off, n_{ex} increases rapidly within a time ~0.3 μsec. Thus, at $T_e \approx 15$ K the characteristic relaxation time of the exciton density is ≲0.5 μsec.

4. EFFECT OF HEATING FIELD ON EHD. MOTION OF EHD UNDER THE INFLUENCE OF NONEQUILIBRIUM PHONONS

We have previously[5] observed that the action of a heating microwave field on a system of free carriers,

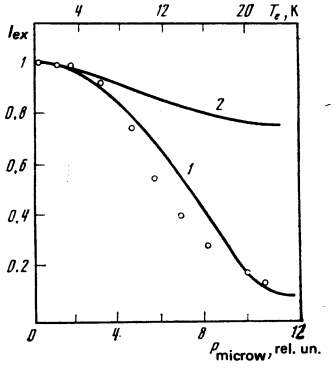

FIG. 2. Dependence of the exciton radiation intensity I_{ex} on the microwave power level, $T = 4.2$ K: 1—calculation by formula (8), 2—the same but with $\gamma_i = 0$; points—experiment.

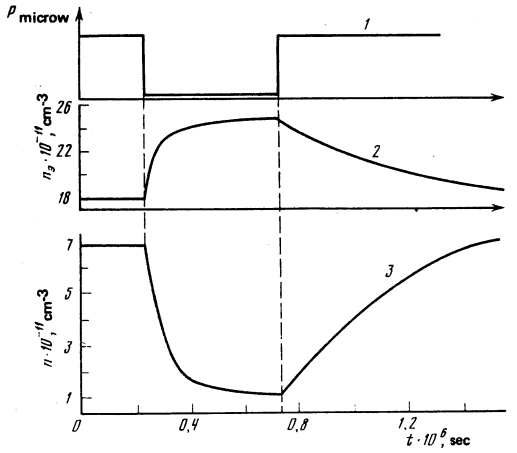

FIG. 3. Kinetics of relaxation of the exciton density (2) and of the electron density (3), calculated by formula (8); $\gamma_i = 1.33 \times 10^{-6}$ cm^3 sec^{-1}, $\alpha = 1.52 \times 10^{-6}$ cm^3 sec^{-1}, $T_0 = 2$ K, $T_e = 15$ K.

excitons, and drops depends essentially on the prior history of the system, i.e., on whether the drops are produced prior to the application of the field or whether a certain value of the heating field is set at $G = 0$ and the pumping G is then increased. In the present section we discuss a situation wherein the drops are produced in the absence of the microwave field. As seen from Fig. 4a, in this case, when the microwave power is increased, the recombination radiation of the drops contained in the crystal increases in a wide range of pumps. At large G, however, the radiation of the drops is extinguished. Figure 4b shows the behavior of the luminescence at various points of the $I(G)$ curves following an increase and subsequent decrease of the microwave power. It is seen that the curve 3 is reversible; at larger G the curves are not reversible and form a hysteresis loop that is larger the larger G.

To explain these phenomena it must be taken into account that free carriers are present in the gas phase. The density can be estimated by using the relations (4) and (5). Assuming $\alpha \approx 2 \times 10^{-5}$ cm^3/sec, $\tau_{ex} = \tau = 5 \times 10^{-6}$ sec, $G \approx 10^{17}$ cm^{-3} sec^{-1} ($G \approx G_+$ at $T = 1.6$ K), we obtain $n_{ex} \approx 4 \times 10^{11}$ cm^{-3} and $n \approx 7 \times 10^{10}$ cm^{-3}. At $G > G_+$ the

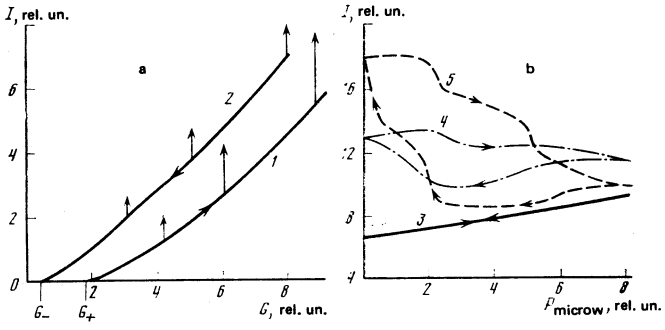

FIG. 4. Influence of heating field on the intensity of the EHD radiation; $T = 1.6$ K. a) Plot of $I(G)$ at low excitation intensity; 1—ascending branch of $I(G)$, 2—descending branch. The arrows show the increase of the luminescence with increasing microwave power up to 200 mW. b) Dependence of I on the microwave power: curve 3 was plotted at $G = 6G_+$, with curves 4 and 5 corresponding to larger G.

electron density can also be determined from (3) and $n \sim G^{1/2}$, since the rate of electron capture by the drops at $G \lesssim 10G_+$ is small compared with n/τ.

We consider now the influence of carrier heater on the intensity of the EHD radiation at $G \gtrsim G_+$. The increase of the intensity of the EHD emission in the heating field is reversible at small $G \gtrsim G_+$. This means that the increase of the radiation signal is due to the increase of the radius of the drops, and not their number. The calculation of the change of the radiation of the drop when the carriers are heated, under the assumption that all the excitons are ionized, i.e., that the heating is large (see Sec. 3), and that the drops are fed by hot electrons and holes having a velocity v_e, leads to the expression

$$\Delta I = I^E - I^0 \sim R(1 - v_T/v_e), \tag{11}$$

i.e., at an initial radiation intensity I_0 the radiation intensity in the field I^E is larger the larger the drop radius R and the carrier velocity v_e.

Thus, in a heating field, when the velocity of the carriers that feed the drops increases, the drop radius increases and, in accord with experiment (see Fig. 4a), the increase of the radiation intensity depends on the initial radius of the drop (cf. curves 1, 2, and 3).

This is the situation at $G \gtrsim G_+$: at higher excitation levels the action of the field is subject to hysteresis; with increasing G the hysteresis loop becomes broader and the radiation signal may not increase at all (curves 4, 5). Thus, the decrease of the EHD radiation intensity is irreversible, which suggests that the number of drops decreases under these conditions. We note that when the temperature is increased the extinction process becomes favored and at $T \geqslant 2$ K hysteresis quenching of the radiation by the field is observed in the entire range of G. The use of surface excitation (light with $\lambda = 0.63$ μm) or of sharper focusing also facilitates the quenching process. On the contrary, the more uniform and the larger the excitation region, the weaker the quenching effect.

It seems to us that the observed quenching is due to the departure of the drops from the excitation region under the influence of the phonon streams produced when the carriers are heated, and consequently to loss of the drops—to a decrease of their number. No new drops can be produced here, inasmuch as the nucleation threshold shifts in a heating field towards larger G (see Sec. 5).

We estimate now the force f exerted on a pair of particles in a drop by the phonon flux[11,12] produced when the carriers are heated:

$$f = \hbar q N_{ph} s \sigma_{ph};$$

here σ_{ph} is the cross section for phonon absorption by an electron in the drop, $n_{ph} \approx 10^{-5}$ cm^2,[11] $\hbar q$ is the characteristic momentum of the phonons emitted by the hot carriers, $\hbar q = (2m^* k T_e)^{1/2}$, N_{ph} is the concentration of the nonequilibrium phonons, and s is the speed of sound. Recognizing that

$$N_{ph} = \frac{dN_{ph}}{dt}\tau_{ph} = \frac{n}{\tau_{eph}}\tau_{ph},$$

where τ_{eph}^{-1} is the frequency of the electron-phonon interactions and τ_{ph} is the lifetime of the long-wave phonons, determined at low temperatures by their departure from the crystal, ($\tau_{ph} \sim L/s$ and L is the dimension of the sample), we have

$$j = \sigma_{ph} n \frac{(2mkT_e)^{1/2}}{\tau_{eph}} L.$$

At $n \approx 10^{11}$ cm^{-3}, $T_e = 20$ K, and τ_{eph} 5×10^{-11} sec, the characteristic value of the force is ~1 meV/cm.

We have shown earlier[13] that the drop is kept in place by the crystal defects[14] and can be set in motion by a force that exceeds a certain critical value that depends on the radius of the drop. Therefore when the density and temperature of the electrons increases with increasing field, as does the radius of the drops, the force may turn out to be sufficient to detach the drop from the defect and to take it out of the excitation region. The larger G, the simpler it is to realize this situation, inasmuch as[1)] $n \propto G^{1/2}$.

5. INFLUENCE OF FREE CARRIERS ON THE EHD NUCLEATION

A. We turn to experiments in which, at a certain microwave power, the generation rate G increases smoothly. Figure 5 shows the hysteresis curves plotted at certain values of the microwave power. We note the main features of these plots: the threshold G_+ of the appearance of the liquid phase in the microwave field increases, whereas the disappearance threshold G_- does not shift when the carriers are heated. As a result, the hysteresis width G_+/G_- in pure germanium samples increases substantially; in some range of the microwave power, the liquid phase appears jumpwise (curves 6–8).

As seen from Fig. 5, the descending branch (curve 4) has a small slope. It will be shown below that the reason is that at $G = G_{max}$ a much lower drop concentration is produced in the heating field. For a more definite determination of the threshold G_-, the descending branch was plotted by applying single pulses from a GaAs laser (see above). In this measuring method, the threshold of G_- was determined distinctly at all values of the microwave power and did not shift in prac-

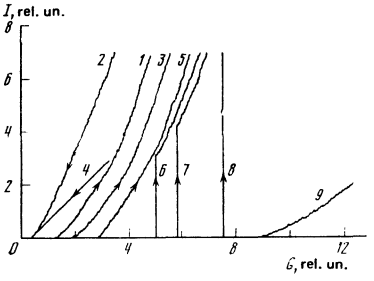

FIG. 5. Influence of heating microwave field on EHD nucleation, $T = 1.6$ K: 1,2—ascending and descending branches of EHD luminescence intensity in the absence of a microwave field. Ascending branches at the following values of P microwave (relative units): 3—0.15; 5—0.2; 6—0.62; 7—0.8; 8—0.92; 9—1.23; 4—descending branch at $P = 1.23$. Curve 8 was obtained at $P_{microw} = 50$ mW (0.25 P_{max}).

tice when the carriers were heated. The quantity G_- characterizes a state of the system wherein the drops reach a radius $R=R_{min}$ and the excitons and electrons have a total density corresponding to this radius. The constancy of G_- following application of a microwave field allows us to state that when the ratio of the electron and exciton densities changes their combined density $n+n_{ex}$ remains unchanged, and consequently the lifetimes of the excitons and electrons are close to each other. A similar conclusion that τ_{ex} and τ are close follows also from the fact described above that the intensity of the EHD radiation increases when the carriers are heated.

We proceed now to a discussion of the nature of the shift of this threshold G_+ in a heating microwave field. Inasmuch as in the course of heating the total concentration $n+n_{ex}$ of the generated pairs remains practically unchanged, an increase of the threshold is evidence that the condensation sets in at a higher density of the nonequilibrium pairs. This effect can be connected with the fact that the free carriers do not participate in the nucleation, and the nucleus is made up only of excitons[5]; in this model the condensation threshold G_+ corresponds to attainment of one and the same exciton density n_{ex}^*, independent of the value of the field. However, as noted above, the characteristic electron temperature T_e at which a shift of the condensation threshold was observed did not exceed ~10–15 K. At this temperature, as seen from Fig. 2, the decrease of the exciton density is negligible. It must therefore be assumed that the condensation threshold $G_+(E)$ in a heating field corresponds to an exciton density higher than n_{ex}^*, i.e., the condensation sets in at an excess supersaturation. We have demonstrated this earlier[8] by direct measurements.

The excess supersaturation of the exciton gas is due to the increase of the density of the free carriers in the heating field. This effect of slowing down the nucleation process with increasing flux of electrons and holes through the nucleus is due to the non-isothermy of the nucleation process, namely, to the superheat of the produced nucleus relative to the surrounding gas, owing to the binding energy released when the particles are trapped to form a drop. The capture of an exciton is accompanied by release of a binding energy $\varphi_0 \sim 2$ meV, and in the case of trapping of an electron and a hole the energy released is much higher, $\varepsilon=\varphi_0+\varepsilon_{ex}\approx 6$ meV, where $\varepsilon_{ex}\approx 4$ meV is the exciton binding energy.[2)] Since the influx of the heat is determined by the particle flux to the surface of the drop, and the outflow is due to the interaction between all the particles in the volume and the acoustic phonons, the superheat phenomenon turns out to be most substantial for small nucleation drops and leads to a decrease in the rate of nucleation.[8,15]

It was shown in Ref. 8 that in a constant magnetic field that magnetizes the motion of the charged particles, when the flux of these particles per nucleus decreases, the threshold of the condensation decreases. Thus, the effect of slowing down the condensation takes place also when the density of the "cold" carriers is increased.

Under the joint action of a heating microwave field and a constant magnetic field H, the largest deceleration of the condensation is observed at values $H_c=m\dagger c\omega/e$ corresponding to conditions of cyclotron resonances of the electrons and holes. Equal shifts of the condensation thresholds are attained in this case at a microwave power much lower than at $H=0$. Consequently one can conclude that the excess supersaturation is due only to an increase in the number of free carriers in the course of heating, and not to the direct influence of the microwave field on the produced nucleus.

Figure 6 shows plots of the EHD radiation intensity at $G\approx 2G_+$ against the magnetic field at various microwave-field intensities. Curve 1 shows that in the absence of heating the radiation intensity increases because of the lowering of the condensation threshold; when the microwave field is turned on (curves 2, 3) the radiation intensity decreases at $H_i=m\dagger c\omega/e$ because of the increase of the condensation threshold, when the carriers become heated in the cyclotron field[3)] (a similar decrease of I was apparently observed also in Ref. 16).

In concluding this section we note that the phenomenon of deceleration of the condensation by the free carriers modifies substantially our notions concerning the hysteresis phenomenon. In fact, when the excitation level is increased, at $G>G_+$, the free-carrier density continues to increase, $n\propto G^{1/2}$, and consequently an ever larger exciton-gas density will be needed for new nuclei to appear on the ascending branch, i.e., the density of the excitons and the radius of the drops must increase beyond the condensation threshold.[3,6,17]

B. We discuss now the jumplike condensation in a strong microwave field. If the microwave field intensity exceeds 5–10 V/cm then, as seen from Fig. 5 (curve 6–8), the threshold G_+ increases quite strongly, and the condensation takes place jumpwise, i.e., a very large number (~10^4) of macroscopic drops is produced in the sample at a fixed value of G. The result, which is natural for an ordinary gas–liquid transition, when a single produced liquid-phase nucleus can eliminate the entire supersaturation of the gas, contradicts the main premises of the kinetics of condensation of a nonequilibrium system (see above) when the condensation threshold corresponds to a rate of nucleation such that each nucleus manages to be transformed into a macroscopic drop prior to the appearance of the next nucleus.

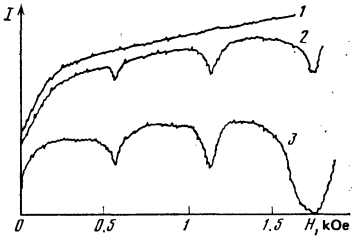

FIG. 6. Dependence of the EHD radiation intensity at $G=2G_+$ on the magnetic field for different P_{microw}: 1—10^{-3} P_{max}, 2—0.1 P_{max}, 3—0.25 P_{max}.

The jumplike growth of the drop radiation intensity means that a large number of viable nuclei is produces simultaneously in the system. This is possible if the rate of appearance of the nuclei is not limited by the growth rate of the drop.

To explain the effect of the jumplike condensation it is necessary, in our opinion, to assume that the process of nucleus formation in pure germanium crystals is homogeneous, i.e., the EHD nuclei are produced as a result of fluctuations in an exciton gas, and not on condensation centers such as impurities. This assumption does not contradict the results of Refs. 13 and 14, where it is stated that the EHD is held back by the defects, inasmuch as in the course of random wandering over the crystal a drop of radius 10^{-5} will be captured by an impurity after a very short time[4] $\tau_d=(\pi R^2 v_T N_i)^{-1}$ $\sim(10^{-5}-10^{-6})$ sec, and will subsequently be held back by the impurity, since the binding energy should amount to ~5 meV.[18]

The explanation of the jumplike condensation lies, in our opinion, to the effects of motion of the EHD, and consists in the following. When the electrons are appreciably heated, the nucleation proceeeds under conditions of the phonon dragging (see above), and therefore the EHD nuclei, being free and mobile formations, are removed from the excited region of the crystal and are annihilated before they grow to macroscopic dimensions.[5] Therefore at $G \geqslant G_*$ no condensation can be observed, and with increasing G, the supersaturation of the exciton gas continues to increase, since there is no outflow of excitons to growing drops, and consequently, the condensation rate increases drastically. The growth rate of the nucleus increases, and the time of capture of the drop by a lattice defect decreases like R^{-2}. Since these two factors arced in the same direction, the nucleation drop is captured at a certain $G=G'$ by the defect, is stopped without managing to leave the excitation region, and consequently can reach macroscopic size. Since the rate of condensation at $G=G'>G_*$ is quite large, the capture conditions are satisfied simultaneously for a large number of drops. At the same time it appears that a certain role can be played also by the fact that the flux of electrons and holes to the captured drops leads to a decrease of the concentration of the free carriers, and by the same token, on the one hand, to a decrease of the phonon dragging force, and on the other, to a weakening of the action of the free carriers on the nucleation. As a result, the conditions for retaining the drop become easier, and "avalanche deceleration" of the newly produced drops sets in.

We note that jumplike condensation is observed only in pure germanium crystals. In samples containing a large impurity density ($N_i \gtrsim 10^{12}$ cm^{-3}), the jump is never observed, and the threshold G_* increases monotonically and smoothly with increasing microwave power. This is evidence either of formation of nuclei at the impurities, or of so fast a capture of the nuclei, that their outward drift is negligible. We note also that jumps in pure germanium are observed only at $T \lesssim 1.9$ K.

6. MEASUREMENT OF THE RATE OF THE NUCLEUS-FORMATION PROCESS AND THE NONSTATIONARY EFFECT

The observed influence of the free carriers on the rate of the condensation process can be used to measure the dependence of the rate of the nucleation on the degree of supersaturation. In the experiments known to us on the kinetics of condensation of ordinary gases (water vapor, ammonia, SF_6, and others),[15,19] performed in expansion chambers in the gas jets one usually determines the temperature dependence of the supersaturation at which the condensation becomes observable.

Experiments similar in principle, aimed at observing the kinetics of exciton condensation in EHD, performed by Westervelt,[3] have shown that the kinetics of nucleation can in this case be described by the classical theory of nucleation with account taken of the finite lifetime of the particles in the drops.[3,4]

It must be emphasized that in each of the studies[3,15,19] the rate of the condensation process was fixed, since both the condensation time Δt and the threshold number N_{min} of drops were left constant. We describe here a different method of determining the condensation rate, in which one varies the time Δt allotted to condensation and one obtains the degree of supersaturation at which an observable number N_{min} of drops is produced.

The method of measuring the condensation rate is based on the already described hysteresis in the action of a heating microwave field on a system of free carriers, excitons, and EHD (Secs. 4 and 5). It was shown that in a heating field the condensation threshold increases (Fig. 5), whereas the microwave field has little effect on the drops present in the system (see Fig. 4). Assume that at $G > G_*$ and in the presence of a microwave field no drops are produced. Then, when the field is turned off for a time θ the exciton density increases somewhat and the concentration of the free carriers decreases by several times (see Fig. 3), leading to a strong increase in the condensation rate. If the exciton density in the system is sufficient to produce viable nuclei in a time θ, then the drops will remain in the sample after the field is turned on, and their dimension will increase somewhat because of the increase of flux of matter from the gas phase. The system remains in this state until the next pulse application of the microwave field, so that no new nuclei can be produced.

Besides the duration θ of a single field-application pulse, it is possible to vary the repetition frequency ν of these pulses; then the condensation time is $\Delta t = \nu\theta\nu\Gamma$, where Γ is the observation time characteristic of our experiment, ~1 sec, and the minimum observable number of drops[6] is [see (2)]

$$N_{min}=\left(\frac{dN}{dt}\right)_{min} V\theta\nu\Gamma=A\exp\left[-\frac{\Lambda}{\ln^2(n_{ex}/n_{T\infty})}\right] V\theta\nu\Gamma. \qquad (12)$$

As seen from (12), when the time allotted to condensation is decreased ($\theta\nu<1$), the condensation will set in at a higher degree of supersaturation than at $\theta\nu=1$.

The condensation rate near the threshold of drop appearance is then higher:

$$dN/dt = \text{const}/\theta\nu.$$

By varying θ and ν we can experimentally determine the dependence of dN/dt on $n_{ex}/n_{T\infty}$.

We have measured the shift of the threshold of the EHD luminescence with changing pulse duration θ. From the experimental plots of $I(G)$, in analogy with Ref. 20, we determined the degree of supersaturation $n_{ex}/n_{T\infty}$ corresponding to the condensation threshold. In the experiments described in Ref. 20, the frequency ν remained constant and equal to ~1 Hz, while the time θ ranged from 10^{-3} to 10^{-7} sec. The results were plots of the average radiation intensity against the pump, which showed that when θ is decreased the degree of supersaturation needed to start the condensation increases.

To compare our results with the theory [Eq. (12)] it was convenient to represent the results in the form $\ln\theta^{-1} = f(\ln^2 n_{ex}/n_{T\infty})$. In this coordinate system, the formula (12) is plotted as a straight line whose slope determines Λ and with it the value of the surface tension σ, while the intercept with the ordinate axis yields the pre-exponential factor A. Figure 7 shows the result of a comparison of our experimental data with the predictions of the theory.

It turned out[20] that in the range 10^{-5} sec $< \theta < 1$ sec the function (12) agrees satisfactorily with experiment. We were able to determine the values of Λ and σ and show[20] that they correlate well with other measurements and calculations.[3] At small $\theta (< 5 \cdot 10^{-6}$ sec), however, as seen from Fig. 7, a deviation of the observed function $\theta^{-1} = f(\ln^2 n_{ex}/n_{T\infty}$ from the theoretical is observed. This could be naturally attributed to the nonstationary character of the condensation process at short times.

The nonstationary condensation process was first considered theoretically by Zel'dovich[21] and subsequently investigated by many workers.[15] On the basis of the analogy of the kinetic equation of nucleation with the diffusion equation, Zel'dovich has shown that the nonstationary condensation rate is

$$\frac{dN}{dt}(\theta) = \left(\frac{dN}{dt}\right)_{stat} \exp\left(-\frac{\tau^*}{\theta}\right), \qquad (13)$$

where τ^* is the characteristic diffusion time of the distribution function of the nuclei over the dimensions in dimension space. The nonstationary character of the condensation process is due to the fact that during the time θ the "average" nucleus cannot grow and exceed the critical dimension corresponding to the degree of supersaturation existing in the system. So far, to our knowledge, the nonstationary effect that follows from (13) has not been observed.

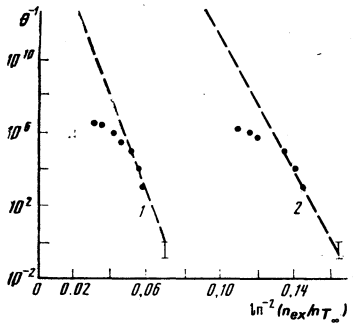

FIG. 7. Dependence of the condensation rate $dN/dt \propto \theta^{-1}$ on the degree of supersaturation: 1—$T = 1.53$ K; 2—$T = 1.85$ K. The dashed straight lines correspond to Eq. (1).

It appears that the results shown in Fig. 7 and in Ref. 20, which pertain to $\theta < 5 \times 10^{-6}$ sec, are due to the nonstationary character of the condensation process. It has turned out, however, that near the condensation threshold the quantity N_{min} is indefinite, since the EHD emission intensity changes jumpwise.[20] The jumps of the emission intensity, discussed above, are observed only in the strong microwave fields and are due to the motion of the nuclei.

To separate the purely kinetic effects from the effects connected with motion, it was necessary to choose a heating-field intensity such that no jumpwise change of the radiation intensity was observed. This, however, decreases the shift of the threshold of the ascending branch, leading to a decrease of the drop of the concentration of the excitons and free carriers in the heating field also when the field is turned off. As a result, at a frequency $\nu \sim 1$ Hz, the condensation threshold in the heating field hardly shifts at $\theta < 10^{-5}$ sec, making it impossible to investigate the nonstationarity effects. It was therefore convenient to modulate the microwave power by pulses with an off-duty cycle 2 and with a period $2\theta = 10^{-3} - 5 \times 10^{-7}$ sec.[7)]

In this case, when the modulation frequency of the heating field is changed, the average time Δt allotted to condensation does not change and is half as long as in the absence of the heating field. The threshold rate of condensation in an alternating field is double that in a zero field. Inasmuch as the change of the degree of supersaturation needed to change dN/dt by a factor of 2 is negligible, the $I(G)$ curves should not be affected by the change of the frequency of the microwave modulation, so long as the exciton density and the electron density manages to follow the pulses of the field, and so long as the rate of the condensation process is stationary, i.e., $\theta \gg \tau^*$.

The results of the experiment are shown in Fig. 8. It is seen that at $\nu < 50$ kHz the $I(G)$ curves do not depend on ν and do not differ from the case when there is no heating field (curve 1 on Fig. 8a). At $\nu > 100$ kHz, a distinct shift of the condensation threshold towards larger pumps is observed (curves 2–9). Finally, at $\nu \approx 1$ MHz the motion of the threshold slows down and with further increase of the modulation frequency to 2 MHz its position becomes unchanged.

Figure 8b shows by way of illustration the function $I(G)$ at a large field intensity, when a jump of the radiation intensity is observed at $\theta = 0$. In this case, when θ and ν are changed, $\theta\nu$ remains constant at 10^{-2}; this has led to an initial shift of the condensation threshold at $\theta = 10^{-3}$ sec.

In our opinion the results can be due only to the non-

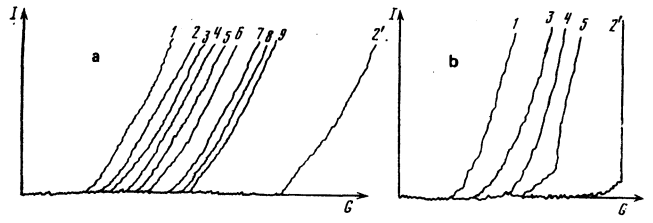

FIG. 8. Ascending branch of $I(G)$ at different regimes of turning-on the microwave power, $T = 1.65$ K. Curves 1—no heating field, 2'—field applied constantly. a) $\nu\theta = 0.5$, microwave field intensity $E \sim 3$ V/cm, ν (MHz): curve 2—0.1, 3—0.2, 4—0.3, 5—0.4, 6—0.6, 7—0.8, 8—1.0, 9—2.0; b) $\nu\theta = 10^{-2}$, $E = 5$ V/cm, ν (MHz): curve 3—10^{-4}–10^{-3}, 4—10^{-2}, 5—$3.3 \cdot 10^{-2}$.

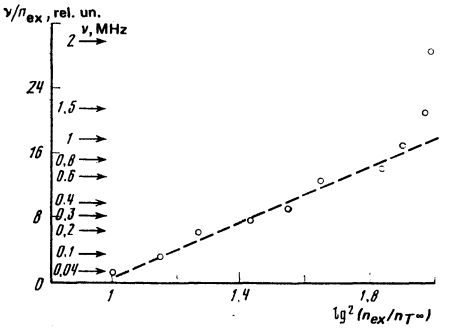

FIG. 9. Comparison of experimental data (Fig. 8a, $\nu\theta = 0.5$) with expression (15). The horizontal arrows indicate the microwave-field modulation frequencies.

stationary character of the condensation process. The only effect that could lead to outwardly analogous results is slow relaxation of the exciton and electron density. We note, however, that the possibility of condensation in the absence of a field is due primarily to the decrease of the concentration of the free carriers, which takes place within the characteristic binding time $\sim 10^{-7}$ sec. When the field is again turned on, the exciton density can decrease by a small amount and quite slowly but a substantial increase of the density of the free carriers will take place quite rapidly (see Fig. 3), so that the nuclei cannot be produced. We note that the effect of slowing down is furthermore strongly nonlinear with respect to the carrier density.[8] All this allows us to assume that the observed effect is not connected with relaxation phenomena, at least up to $\nu \lesssim 1.5$ MHz.

We have thus observed that nucleation characterizes by a certain in time τ^* the nucleus does not manage to turn into a viable drop. To explain the physical meaning of the time τ^*, we note that in the course of its production and growth the nucleus retains for the longest time a range of dimensions near the critical value R^*. The width δR of this region is determined from the condition that the change of the thermodynamic potential Φ near its maximum is of the order of kT;

$$\Phi(R^*) - \Phi(R^* \pm \delta R/2) \sim kT.$$

The change of the dimension of the nucleus near R^* can be regarded as diffusion in dimension space,[2,15] since $(d\Phi/dR)_{R^*} = 0$; the time necessary to pass through the region $R^* - \delta R/2 < R^* < R^* + \delta R/2$ is $\tau^* = (\delta R)^2/4D$, where D is the corresponding "diffusion coefficient."

To compare the behavior G_+ when θ is varied (see Fig. 8) with Eq. (13) we must express τ^* in terms of experimental quantities. Using the expressions for $(\delta R)^2$ in D, given in Frenkel's book,[2] we obtain

$$\tau^* = \frac{4\sigma}{n_{ex} v_T kT \ln^2(n_{ex}/n_{T\infty})}. \qquad (14)$$

An estimate of the value of τ^* for EHD yields $\sim 10^{-6}$ sec. Using (14), we obtain from (13) after taking the logarithms ($\theta = 1/2\nu$)

$$b = a\nu/n_{ex} + \ln^2(n_{ex}/n_{T\infty}), \qquad (15)$$

where b and a are constants of appropriate dimensionality. The quantity $n_{ex}/n_{T\infty}$, which corresponds to the condensation threshold at $\nu < 10^5$ Hz, was assumed equal to 10, in accordance with Ref. 3; it was assumed further that $n_{ex} \sim G_+$.

Figure 9 shows the results of a comparison of the experimental data (Fig. 8a) with Eq. (15). It is seen that the experimental points fit a straight line up to $\nu \approx 1$ MHz. The deviation from the theoretical relation at $\nu \gtrsim 1.5$ MHz is possibly due to the relaxation process in the exciton-electron plasma.

The nonstationary effect of nucleation observed by us in the time interval 10^{-5} sec $> \theta > 5 \times 10^{-7}$ sec is thus satisfactorily described by the equation of diffusion of the distribution function of the nuclei in dimension space. The most substantial is the set of nucleus states near R^*, where the thermodynamic force acting on the nucleus is $\partial\Phi/\partial R = 0$.

CONCLUSION

We have observed, for the first time ever, the influence of free carriers on the processes of nucleation, growth, and motion of EHD. The observed effect of slowing down the condensation with increasing free-carrier density, due to the non-isothermy of the condensation process, is a convenient method of studying the kinetics of nucleation. We were able to observe with its aid a non-stationary effect of nucleation and compare it with the classical theory of condensation.

The hysteresis effect in the action of a microwave field on the condensation of an exciton-electron plasma, investigated in this paper, uncovers a number of new possibilities for the study of the regularities of a phase transition of the gas-liquid type. At the same time it makes it possible to obtain a very broad and controllable hysteresis, which may be of interest for technical applications.

[1)] A unique effect of quenching of the EHD luminescence is observed when a sample placed in a heating field is subjected to additional illumination pulses. If a pump $G > G_-$ is used such that even at the maximum microwave field intensity there is no escape of drops (see Fig. 4a), and the sample is then illuminated by a single pulse from a GaAs laser of duration $\sim 10^{-6}$ sec, the radiation from the drops stops. The

explanation of this effect is that pulsed excitation produces a large concentration of hot carriers and consequently the phonon dragging force turns out to be very large; previously produced drops are carried out of the excitation region, and new ones cannot penetrate there. We note that in the absence of a heating field pulsed illumination leads to an increase of the luminescence signal.[5]

[2)]The kinetic energy of a hot carrier for $T = 15$ K amounts to $\varepsilon_k \sim 1.3$ meV, $\varepsilon_k < \varphi_0 + \varepsilon_{ex}$.

[3)]It turned out that to obtain identical effects of the shift of the condensation threshold in the case of cyclotron resonance the heaters must be more strongly heated than at $H = 0$. This is understandable if it is recognized that in a constant magnetic field the flux of free particles per nucleus decreases by almost a factor of 3, and consequently the same slowing-down effect calls for a much higher carrier density.

[4)]This time was obtained under the assumption that in the interval between the collisions with the impurities the drop moves like a freely diffusing particle.

[5)]The time for the nucleus to grow to $R \sim 10^{-5}$ cm amounts to $\sim 10^{-6}$ sec and is determined mainly by the time of its passage through the barrier of the thermodynamic potential, while the time of transformation into a stable macroscopic drop of radius 10^{-4} amounts to $\sim 10^{-4}$ sec at $n_{ex} = \varkappa n_{T\infty}$.

[6)]We note that in measurements of the rate of the condensation processes in Refs. 3 and 19, when $\theta\nu = 1$, the condensation time Δt is equal to the characteristic observation time.

[7)]It turned out that at a high repetition frequency ($\nu \gtrsim 10^3$ Hz) there are usually no jumps of the radiation intensity even in a stronger field; if at the same time the slope of the $I(G)$ plot does not differ from the slope in a zero field (Figs. 8b), then it can be assumed that the effects of EHD motion do not influence the results of the experiment.

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