Metastable states in the phase transition of an exciton gas into an electron-hole liquid and abrupt nucleation of this liquid

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If, during the phase transition of an exciton gas into an electron-hole liquid, the liquid is nucleated homogeneously, and droplets escape from the sample through diffusion or drift, the observed nucleation threshold will be very high and dependent on the boundary conditions on the system but not on the temperature. An important role here is played by the capture of droplets by point centers and the detachment of droplets from these centers by external forces. If the droplets which arise in a crystal cannot be captured at centers, the state of the highly supersaturated gas and of the liquid droplets which are moving through the sample will be metastable. The system may go into a stable state with a low supersaturation and with all the droplets trapped in the volume. At a transition of this type, the density of the liquid should increase abruptly. An approximate method has been developed for solving the steady-state kinetic equation of the nucleation.

At the gas-liquid phase transition in a pure system the threshold for the nucleation of the liquid shifts to a gas density higher than the thermodynamic boundary for the transition. This shift and the appearance of a metastable supersaturated state of the gas result from the low rate at which viable nucleating regions of the liquid phase appear.[1,2] One such transition is the condensation of excitons into an electron-hole liquid in pure germanium.[3]

On the other hand, there are situations in which the nucleation rate (in the ordinary sense of the term), is high but the metastable state is long-lived. A situation of this sort arises when the mobile nucleating regions ("nuclei") of the liquid phase can escape rapidly from the excitation region or can recombine at the wall of the sample.[4,5] In such cases the appearance of a viable nucleus is a consequence not of a competition between the evaporation and attachment of excitons but of a "successful" random walk of a nucleus through the sample or of the capture of a nucleus by a fixed force center. The nucleation threshold is essentially independent of the temperature, being determined by, for example, the thickness of the sample and the impurity concentration. The threshold exciton density may exceed the density of supersaturated vapor by two or three orders of magnitude.

It has been found experimentally[4–6] that the liquid can appear abruptly near the threshold; i.e., the observed amount of liquid increases sharply in a short time and upon a small increase in the pump level (Fig. 1). An abrupt behavior of this sort, which is natural for systems with an infinite lifetime "when one nucleus which has successfully grown reaches a macroscopic size and removes the supersaturation from the entire system,"[2] is unusual indeed for an electron-hole liquid. In the latter case the stable size of the droplet is bounded, and an increase in the density of the liquid is seen as a sequence of microscopic jumps which, at the ordinary experimental sensitivity, merge into a smooth curve. For condensation to occur abruptly, a large number of "supercritical" droplets would have to form essentially simultaneously.

We will begin with a qualitative explanation of abrupt nucleation. We will then (§§1,2) derive a system of kinetic equations describing steady-state nucleation, and we will point out a simple method for finding an approximate solution of this system. In §3 we return to abrupt nucleation and calculate the contributions of time-varying effects.

We consider a sample of pure germanium at whose surface electrons and holes are excited by an external light source. These electrons and holes are bound into excitons. Droplets of an electron-hole liquid are nucleated near the illuminated surface and are drawn into the interior of the sample by force due to, for example, the "phonon wind"[7] or a nonuniform strain.[8] When they reach the opposite face of the sample, the droplets adhere to it and rapidly recombine.

If the nucleation occurs homogeneously, and a mobile nucleus in the sample manages to collide with a force center, is captured by this center[9] and grows into a macroscopic

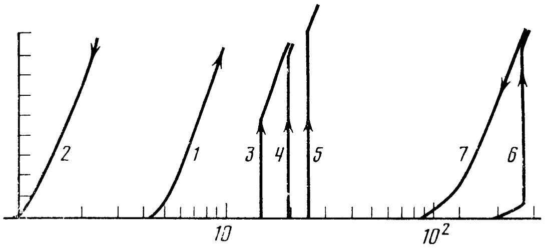

FIG. 1. Intensity of the emission from an electron-hole liquid, J, versus the rate of the external pumping, G. Curves 1–5—germanium sample 0.5 mm thick with an impurity density $N = 3\cdot 10^{10}\ \text{cm}^{-3}$ in a microwave field of power P, which heats free carriers and creates a phonon wind[4]; 1—$P = 0$; 3—34 mW; 4—43 mW; 5—mW; 2—stable branch of the hysteresis function $J(G)$. Curves 6 and 7 (Ref. 5)—germanium sample 10^{-3} cm thick with an impurity density $N = 3\cdot 10^{10}\ \text{cm}^{-3}$; 6—metastable branch of $J(G)$; 7—stable branch of $J(G)$.

droplet containing ν_s particles (ν_s increases with increasing exciton density).

The binding energy of a droplet with a point center does not depend on the size of the droplet, while the force which pulls the droplet into the interior of the sample is usually proportional to the number (ν) of particles in the droplet, so that there exists a number $\tilde{\nu}$ such that at $\nu > \tilde{\nu}$ a center cannot hold the droplet.

At $\nu_s > \tilde{\nu}$ a droplet which has been captured by a center grows, breaks away from the center and escapes from the sample without reaching a stable size. Near the nucleation threshold the average density of the liquid is low, since the average number of droplets which have been captured by centers is low at any instant. This state is metastable, and if an external source is used to produce a large number of droplets at force centers (e.g., an intense and brief illumination of the sample[10]) the system will go into a stable state in which all the droplets are captured by centers, the exciton density is low, and $\tilde{\nu} > \nu_s$.

We now take into account the unavoidable fluctuations in the nucleation rate in the metastable state (these fluctuations might be caused by, for example, fluctuations of the external pump). If many droplets arise simultaneously, the exciton density and the value of ν_s decrease sharply, and we may find a situation with $\nu_s < \tilde{\nu}$. In this case all the droplets which are nucleated remain inside the sample; i.e., the system "remembers" a single, sufficiently large fluctuation. The system may therefore be in two different states at the same average pump level: a "static" state, in which the droplets are at rest, and there is essentially no nucleation; and a "current" state, in which the nucleation rate is high, and the droplets are captured by centers, grow to $\nu > \tilde{\nu}$, detach from the centers, and move off to the wall of the sample.

It is a transition from the current state to the static state, due to the competition between capture and detachment, which constitutes the abrupt nucleation of a liquid.

§1. SYSTEM OF EQUATIONS

A droplet of an electron-hole liquid consisting of ν particles changes size as a result of the attachment or loss of an exciton and thereby undergoes a transition from the class (ν) to the class ($\nu \pm 1$) (Refs. 2 and 3). The droplet disappearance as a whole is described in the kinetic equation by the term[5,11] $-g_\nu/T_\nu$, where g_ν is the number of droplets consisting of ν particles, and T_ν is the time required for a droplet to escape from the sample.

Taking capture and detachment into account, we can write the equations of motion of a droplet along the ν axis as

$$\frac{dg_\nu}{dt}=\alpha_{\nu-1}g_{\nu-1}-(\alpha_\nu+\beta_\nu)g_\nu+\beta_{\nu+1}g_{\nu+1}-\frac{g_\nu}{T_\nu}, \tag{1}$$

$$T_\nu=T_0\exp(NdS_\nu) \quad \text{for} \quad \nu<\tilde{\nu}, \qquad T_\nu=T_0 \quad \text{for} \quad \nu>\tilde{\nu}, \tag{2}$$

$$\frac{dg_1}{dt}\equiv\frac{dn}{dt}=G-\frac{n}{\tau_e}-\pi r_0^2 v_T\sum_{\nu=2}^{\infty}[n-n_s(\nu)]\nu^{2/3}g_\nu; \tag{3}$$

the coefficients α_ν and β_ν are given by the known expressions[2,3]

$$\alpha_\nu=\pi r_0^2\nu^{2/3}v_T n, \quad \beta_\nu=\alpha_\nu\frac{n_s}{n}\exp(\gamma\nu^{-1/3})+\frac{\nu}{\tau_0},$$

where $r_0=(3/4\pi n_0)^{1/3}$, n_0 is the density of particles in a droplet, v_T and τ_e are the thermal velocity and lifetime of the excitons, n is their density, n_s is the saturation vapor density, $\gamma = 2\sigma/n_0kTr_0$, σ is the surface tension of the liquid, and τ_0 is its lifetime.

In Eq. (1) for the size distribution of the droplets at $\nu < \tilde{\nu}$, the last term contains a factor $\exp(-NdS_\nu)$, which makes it possible to take into account the capture of droplets to fixed centers. The density of centers is N; the thickness of the sample (or the width of the excitation region) is d; and the capture cross section is $S_\nu = \pi r_0^2\nu^{2/3}$. The capture of droplets is described by analogy with the absorption of particles moving through a medium with an absorption length $\lambda = (NS)^{-1}$. At $\lambda \gg d$, the capture can be ignored, while at $\lambda \ll d$ the term describing the removal of droplets is negligibly small; i.e., the nucleation becomes essentially heterogeneous. The time required for the escape of a droplet of a sample is $T_0 = d/v$, where v is the drift velocity (to simplify the discussion we are ignoring the diffusion of droplets, which plays a major role in thin germanium samples[5]). To find a rough estimate of $\tilde{\nu}$ we can equate the binding energy of a droplet with an impurity, ε, to the kinetic energy acquired by a droplet due to the force $F = \nu f$ over a distance equal to the "width of the surface layer," i.e., a distance on the order of the first Bohr radius of an exciton, a_B. From $\varepsilon \sim f\nu a_B$ we find

$$\tilde{\nu}\sim\varepsilon\tau_p T_0/m^* d a_B.$$

Here we have used the substitution $v = f\tau_p/m^*$, where τ_p is the scale time for the momentum relaxation of a droplet through collisions with acoustic phonons. Substituting in the values $\varepsilon \approx 5\cdot10^{-3}$ eV (Ref. 12), $\tau_p \sim 10^{-9}$ s (Refs. 13 and 14), and $a_B \sim 10^{-6}$ cm, we find $\tilde{\nu} \sim 3\cdot10^{10}/v$. At typical velocities of the drift motion of droplets, $v = 10^3$–10^5 cm/s, we find $\tilde{\nu}$ to be $10^5 - 10^8$ in order of magnitude, i.e., comparable to the stable size of the droplets, ν_s at which the condition $\alpha_s = \beta_s$ holds.

Equation (4) describes the kinetics of the formation, recombination, capture, and evaporation of excitons.

§2. STEADY-STATE DISTRIBUTION FUNCTION

We consider the steady-state situation near the nucleation threshold; i.e., we set $dg_\nu/dt = 0$, $dn/dt = 0$, and $G \gtrsim n/\tau_e$. In this case the exciton density n is independent of g. Following Ref. 11, we sum Eqs. (1) from ν to ∞:

$$\alpha_{\nu-1}g_{\nu-1}-\beta_\nu g_\nu=\sum_{i=\nu}^{\infty}\frac{g_i}{T_i}\equiv j_\nu. \tag{4}$$

The quantity j_ν is proportinal to the resultant flux of droplets containing more than $\nu - 1$ particles to the wall of the sample. Since g_i and T_i are positive quantities, the function j_ν falls off monotonically with increasing ν. We note that

$$g_{\nu-1} \equiv T_{\nu-1}\left(\sum_{i=\nu-1}^{\infty}\frac{g_i}{T_i}-\sum_{i=\nu}^{\infty}\frac{g_i}{T_i}\right)=T_{\nu-1}(j_{\nu-1}-j_\nu),$$

$$g_\nu = T_\nu(j_\nu - j_{\nu+1}). \tag{5}$$

Substituting (5) into (4), we find an equation equivalent to (1):

$$j_{\nu-1}-(1+a_{\nu-1}+b_{\nu-1})j_\nu+a_{\nu-1}j_{\nu+1}=0, \tag{6}$$

where

$$a_{\nu-1}=\frac{\beta_\nu T_\nu}{\alpha_{\nu-1}T_{\nu-1}}, \quad b_{\nu-1}=\frac{1}{\alpha_{\nu-1}T_{\nu-1}}.$$

To find an approximate of (6), we introduce[5,11] the function ψ_ν by means of $j_\nu = \exp(-\psi_\nu)$. We can then write

$$j_{\nu\pm1}\approx \exp(-\psi_\nu \mp d\psi/d\nu). \tag{7}$$

Subsituting (7) into (6), and solving the quadratic equation for $z = \exp(d\psi/d\nu)$, we find

$$j_\nu \propto \exp\left(-\int_1^\nu \ln z \, d\nu\right),$$

$$z_{1,2}=\frac{1+a+b}{2}\pm\left[\left(\frac{1+a+b}{2}\right)^2-a\right]^{1/2}. \tag{8}$$

Since $j(\nu)$ is a monotonic function, the expansion in (7) is valid along the entire ν axis. The boundary condition $j \to 0$ in the limit $\nu \to \infty$ determines the choice of sign (+) in solution (8). Combining (5), (7), and (8), we find

$$g(\nu)\approx\frac{T_\nu \ln z}{T_1 \ln z_1} n \exp\left(-\int_1^\nu \ln z\, d\nu\right), \tag{9}$$

where the factor $n/T_1 \ln z_1$ has been introduced to satisfy the boundary condition $g_1 = n$. Expression (9) gives the approximate WKB solution of steady-state equation (1). We turn now to some particular cases.

We first assume tht $T_\nu = T_0$ does not depend on ν, i.e., that there are no effects resulting from the capture and detachment of droplets. Figure 2 shows the function $g(\nu)$ calculated for various values of T_0 and for an exciton density near the "thermodynamic" nucleation threshold; in these calculations we assumed that under the conditions $a < 1$ and $b \ll 1$ (at large T_0) we would have $\ln z \approx b$ (with $a > 1$ and $b \ll 1$ we have $z \approx a$).

At large values of T_0 (i.e., at $\alpha T_0 = b^{-1} \gg 1$) the function $g(\nu)$ decreases sharply up to $\nu \sim \nu_c$ (curve 1 in Fig. 2), where the critical size ν_c is the size at which the rates of evaporation and attachment are equal: $\alpha_c = \beta_c$. At $\nu \gtrsim \nu_c$, the functional dependence of g on ν becomes a power law: $g \propto \nu^{-2/3}$.

Near the stable size ν_s ($\alpha_s = \beta_s$), at which the attachment rate is nearly equal to the recombination rate, the function $g(\nu)$ has a sharp maximum, g_s, as shown (in exaggerated form) by curve 1 in Fig. 2. The width of this maximum is $\delta\nu \sim \nu_s^{1/2}$, so that it appears as a vertical line in the scale of Fig. 2.

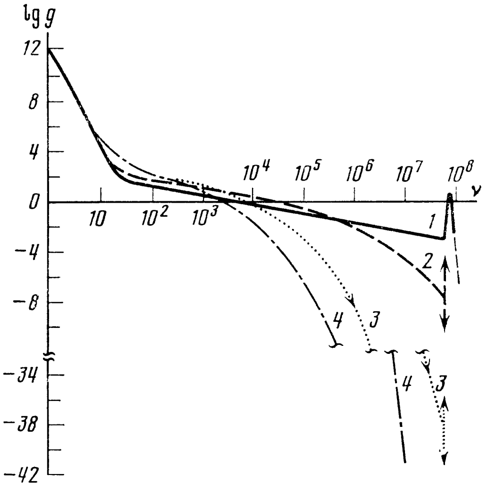

FIG. 2. Steady-state droplet size distribution $g(\nu)$ ($T = 1.9$ K, $N = 0$, $n = 2 \cdot 10^{12}$ cm^{-3}) at several values of T_0: 1—10^{-4} s; 2—$2\cdot10^{-5}$ s; 3—$2\cdot10^{-6}$ s; 4—10^{-6} s.

That simple expression (9) can be used to describe the rapidly varying function $g(\nu)$ along the entire ν axis is an advantage of this method solving Eq. (1) in comparison with other methods.[1)] On the other hand, this method is not free of disadvantages: Near ν_c the curves corresponding to different values of T_0 intersect, but this could of course not happen for real functions $g(\nu)$. Furthermore, the maximum g_s shifts slightly upward on the ν scale from the actual extremum point ν_s.

As T_0 increases, the distribution function essentially retains its shape; there is only an increase in the height of the maximum g_s. The physical meaning of this result is that at large values of T_0 the neighborhood of the critical nucleus ν_c is a "source" which furnishes droplets to the state ν_s (§3), and if the time required for the transition of one droplet from ν_c to ν_s is short in comparison with T_0 then we would have

$$g_c/\tau_c \sim g_s/T_0, \tag{10}$$

where τ_c is the Zel'dovich time,[2] equal to the time required for a droplet to traverse the critical region (the usual values are[4] $\tau_c = 10^{-5}$–10^{-6} s). A decrease in T_0 leads to a change in $g(\nu)$ in the region $\nu \gtrsim \nu_c$; in the interval $\nu_c < \nu < \nu_s$ at small values of T_0 the denisty of macroscopic droplets falls off sharply (curve 3 in Fig. 2). As has been pointed out elsewhere,[5] we are seeing here a difference between the recombination of a droplet as a whole and recombination due to the finite lifetime of the particles in a droplet.

When capture and detachment are taken into account, we find fundamental changes in the function $g(\nu)$ in the region $\nu \sim \nu_s$. If the large droplets "stick" in a sample (because of capture by defects, decrease in the velocity toward the boundary, etc.), the density g_s will increase exceedingly rapidly. Curve 2 in Fig. 3 shows the changes in the distribution function caused by introducing $3 \cdot 10^9$ impurity per cubic centimeter in a sample 1 mm thick: At $\nu \gtrsim 10^3$ the density becomes the same as a large values of T_0, and near ν_s the height

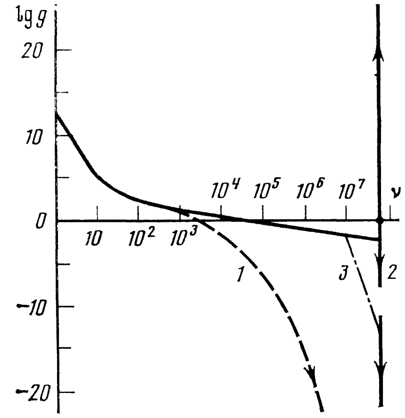

FIG. 3. Effect of capture and detachment on the steady-state distribution function $g(\nu)$ ($T = 1.9$ K, $n = 2\cdot 10^{12}$ cm^{-3}, $T_0 = 10^{-6}$ s, sample thickness $d = 1$ mm). 1—Pure sample ($N = 0$); 2—density of capture centers $N = 3\cdot 10^{9}$ cm^{-3}, $\tilde{\nu} > 10^{8}$; 3—$N = 3\cdot 10^{9}$ cm^{-3}, $\tilde{\nu} = 10^{7}$.

of the maximum turns out to be prodigious, $g_s \sim 10^{62}$ [this result becomes understandable when we note that[2)] $T_s = T_0 \exp(NdS) \sim 10^{57}$ s. Using (10), we find $g_s \sim 10^{64}$, in order-of-magnitude agreement with the calculation from (9)].

Finally, if we assume that droplets containing more than $\tilde{\nu} = 10^{7}$ particles cannot be captured by impurities and escape from the sample in a time T_0, we find curve 3 in Fig. 3.

To conclude this section of the paper we note that the solution of the steady-state problem of nucleation with escape of the liquid from excitation region leads to a distribution function $g(\nu)$ which is physically reasonable and which can be arranged experimentally. The analogous solution for the limit[3] $T \to \infty$ yields excessively large values $g_s \sim \exp(\nu_s)$, showing that time-varying effects must be taken into account.

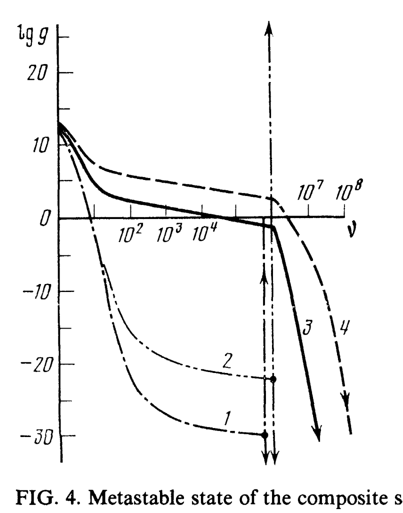

FIG. 4. Metastable state of the composite system consisting of excitons and electron-hole liquid ($T = 1.9$ K, $N = 3\cdot 10^{10}$ cm^{-3}, $T_0 = 10^{-6}$ s; $d = 1$ mm, $\tilde{\nu} = 10^{6}$). 1—$n = 5\cdot 10^{11}$ cm^{-3}, $g_s \sim 10^{-1}$; 2—$n = 6\cdot 10^{11}$ cm^{-3}, $g_s \sim 10^{24}$; 3—$n = 2\cdot 10^{12}$ cm^{-3}; 4—$n = 4\cdot 10^{12}$ cm^{-3}

§3. METASTABLE STATES AND ROLE PLAYED BY TIME-VARYING PROCESSES

Figure 4 shows steady-state functions $g(\nu)$ for $T_0 = 10^{-6}$ s and $\tilde{\nu} = 10^{6}$. We first look at curves 1 and 2, for which the density of macroscopic droplets, g_s, is large, but the density of droplets in the intermediate region ($\nu_c < \nu < \nu_s$) is very small. If an experiment is carried out in such a manner that the pump (and thus the exciton density) increases gradually from zero, then each droplet must spend some time in this region before it reaches the size ν_s. Consequently, the high steady-state density g_s occurs because the droplets accumulate for a long time at the point ν_s, and if the duration of the experiment, θ, is short in comparison with the accumulation time, $T_s = T_0 \exp(NdS)$, the term dg_ν/dt, rather than g_ν/T_ν, will dominate Eq. (1).

The solution of time-varying equation (1) is not known for arbitrary α_ν, β_ν, and T_ν. For a qualitative analysis of the transient processes, we transform from (1) to the second-order differential equation

$$\frac{\partial g}{\partial t} = \frac{\alpha+\beta}{2}\frac{\partial^2 g}{\partial \nu^2} + (\beta-\alpha)\frac{\partial g}{\partial \nu} + \left(\frac{d\beta}{d\nu} - \frac{d\alpha}{d\nu} - \frac{1}{T}\right) g. \qquad (11)$$

If we ignore the derivative $d\beta/d\nu$ and $d\alpha/d\nu$ in comparison with $1/T$, we find that Eq. (11) describes a mixed diffusion and drift with a finite lifetime T. Far from the points $\beta = \alpha$, Eq. (11) reduces to a first-order equation,

$$\partial g/\partial t = (\beta-\alpha)\,\partial g/\partial \nu - g/T,$$

i.e., the equation of the drift motion of an arbitrary perturbation with a finite lifetime and a "velocity" $\beta - \alpha$. We can thus find the scale time for the motion of the droplet over the intervals $1 < \nu < \nu_c$ and $\nu_c < \nu < \nu_s$ in the form $t \sim \int d\nu/(\beta - \alpha)$. For the region $1 < \nu < \nu_c$ this time, equal to the evaporation time of a critical nucleus, is approximately equal to the scale time for relaxation to a steady state; with $\nu_c \sim 10^2$ and $n \sim 10^{12}$ cm^{-3} it is $\lesssim 10^{-6}$ s in order of magnitude. The scale time for the motion of an individual droplet out of the supercritical region is also extremely short ($\sim 5\cdot 10^{-5}$ s for $n \sim 10^{122}$ cm^{-3} and $\nu_s \sim 10^{7}$). If, however, the density of stable droplets, g_s, is large in comparison with g_c, the time required for relaxation to a steady state, t_1, will be large in comparison with t. If, at $\nu > \nu_c$, we have a parameter value $b = \alpha^{-1} T^{-1} \ll 1$, then we can find the time t_1 approximately as the ratio of the steady-state density g_s to the flux g_c/τ_c, of droplets through the critical region, where τ_c is the time required for droplets to traverse the critical region. This time was found in Ref. 2 in the diffusion approximation. As we show in the Appendix to the present paper, the solution of the diffusion-drift equation of motion of a nucleus near $\nu = \nu_c$ gives a result essentially the same as that found in Ref. 2.

Accordingly, we can use (10) to estimate t_1. If the condition $b \gtrsim 1$ holds in the supercritical region, i.e., if the time T_ν is short, we would have to replace g_c by the minimum value of g on the interval $\nu_s > \nu > \nu_c$. If $t_1 \gg \theta$, the system is metastable (no significant amount of liquid accumulates in the system over the time θ, although the steady-state density g_s may be very high). For example, a time $\theta \sim 10^{15}$ s would be required for a single droplet to reach the state ν_s in the case shown by curve 2 in Fig. 4. The exciton density would have to be increased for a detectable amount of liquid to form over a realistic experimental time $\theta \lesssim 10^5$ s. At $n > 6\cdot 10^{11}$ cm^{-3}, however, the relation between ν_s and $\tilde{\nu}$ changes ($\nu_s > \tilde{\nu}$); the relation between the typical time θ and the value of T_s also changes ($\theta \gg T_s$). For this reason, experiments deal with a steady-state function $g(\nu)$, and the value of g_s may be exceedingly small (curves 3 and 4). Near the nucleation threshold ($2\cdot 10^{12}$ cm$^{-3} < n < 4\cdot 10^{12}$ cm^{-3}) the liquid phase consists primarily of small droplets, and the exciton gas is highly supersaturated.

Using the functions $g(\nu)$ shown in Fig. 4 along with Eq. (3), we easily find that for a given external pump G there can be two different steady states: a static state ($5\cdot 10^{11} \lesssim n \lesssim 6\cdot 10^{11}$ cm^{-3}, $\nu_s \lesssim 10^6$) and a current state ($2\cdot 10^{12} \lesssim n \lesssim 4\cdot 10^{12}$ cm^{-3}). A transition from the current state to the static state would require the simultaneous (on a scale $\sim T_0$) fluctuational nucleation of $\sim 10^6$ liquid droplets.

CONCLUSION

In this paper we have analyzed the nucleation of the liquid for the case in which, in contrast with the Becker-Döring model,[1,2] not only the critical nuclei but the droplets of all sizes escape from the sample over a finite time. In this case the steady-state nucleation rate depends strongly on the mobility of the macroscopic droplets; if this mobility is high, there are essentially no large droplets in the system.

It is convenient to describe the nucleation process qualitatively as a diffusion-drift motion of a nucleus having a finite lifetime in size space.

In the metastable state the exciton gas is highly supersaturated, and the liquid which has been nucleated "flows" through the sample. A transition from the metastable state to the ground state requires the capture of a large number of droplets by force centers. Experimentally, this transition is seen as an abrupt increase in the total volume of liquid.

The abrupt nucleation is a fluctuational process in an open system far from equilibrium, in which a nucleation barrier is erected not by the "thermodynamic force," i.e., by the competition between the evaporation and attachment of gas particles, but by the kinetics of the flow and capture of liquid droplets. In a sense, there are two successive first-order transitions. The first is the nucleation of mobile droplets (this transition is essentially not observed unless the experimental apparatus is very sensitive), while the second is the abrupt appearance of a large number of immobile droplets; here the droplets which are moving through the sample play the role of the atoms of a gas, while the entire ensemble of rather small liquid droplets which have stopped plays the role of a single droplet of the new phase.

APPENDIX

To determine τ_c we work from Eq. (11). Near the point $\nu = \nu_c$ we can set $\beta - \alpha \approx (\beta' - \alpha')(\nu - \nu_c)$, and $(\alpha + \beta)/2 = \alpha_c$. We transform to the dimensionless variables

$$x = (\nu - \nu_c)\left(\frac{\alpha' - \beta'}{2\alpha_c}\right)^{1/2}, \quad \zeta = \frac{\alpha' - \beta'}{2}t.$$

Equation (11) can then be written

$$\frac{\partial g}{\partial \zeta} = \frac{\partial^2 g}{\partial x^2} - 2x\frac{\partial g}{\partial x} - 2\left(1 + \frac{1}{\eta}\right)g, \tag{A.1}$$

where $1/\eta = 2/T(\alpha' - \beta')$. In Eq. (A.1) it is convenient to use the substitution

$$p = g\exp(-x^2/2),$$

so that this equation becomes

$$\frac{\partial p}{\partial \zeta} = \frac{\partial^2 p}{\partial x^2} - \left(1 + x^2 + \frac{1}{\eta}\right)p. \tag{A.2}$$

Since we are intereted in the crossing of the barrier by an individual particle, we supplement Eq. (A.2) with the initial condition $p(x,0) = \delta(x - x_0)$. We solve Eq. (A.2) by Fourier transforms,[13] setting

$$p(x,t) = \int_{-\infty}^{\infty} f(k,t)\, e^{ikx}\, dk,$$

$$f(k,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} p(x,t)\, e^{-ikx}\, dx.$$

For the transform $f(k,t)$ we then find the equation

$$df/d\zeta = -(k^2 + 1 + x^2 + 1/\eta) f.$$

Integrating this equation,

$$f = f_0 \exp[(-k^2 - 1 - x^2 - 1/\eta)\zeta],$$

using the initial condition to determine f_0, and returning to the function $g(x,\zeta)$, we find

$$g(x,\zeta) = (4\pi\zeta)^{-1/2}\exp\left[\frac{x^2}{2} - \frac{(x - x_0)^2}{4\zeta} - \left(1 + x^2 + \frac{1}{\eta}\right)\zeta\right]. \tag{A.3}$$

Setting $|x| = |x_0|$, where $x - x_0 = 2x_0$, we consider the transition of a particle to the equivalent position $(-x_0)$ on the opposite side of the barrier:

$$g(-x_0,\zeta) = (4\pi\zeta)^{-1/2}\exp\left[x_0^2\left(\frac{1}{2} - \frac{1}{r}\right) - \zeta\left(1 + x_0^2 + \frac{1}{n}\right)\right].$$

The maximum of g is reached after a "time" ζ_c given by

$$\zeta_c \approx x_0(1 + x_0^2 + 1/\eta)^{-1/2}. \tag{A.4}$$

In the limit $\eta \to \infty$, i.e., in the absence of a current, we have $\zeta_c \approx 1$ at $x_0 \gtrsim 1$. If we identify the time interval found here, $\tau = 2\zeta_c/(\alpha' - \beta')$, with the value of τ_c, we find $\tau_c \approx 2/(\alpha' - \beta')$, in essential agreement with the time introduced by Zel'dovich.[2]

[1)] Equation (1) was solved directly in Refs. 5 and 11. Since the function $g(\nu)$, in contrast with $j(\nu)$, has extrema, the expansion in (7) is not valid near ν_c and ν_s, and it becomes necessary to match the approximate solutions obtained in different ν regions or to resort to numerical methods.

[2)] This estimate is on the high side. If we take into account the possibility of the detachment of a droplet from a force center as a result of a thermal fluctuation, we find that the "settled" droplet lifetime is $r_0 v_s^{5/6}(m^*/3kT)^{1/2}\exp(\varepsilon/kT) \sim 10^8 - 10^{16}$ s.

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