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# Time evolution of the electron-hole plasma nucleation: An analytical approach

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An analytical solution of the time evolution of the electron-hole plasma nucleation is presented. There are essentially two regimes of nucleation: a fast one and a slow one, depending on the excitation. The variation with the excitation of the exciton density, the amount of electron-hole liquid, the density of drops, and their size are compared with experiments.

## I. INTRODUCTION

Since the first suggestion by Keldysh of the existence of the electron-hole (e-h) plasma, a large amount of work has been devoted to the study of its thermodynamical properties,[1] and more recently to the nucleation[2-8] of such a liquid. The electron-hole condensation is very similar to a gas-liquid transition, except that the particles have a finite lifetime. The main consequence is that the electron-hole droplets cannot grow to form an ocean, but rather stop at a finite size when the amount of collected excitons on the surface compensates the decay of electron-holes in the drop, which increases as the volume. This stable size depends on the exciton density which must be larger than the thermodynamical density due to the electron-hole decay. The consideration of a steady-state situation implies that particles are continuously created, and the characteristics of the steady state depend on how it is reached; a given generation rate can *a priori* compensate the particle decay of a small number of large drops in a dense exciton gas, or a larger amount of smaller drops in a less dense gas.

In this paper we want to study the time evolution of the electron-hole liquid formation as a function of the exciton generation rate. Such a problem has been considered in the case of germanium by Staehli,[6] who uses, as most of the theoretical works on electron-hole droplet nucleation do, a stochastic approach where all the stages of "drops" size between the stable one to the exciton are considered. He then solves his rate equation numerically. We address ourselves to the analytical solution of this problem, which has been made possible because of approximations consistent with the extremely sharp dependence of the nucleation current $J(N_x)$ with respect to the exciton density $N_x$.

In Sec. II we write a set of equations, similar to the ones used by Keldysh,[7] giving the time evolution of the amount of excitons and electron-hole liquid. In Sec. III, we analyze its solution when $J$ is linearized around a fixed value of the exciton density. This crude approximation has the advantage that we can solve our equations exactly over the whole range of excitation. The solution exhibits the fact that there are essentially two regimes of nucleation, a slow and a fast one, depending on the value of an important parameter, the nucleation ratetime $\tau_\nu$, which depends on the exciton density as $1/J(N_x)$.

In Secs. IV and V, we study separately the regimes of slow nucleation and fast nucleation. In the light of the first analysis, we write two simplified sets of time evolution equations, valid in each regime. These simplifications are based on the fact that when the nucleation is very slow, the system is always very close to steady state and the decay terms are preponderant, while when the nucleation is very fast, the decay terms can be neglected compared to the change with time of the amount of excitons and liquid. We obtain in each case the dependences, on the laser excitation, of the number of drops, the radius of the drops, and the exciton density at steady-state equilibrium. The results are summarized in Sec. VI and compared with the experiments. We would like to emphasize that this calculation, as the preceding ones, applies to an ideal situation where excitons are created homogeneously inside the sample. Experimentally, this is not usually the case. One consequence is that the excitons diffusion inside the sample would make the quantity of excitons increase, even if the density of excitons in the droplet region decreases. So the exciton luminescence from the whole sample is not a measure of the exciton density. This tends to hide the observation of the overshoot.[5]

## II. RATE EQUATIONS

We consider a laser excitation which creates a density $N_0(t)/\tau_x$ of electron holes per unit time. The problem is to obtain the exciton density $N_x$, the amount $D$ of electron holes in the drops, the density of drops $\mathfrak{N}$ and their size $n=r^3$ as a func-

tion of $N_0(t)$. Four equations are needed; they will be Eqs. (1), (2), (6), and (7).

(a) *The conservation of electron holes reads*

$$N_0(t)/\tau_x = \dot{N}_x + N_x/\tau_x + \dot{D} + D/\tau_d . \quad (1)$$

(b) *The time evolution of the number of electron holes inside a drop* being ruled by the collection of excitons, their evaporation, and the electron-hole decay, reads[9]

$$\dot{n} = bN_x n^{2/3} - an^{2/3}\exp(-\tilde{\phi}_\infty + \tilde{s}n^{-1/3}) - n/\tau_d ,$$

where $\phi_\infty = \tilde{\phi}_\infty kT$ is the electron-hole liquid binding energy. The surface tension $S$ appears in the coefficient $\tilde{s}$ as

$$s = 2S(\tfrac{4}{3}\pi\rho)^{1/3}/\rho = \tilde{s}kT ,$$

where $\rho$ is the liquid density. The coefficient $b$ is given by

$$b = 4\pi(3/4\pi\rho)^{2/3}(kT/2\pi m_x)^{1/2} ,$$

where $m_x$ is the exciton center-of-gravity mass.

Let us introduce $\tau_c$ defined by

$$1/\tau_c = bN_{0x} \equiv a\exp(-\tilde{\phi}_\infty) ,$$

where $\tau_c$ is the exciton collection time per unit surface at thermodynamical density $N_{0x}$;

$$N_{0x} = g(2\pi m_x kT/h^3)^{3/2} e^{-\tilde{\phi}_\infty} = N_{00} e^{-\tilde{\phi}_\infty} ,$$

where $g$ is the exciton degeneracy. Using $n = r^3$, one can then give to the equation for $\dot{n}$ a simpler form:

$$\dot{r} = (1/3\tau_c)(R_x - e^{\tilde{s}/r}) - r/3\tau_d , \quad (2)$$

where the supersaturation $R_x$ is defined as

$$R_x = N_x/N_{0x} .$$

Equation (2), shown in Fig. 1, has three important consequences:

(i) There exist two equilibrium sizes given by $\dot{r} = 0$, a stable one $r_x$ and an unstable one $r^*$:

$$r_x \sim (\tau_d/\tau_c)(R_x - 1) ,$$
$$r^* \sim \tilde{s}/\ln R_x . \quad (3)$$

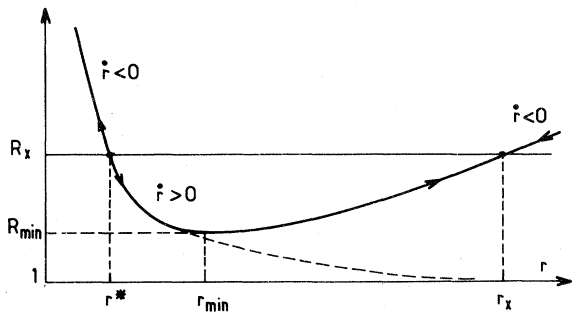


FIG. 1. Stable and critical size for e-h droplets.

$r^*$ is usually called the critical embryo size, and is introduced as the size corresponding to the maximum in the change in the enthalpy $G$ between a cluster of $n$ electron-hole pairs and $n$ excitons in the gas at density $N_x$, i.e.,

$$\Delta G(n) = (-n\phi_\infty + \tfrac{3}{2}sn^{2/3}) - nkT\ln(N_x/N_{00}) .$$

(ii) In order to have stable drops (or find a solution to $\dot{r} = 0$), one needs $R_x$ to be larger than a minimum value

$$R_{\min} \simeq 1 + (4\tilde{s}\tau_c/\tau_d)^{1/2} , \quad (4)$$

which corresponds to a minimum size $r_{\min} = \frac{1}{2}(\tau_d/\tau_c)(R_{\min} - 1)$ (that differs from $r_x$ by a factor of $\frac{1}{2}$). To obtain these quantities, we have replaced $e^{\tilde{s}/r}$ by $1 + \tilde{s}/r$ which is valid if $r_{\min}$ is large enough.

(iii) For large drops ($e^{\tilde{s}/r} \sim 1$) far from $r^*$, $r$ increases at constant $R_x$ as[10]

$$r \simeq r_x[1 - \exp(-t/3\tau_d)] . \quad (5)$$

(c) In order to obtain the density of drop $\mathfrak{N}$, we need to introduce the nucleation current $J$. Equation (2) would not allow an embryo to grow from $r = 1$ to $r^*$, because $\dot{r}$ is negative for $1 < r < r^*$, and no macroscopic drops should be formed from an exciton gas. In fact, the sizes between 1 and $r^*$ are populated by fluctuations and there indeed exists a nucleation current $J(t)$ which is the density of embryos passing the critical size $r^*$ by unit time. It is proportional[11] to the density $N^*$ of critical embryos which depends on the energy barrier $\Delta G(n^*)$ as $\exp[-\Delta G(n^*)/kT]$, if we assume that all the embryos between 1 and $r^*$ are in thermodynamic equilibrium

$$J = N^*/\tau' = j_0\exp(-\tilde{s}^3/2\ln^2 R_x) . \quad (6)$$

Such a relation between the number of embryos passing the neck point, and the density of excitons measured at the same moment is valid for phenomena changing on a time scale large compared with the time necessary to excitons to form a critical embryo. If one takes into account only exciton collection $\dot{r} = R_x/3\tau_c$, then this leads to an estimate of the minimum time to reach equilibrium between excitons and critical embryos:

$$\tau_{\inf} = (3r^*/R_x)\tau_c .$$

We will check this condition in Sec. V.

A reliable theory for the prefactor $j_0$ is not easy to find. One can get to within an order of magnitude by dimensional arguments: $j_0$ is in the order of the density of excitons divided by the time $\tau'$ necessary for an embryo with size $n^*$ to collect one exciton, which from Eq. (2) is $\tau_c/R_x n^{*2/3}$. This gives[12]

$$j_0 \sim N_x/\tau' = (N_{0x}/\tau_c)R_x^2 r^{*2} .$$

We will call a drop any embryo larger than $r^*$, so that the density of drops $\mathfrak{N}$ and the density $D$ of e-h in drops are related to $J$ by

$$\mathfrak{N}(t)=\int_0^t J(t')\,dt',$$
$$D(t)=\int_0^t J(t)r_{t'}^3(t)\,dt', \tag{7}$$

where $r_{t'}(t)$ is the size at time $t$ of a drop made at time $t'$ [with $r_t(t)\simeq r^*$].

(d) At that stage, it is important to figure out what are the numerical values of the various quantities. Table I give the bare parameters of Ge and Si. In Table II, we calculate some elaborated parameters at various temperatures. Note that the ones for silicon at 2 °K look crazy. We will come back to this point in Sec. VI. Table III gives the values of $J$, $r^*$, and $r_x$ for various $R_x$ at some fixed temperature. Note how $J$ is sensitive to the supersaturation $R_x=N_x/N_{0x}$.

The reader, essentially interested in the results, can skip Sec. III whose purpose is mainly to show how one goes from the slow-nucleation regime to the fast-nucleation regime, and to justify the approximations made for these two regimes in Secs. IV and V.

## III. ANALYSIS OF THE POSSIBLE SOLUTIONS

Combining Eqs. (1), (2), and (6) we obtain an integrodifferential equation for $N_x$ [the electron-hole drops (EHD) decay terms of (1) and (2) cancel exactly]:

$$N_0(t)/\tau_x=\dot{N}_x+N_x/\tau_x+J(t)n^*+\frac{1}{\tau_c}\int_0^t dt'\,J(t')r_{t'}^2(t)(R_x-e^{\tilde{s}/r}). \tag{8}$$

The third term is the amount of particles passing the neck size $n^*$, and the last term is the exchange of excitons at the surface of the drops made previously. Our problem is to solve Eq. (8). The nasty term is the last one, because it requires the story of $N_x$. But the form of $J$ is extremely sensitive to $N_x$ and this property will be of great help.

The dependence of $J$ on $R_x$ is so fast that if the

TABLE I. Values of the exciton lifetime $\tau_x$, drop lifetime $\tau_d$, work function $\phi_\infty$, surface tension $S$, electron-hole liquid density $\rho$, exciton mass $m_x$, and degeneracy $g$ for Ge and Si.

| | Ge | Si |
|---|---|---|
| $\tau_x$ ($\mu$sec) | 10 | 2 |
| $\tau_d$ ($\mu$sec) | 40 | 0.2 |
| $\phi_\infty$ (°K) | 23 | 80 |
| $S$ ($10^{-4}$ erg/cm$^2$) | 3.5 | 80 |
| $\rho$ ($10^{18}$ part/cm$^3$) | 0.23 | 3.7 |
| $m_x/m_{\text{electron}}$ | 0.33 | 0.6 |
| $g$ | 16 | 24 |

TABLE II. Numerical values of some elaborate parameters defined in Sec. II for Ge and Si.

| | Ge | Si |
|---|---|---|
| $s$ (°K) | 22 | 78 |
| $b$ (m$^{-3}$sec$^{-1}$) | $3.5\times10^{-12}\sqrt{T}$ | $4.0\times10^{-13}\sqrt{T}$ |
| $N_{0x}$(part/cm$^3$) | $7.3\times10^{15}T^{3/2}e^{-23/T}$ | $2.7\times10^{16}T^{3/2}e^{-80/T}$ |
| | $2.6\times10^{14}$ (at 4.2 °K) | $2.9\times10^{14}$ (at 10 °K) |
| | $2.1\times10^{11}$ (at 2 °K) | $3.2\times10^{-1}$(!) (at 2 °K) |
| $\tau_c$ (sec) | $3.9\times10^{-11}e^{23/T}/T^2$ | $0.91\times10^{-10}e^{80/T}/T^2$ |
| | $5.3\times10^{-10}$ (at 4.3 °K) | $2.7\times10^{-9}$ (at 10 °K) |
| | $9.6\times10^{-7}$ (at 2 °K) | $5.3\times10^{6}$(!) (at 2 °K) |
| $r_{\min}$ | 630 (at 4.2 °K) | 24 (at 10 °K) |
| | 21(25) (at 2 °K) | $\sim10^{-3}$(!) (at 2 °K) |
| $R_{\min}-1$ | $1.7\times10^{-2}$ (at 4.2 °K) | 0.65 (at 10 °K) |
| | 1.0(1.1) (at 2 °K) | $\sim10^{7}$ for $r=1$ (!) (at 2 °K) |
| $j_0/N_{0x}$(sec$^{-1}$) | $5.2\times10^{10}(R_x/\ln R_x)^2$ (at 4.2 °K) | $2.2\times10^{10}(R_x/\ln R_x)^2$ (at 10 °K) |
| | $1.3\times10^{8}(R_x/\ln R_x)^2$ (at 2 °K) | $2.9\times10^{-4}(R_x/\ln R_x)^2$ (at 2 °K) |
| $J/j_0$ | $e^{-72/\ln^2 R_x}$ (at 4.2 °K) | $e^{-237/\ln^2 R_x}$ (at 10 °K) |
| | $e^{-665/\ln^2 R_x}$ (at 2 °K) | $e^{-30\,000/\ln^2 R_x}$ (!) (at 2 °K) |

TABLE III. Variations with the supersaturation $R_x$ of the stable size $n_x$ and unstable size $n^*$, of the nucleation current $J$ over the prefactor $j_0$, and of the nucleation rate time $\tau_\nu$ for Ge at $T=4.2$ °K and $T=2$ °K.

| $T$ (°K) | 4.2 | 4.2 | 4.2 | 4.2 | 2 | 2 | 2 |
|---|---|---|---|---|---|---|---|
| $R_x$ | 2 | 3 | 4 | 5 | 10 | 100 | 500 |
| $n_x^{1/3}$ | $0.75\times10^5$ | $1.5\times10^5$ | $2.2\times10^5$ | $3\times10^5$ | 380 | $4.2\times10^3$ | $10^4$ |
| $n^*$ | 420 | 100 | 51 | 33 | 110 | 14 | 5 |
| $J/j_0$ | $8.3\times10^{-66}$ | $1.2\times10^{-26}$ | $5.4\times10^{-17}$ | $8.5\times10^{-13}$ | $3.4\times10^{-55}$ | $2.4\times10^{-14}$ | $3.3\times10^{8}$ |
| $\tau_\nu$ (sec) | $3\times10^{36}$ | $2\times10^{-3}$ | $3\times10^{-13}$ | $1.3\times10^{-17}$ | $2\times10^{36}$ | $6.6\times10^{-8}$ | $3.8\times10^{-16}$ |

exciton density reaches a certain value $N_\nu$, the amount of drops created for $N_x<N_\nu$ is negligibly small and if the nucleation current is "sizable" for $R_\nu$, a further increase of $N_x$, implying a very large increase of $J$, is not possible because of the conservation equation (1).

This remark leads to expanding $J$ around a given value $N_\nu$ (the choice of the appropriate $N_\nu$ will be discussed later; for the moment, it is said that $N_\nu$ will have to be close to the maximum value of $N_x$):

$$J\simeq j_0\exp\left(-\frac{\tilde{s}^3}{2\ln^3 R_\nu}\right)\left(1+\frac{\tilde{s}^3}{\ln^3 R_\nu}\frac{N_x-N_\nu}{N_\nu}+\cdots\right)$$

$$\simeq J_\nu n_\nu^*(N_x-N_\nu')/N_\nu,$$

where $N_\nu'=N_\nu(1-n_\nu^{*-1})\sim N_\nu$ and $J_\nu, n_\nu^*$, and $n_\nu$ are the nucleation current, the critical size, and the stable size at $N_\nu$.

Here *we approximate J by its tangent at* $N_\nu$ (see Fig. 2):

$$J\sim J_\nu n_\nu^*\frac{N_x-N_\nu'}{N_\nu}\Theta(N_x-N_\nu'). \tag{9}$$

This expression includes the facts that if $N_x$ reaches $N_\nu'$, the number of drops created previously is very small and if $N_x$ continues to increase, $J$ will increase extremely rapidly, because the slope of the curve is very sharp.

(a) For $N_x<N_\nu'$, and $J=0$, Eq. (8) reduces to the simple evolution of an exciton gas under the laser excitation; we will take it in the form

$$N_o(t)=N_o(1-e^{-t/\tau_L}), \tag{10}$$

with $\tau_L$ being the laser rise time. $N_x$ reaches $N_\nu'$ at time $t_\nu$; then the nucleation starts.

(b) In order to put Eq. (8) in a solvable form for $J\neq0$, one needs to do a few simplifications. Each time $J$ is different from zero, the value of $N_x$ will be close to $N_\nu$ (i.e., almost constant), so that one can replace $n^*$ by $n_\nu^*$, $r_{t'}(t)$ by [see Eq. (5)] $r_\nu[1-\exp[(t'-t)/3\tau_d]]$, and $(R_x-e^{\tilde{s}/r})$ by $(R_\nu-1)$. These last two simplifications make the critical embryo grow faster than they really do. This transforms Eq. (8), in the following equation for the change in the supersaturation

$$\Delta R=R_x-R_\nu'=N_x/N_{ox}-N_\nu'/N_{ox}$$

during the nucleation process (i.e., for $\Delta R>0$)

$$\frac{\Delta R_0}{\tau_x}-\frac{R_0}{\tau_x}\exp[-(\tilde{t}+t_\nu)/\tau_L]$$
$$=\Delta\dot{R}+\Delta R\left(\frac{1}{\tau_x}+\frac{1}{\tau_\nu}\frac{n_\nu^*}{n_\nu}\right)$$
$$+\frac{1}{\tau_\nu\tau_d}\int_0^{\tilde{t}}\Delta R(\tilde{t}')\{1-\exp[(\tilde{t}'-\tilde{t})/3\tau_d]\}^2\,d\tilde{t}', \tag{11}$$

where $R_0=N_0/N_{0x}$.

We have rescaled the origin of time $t=\tilde{t}+t_\nu$ and *introduced a nucleation rate time* $\tau_\nu$ which depends on the nucleation current $J_\nu$ (see Fig. 2) as

$$1/\tau_\nu=J_\nu n_\nu^* n_\nu/N_\nu. \tag{12}$$

Four characteristic times appear in Eq. (11): the laser rise time $\tau_L$ (which will play a role if $t_\nu$ is not much larger than $\tau_L$, i.e., the laser excitation still increases during the nucleation), the exciton lifetime $\tau_x$, the drops rise time $3\tau_d$, and the nucleation rate time $\tau_\nu$. One expects the behavior of the solution to differ if $\tau_\nu\ll\tau_x$, $\tau_d$ or $\tau_\nu\gg\tau_x$, $\tau_d$. $\tau_\nu$ is very sensitive to $R_\nu$ as shown in Table III, because the number of critical embryos $N^*$ ready

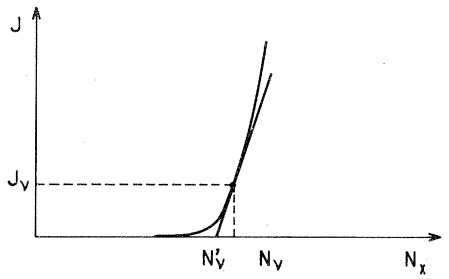


FIG. 2. Linear approximation for the nucleation current.

to pass the neck size every $\tau'$ seconds increases very fast with $R_\nu$. The general form of the solution of Eq. (11) is

$$\Delta R(\tilde{t}) = \Delta R_0\left(a_L e^{-\tilde{t}/\tau_L} + \sum_{i=1}^{4} b_i e^{-\tilde{t}/T_i}\right). \tag{13}$$

Feeding Eq. (11) with (13) and writing that the equation is true for any $\tilde{t}$, one gets a fourth-order equation for the $T_i$,

$$\frac{\frac{1}{T}\left(\frac{1}{T}-\frac{1}{3\tau_d}\right)\left(\frac{1}{T}-\frac{2}{3\tau_d}\right)\left(\frac{1}{T}-\frac{1}{\tau_x}\right)}{\frac{n_\nu^*}{n_\nu}\frac{1}{T}\left(\frac{1}{T}-\frac{1}{3\tau_d}\right)\left(\frac{1}{T}-\frac{2}{3\tau_d}\right)-\frac{6}{(3\tau_d)^3}} = \frac{1}{\tau_\nu}, \tag{14}$$

and four more equations between $a_L$ and the $b_i$ which will determine these five coefficients if one adds the initial condition $\Delta R(\tilde{t}=0)=0$. The left-hand side of Eq. (14) is shown as a function of $1/T$ in Fig. 3.

The intersections of that curve with $1/\tau_\nu$ lead to roughly three regimes for the solutions $T_i$.

If $\tau_\nu$ is very large (compared to $\tau_x, \tau_d$), i.e., the nucleation current is very small, then $1/\tau_\nu$ is almost zero, so that Eq. (14) has four real solutions:

$$\frac{1}{T_1}\simeq\frac{\tau_x}{\tau_\nu\tau_d},\quad \frac{1}{T_2}\simeq\frac{1}{3\tau_d},\quad \frac{1}{T_3}\simeq\frac{2}{3\tau_d},\quad \frac{1}{T_4}\simeq\frac{1}{\tau_x}. \tag{15}$$

If $\tau_\nu$ is very small, Eq. (14) has two real and two imaginary solutions:

$$\frac{1}{T'_\phi}=\frac{1}{\tau_{d\nu}e^{i\phi}}$$

with

$$\phi=0,\ \frac{2\pi}{3},\ \frac{4\pi}{3}\ \text{and}\ \tau_{d\nu}=3\tau_d\left(\frac{n_\nu^*}{6n_\nu}\right)^{1/3}, \tag{16}$$

$$\frac{1}{T'_4}=\frac{1}{\tau_\nu}.$$

When $\tau_\nu$ is neither very large nor very small, the four solutions are imaginary. Close to the small $\tau_\nu$ region, the $T_i$ have the simple expression

$$\frac{1}{T_{\phi_n}}=\frac{1}{T_0 e^{i\phi_n}}\ \text{with}\ \phi=\frac{\pi}{4}+n\frac{\pi}{2},\ \text{and}$$

$$T_0^4=\tfrac{9}{2}\tau_\nu\tau_d^3,\ n=1,2,3,4. \tag{17}$$

Turning to the shape of $R_x$ we find that *in the slow-nucleation regime* ($\tau_\nu$ very large), the exciton density rises to to $N_0$ in the time $\tau_x$ as if there were no drops because the amount of drops, created during $\tau_x$, is negligible. But, if one waits long enough, the amount of drops will become sizable and $R_x$ will finally decrease down to $R'_\nu$,

$$\Delta R = \Delta R_0 e^{-t\tau_x/\tau_\nu\tau_d}, \tag{18}$$

where the nucleation stops (due to the approximation used for $J$ which is zero for $N_x < N'_\nu$). This produces an extremely slow rise time $\sim T_1 \sim \tau_\nu\tau_d/\tau_x$ of the amount of e-h in drops and of the number of drops [see Fig. 4(a)].

*In the extremely fast-nucleation regime* ($\tau_\nu$ very small), the imaginary values of $T_i$ produce a maximum for $N_x$ very close to $N'_\nu$. The time during which the nucleation takes place (which corre-

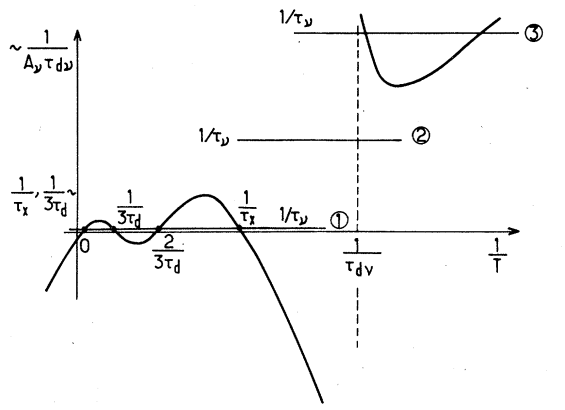


FIG. 3. Graphical solutions for the time $T_i$ of Eq. (14).

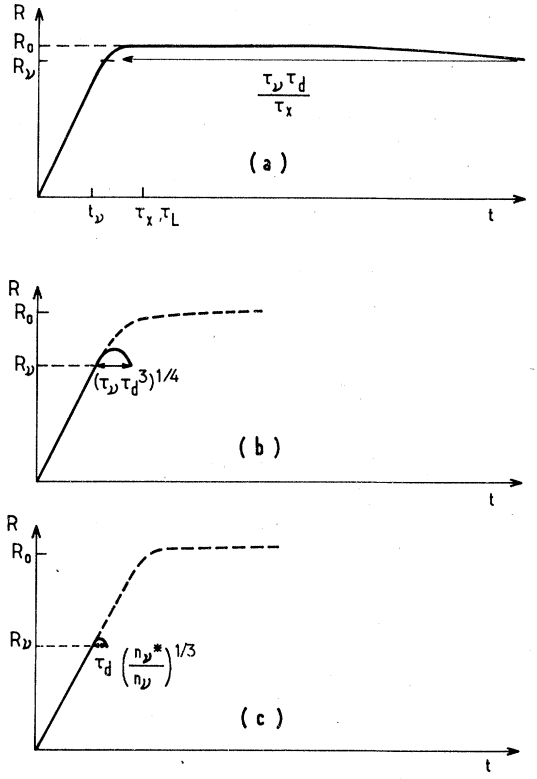


FIG. 4. Exciton supersaturation $R = N_x/N_{0x}$ as a function of time in the slow- (a), fast- (b) and very fast- (c) nucleation regime, obtained for increasing excitation $R_0 = N_0/N_{0x}$.

sponds to $\Delta R > 0$) is of the order $\tau_{d\nu}$. It is much shorter than the time $3\tau_d$ necessary for a drop to grow, so that the exciton density will present an overshoot due to the further growth of the baby drops.

A similar behavior is found in *the fast-nucleation* (or intermediate) *regime*:

$$\Delta R = \Delta R_0 \frac{T_0^2}{2\tau_x \tau_L} \sin \frac{t}{\sqrt{2}\, T_0} (e^{t/\sqrt{2}T_0} - e^{-t/\sqrt{2}T_0})$$

$$(\text{if } T_0 < \tau_L).$$

The nucleation lasts $T_0$ and the maximum of the exciton density is also very close to $N_\nu'$ [Fig. 4(b)].

Let us now turn to *the appropriate choice of* $N_\nu$. For that we follow the increase of $N_x$. If $N_\nu$ is chosen very low, the solution of the slow-nucleation regime implies that $N_x$ will continue to increase up to $N_0$. If the slow-nucleation regime remains up to $N_0$, then it will be the one observed with the characteristics of the size and rise time calculated for $N_\nu \sim N_0$ (as $N_\nu$ must be chosen close to the maximum).

If $N_0$ is high enough so that in the increase of $N_x$ up to $N_0$, one reaches a regime corresponding to the "intermediate" one, $N_x$ will have a maximum close to $N_\nu'$ and then decrease so that no higher value of $N_x$ will be reached, and as a consequence, the "extremely fast" nucleation regime will never exist. $N_x$ will in fact, stop at a value where an imaginary solution for $T_i$ exists (i.e., $\tau_\nu \geq \tau_x, 3\tau_d$) which produces an oscillatory solution (i.e., a maximum) for $N_x$. This condition is independent of $N_0$ and so the maximum of $N_x$ will not depend on the excitation.

(c) *After the nucleation takes place*, in the case of fast nucleation, the exciton density continues to decrease due to the growth of a constant number $\mathfrak{N}$ of the baby drops; this produces an overshoot in the observed $N_x$ with a characteristic time $3\tau_d$ due to the growth of the drops. This growth will be studied more precisely in Sec. V.

## IV. SOLUTION FOR A SLOW-NUCLEATION REGIME

We have found for long nucleation ratetime $\tau_\nu$ defined by Eq. (12), the exciton density rises first up to $N_0$, as if there were no drops, and then decreases slowly due to the slow formation of drops according to $N_x \sim \exp(-t\tau_x/\tau_\nu\tau_d)$. This can be obtained simply, noting that in such a regime the system is always very close to equilibrium. In that case, one can neglect in the conservation equation (1) the change with time of $N_x$ and $D$, so that Eq. (1) reduces to

$$\frac{N_0}{\tau_x} = \frac{N_x}{\tau_d} + \frac{n_x}{\tau_d} \int_0^t J(t')\, dt', \tag{19}$$

all the drops having the same size $n_x$. The laser excitation just compensates the decay in the exciton gas and in the drops. We can then solve Eq. (19) in a more appropriate way. By derivation of Eq. (19), one obtains

$$0 = \frac{\dot{N}_x}{\tau_x} + \frac{\dot{n}_x}{\tau_d} \int_0^t J(t')\, dt' + \frac{n_x}{\tau_d} J(N_x). \tag{20}$$

The integral can be evaluated from Eq. (19), and $\dot{n}_x$ from the fact that

$$n_x^{1/3} = (\tau_d/\tau_c)(N_x/N_{0x} - 1).$$

This transforms Eq. (20) into a first-order differential equation for $N_x$:

$$\dot{N}_x \left(1 + 3\frac{N_0 - N_x}{N_x - N_{0x}}\right) + \frac{\tau_x \tau_d^2}{\tau_c^3} \left(\frac{N_x}{N_{0x}} - 1\right)^3 J(N) = 0,$$

whose solution is readily

$$t\frac{\tau_x}{\tau_d} = \int_{N_x}^{N_0} \frac{1 + 3\,\dfrac{N_0 - N}{N - N_{0x}}}{\dfrac{\tau_d^3}{\tau_c^3}\left(\dfrac{N}{N_{0x}} - 1\right)^3 J(N)}\, dN.$$

One can evaluate this integral for $N_x$ close to $N_0$ using the fact that $J(N)$ is the quantity which mostly varies, so that one can replace $N$ by $N_0$ except in $J$. Noting from Eqs. (6) and (3) that

$$\frac{d(1/J)}{dN} = -\frac{1}{J}\frac{n^*}{N}, \tag{21}$$

one gets

$$t\frac{\tau_x}{\tau_d} \sim \int_{N_x}^{N_0} \frac{dN}{n_0 J} \sim \frac{N_0}{n_0^* n_0} \int_{N_0}^{N_x} d\left(\frac{1}{J}\right),$$

with $n_0$ and $n_0^*$ being the stable and unstable drops size at $N_0$. By integration we find

$$\frac{1}{J(N_x)} - \frac{1}{J(N_0)} \simeq \frac{n_0 n_0^*}{N_0} \frac{\tau_x}{\tau_d} t.$$

*The number of drops and the amount of* e-h *in drops* are obtained from the above expression of $J$:

$$\mathfrak{N} = \int_0^t J(t')\, dt' = \int_0^t dt' \frac{J_0}{1 + (\tau_x t'/\tau_0 \tau_d)}$$

$$= \frac{N_0}{n_0 n_0^*} \frac{\tau_d}{\tau_x} \ln\left(1 + \frac{\tau_x \tau}{\tau_0 \tau_d}\right),$$

$$D \simeq n_0 \mathfrak{N}, \tag{22}$$

where $J_0 = J(N_0)$ and $\tau_0$ is the nucleation ratetime $N_0/n_0 n_0^* J_0$ [as introduced in Eq. (12)]. We find that the amount of e-h liquid increases on a very long time scale of the order $\tau_0\tau_d/\tau_x$ and that when the amount of liquid is detectable, it varies with the excitation and time as

$$\sim R_0 \ln^3 R_0 \ln(t\tau_x/\tau_0\tau_d + 1)\,.$$

The $\ln t$ dependence for the increase of liquid with time was already obtained by Westerwelt.[5] One gets $N_x$ either directly from the conservation equation (19), or from (21) using the dependence of $J(N_x)$ in $N_x$ where we would set

$$\ln\left(1+\frac{\tau_x t}{\tau_0\tau_d}\right) \ll \frac{\tilde{s}^3}{2\ln^2 R_0}$$

consistent with the approximation made to obtain (21). We find that

$$N_x = N_0\left[1-\frac{1}{n_0^*}\ln\left(1+\frac{\tau_x t}{\tau_0\tau_d}\right)\right]. \tag{23}$$

Note that for very small $\tau_x t/\tau_0\tau_d$, Eq. (23) gives $N_x - N_0 \sim J_0 n_0(\tau_x/\tau_d)t$ which is nothing other than the expansion of the solution (18) of Sec. III for small $t$. The behavior of the exciton density and the amount of e-h liquid is shown in Fig. 5 in the case of a slow-nucleation regime. The rise time of $D$ is ruled by $\tau_0$. From Table III, we see that at 4.2 °K for $R_0 = 2$, the nucleation rate time $\tau_0$ is $10^{18}$ times the age of the universe (!) but for $R_0 = 3$, it is only a msec and for $R = 4$, it is already much smaller than the lifetimes. So, close to the cross-over region a very small change in $R_0$ produces a very large change in the observed rise time of the e-h liquid, in agreement with the Shah *et al.*[8] results. In that region with a 10% change in $R_0$, one goes from a slow-nucleation regime to a fast-nucleation one. This may have some consequences in the case of the usual experimental conditions, where the excitation is far from being homogeneous. We also see from Table III that the amount of electron hole in a drop produced in the slow-nucleation regime should be huge ($\sim 10^{15}$), if the processes controlling the drops size are the ones considered in Eq. (2). There is no experimental report on the existence of such large drops. Most probably they explode before reaching such a size, due to the interaction between the e-h liquid and the phonons emitted from the e-h decay in the drop as proposed by Keldysh.[13] To take this effect into account, one can simply replace in Eq. (19) $n_x$ by the limit size $n_{\mathrm{lim}}$, which will change $\tau_0$ by a factor $n_x/n_{\mathrm{lim}}$.

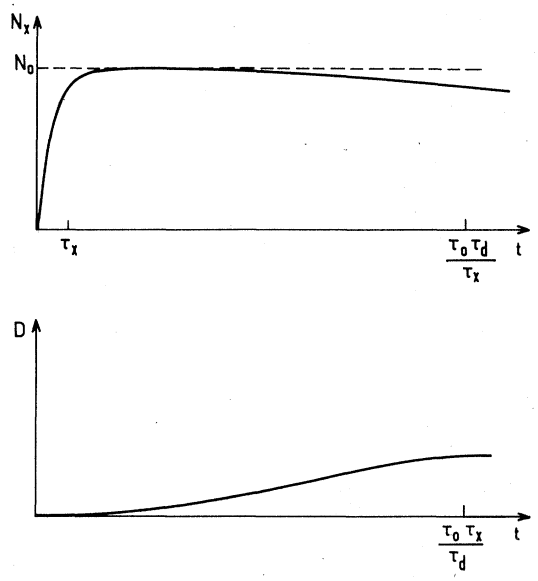


FIG. 5. Time dependence of the exciton density and amount of e-h liquid in the slow-nucleation regime.

Table III also shows that the supersaturation needed to reach the fast-nucleation regime increases significantly when the temperature decreases.

## V. SOLUTION FOR A FAST-NUCLEATION REGIME

(a) In the slow-nucleation regime we kept only the decay terms in the conservation equation (1). Similarly, in the fast-nucleation regime, we only keep, during the nucleation, the change in $N_x$ and $D$, the evolution of the system being expected much faster than $\tau_x$ and $\tau_d$ so that one can neglect the decay of the e-h.

In that regime, during the nucleation, Eq. (1) reduces to

$$N_0(t)/\tau_x \simeq \dot{N}_x + \dot{D}\,. \tag{24}$$

We know from Sec. III that the exciton density has a maximum $N_M$, and that the nucleation takes place around this maximum. Except at the very beginning, the drop radius increases [cf. Eq. (2)] with a velocity

$$\dot{r} \sim \dot{r}_M \sim (1/3\tau_c)(R_M - 1) = r_M/3\tau_d$$

($r_M^3 = n_M$ being the stable size at $N_M$). During the nucleation one can approximate $r_{t'}(t)$ by $\dot{r}_M(t-t')$, in agreement with the fact that $t-t'$ is small compared to $3\tau_d$.

We also know from Sec. III that the amount of e-h going into the critical embryos $Jn^*$ is only important in the very fast-nucleation regime which cannot be reached. Using Eq. (7), we finally rewrite Eq. (24) as

$$\frac{N_0(t)}{\tau_x} = \dot{N}_x + \int_0^t J(t')3r_{t'}^2(t)\dot{r}\,dt'$$

$$\sim \dot{N}_x + \alpha\int_0^t J(t')(t-t')^2\,dt'\,, \tag{25}$$

with $\alpha = 3\dot{r}_M^3 = n_M/9\tau_d^3$. For $N_0(t)$ constant or linear in $t$, Eq. (25) is in fact a fourth-order differential equation for $N$:

$$0 = \dddot{N} + 2\alpha J(N)\,,$$

which can be integrated once, using Eq. (21), as

$$\dot{N}\dddot{N} - \tfrac{1}{2}\ddot{N}^2 + 2\alpha(N/n^*)J = 0\,, \tag{26}$$

but no more. However, this equation gives an exact relation between the curvature $\ddot{N}$ at the maximum and the value of the maximum $N_M$. One can then think to expand $J$ around this maximum reached at time $t_M$:

$$J(t) = J_M + \left(\frac{dJ}{dN}\dot{N}\right)_{\max}(t - t_M) + \tfrac{1}{2}\left(\frac{d^2J}{dN^2}\dot{N}^2 + \frac{dJ}{dN}\ddot{N}\right)_{\max}(t - t_M)^2 + \cdots\,.$$

With $\dot{N} = 0$ and using Eq. (21), this gives

$$J(t) \simeq J_M(1 - u^2/\delta^2)\,,$$

with $t = u + t_M$ and

$$u^2 < \delta^2 = \frac{2N_M}{|\ddot{N}_M|\, n_M^*}\,. \tag{27}$$

Going back to Eq. (25) we have, for $t < t_M - \delta$, no drop and

$$N_x = \int_0^t \frac{N_0(t')}{\tau_x}\,dt'\,. \tag{28}$$

For $t_M - \delta < t < t_M + \delta$, Eq. (25) gives

$$\frac{N_0(t)}{\tau_x} = \dot{N}_x + \alpha J_M \times\left(\frac{(u+\delta)^3}{3} - \frac{u^5}{30\delta^2} - \frac{u^2\delta}{3} - \frac{u\delta^2}{2} - \frac{\delta^3}{5}\right). \tag{29}$$

From these two equations, the derivative, and the integration of (29), one can obtain the three relations which will determine the position of the maximum $(N_M, t_M)$ and the time $\delta$ of the nucleation.

Eq. (29) gives at the maximum, i.e., for $u = 0$:

$$N_0(t_M)/\tau_x = \tfrac{2}{15}\,\alpha\, J_M \delta^3\,. \tag{30}$$

Its derivative, also taken for $u = 0$, gives

$$\dot{N}_0(t_M)/\tau_x = \ddot{N}_M + \alpha\, J_M \tfrac{1}{2}\delta^2\,.$$

Writing $\ddot{N}_M$ in terms of $\delta^2$ from Eq. (27), this gives a second-order equation for $\delta^2$ whose solution is

$$\delta^2 = \frac{1}{\alpha\, J_M}\left\{\frac{\dot{N}_0(t_M)}{\tau_x} + \left[\left(\frac{\dot{N}_0^2(t_M)}{\tau_x}\right)^2 + 4\alpha\, J_M \frac{N_M}{n_M^*}\right]^{1/2}\right\}. \tag{31}$$

Finally, if one integrates Eq. (29), one gets $N_x(u)$ which, for $u = -\delta$, should coincide with the expression (28) of $N_x$ calculated at time $t_M - \delta$. This leads to the relation

$$N_M = \int_0^{t_M} \frac{N_0(t)}{\tau_x}\,dt - \alpha J_M \frac{\delta^4}{36}\,. \tag{32}$$

The solution of Eqs. (30)–(32) will depend on whether $\dot{N}_0(t_M)$ is zero or not, i.e., if the laser rise time is very short or not. Before solving this set of equation in both cases, one can calculate the number of drops:

$$\mathfrak{N} = \int_{-\delta}^{+\delta} J_M\left(1 - \frac{u^2}{\delta^2}\right)du = \tfrac{4}{3}\, J_M \delta\,. \tag{33}$$

Some more drops are surely created after $t_M + \delta$ but their amount is negligibly small and will not affect $\mathfrak{N}$.

The baby drops, created between $t_M - \delta$ and $t_M + \delta$, will grow with a characteristic time $3\tau_d$, and a steady state is reached after a time larger than $\tau_L$, $\tau_x$, $3\tau_d$. It will correspond to an exciton density $N_{x\infty}$ and $\mathfrak{N}$ drops with size $r_{x\infty}$ which verify the conservation equation written in that limit:

$$N_0/\tau_x = N_{x\infty}/\tau_x + (r_{x\infty}^3/\tau_d)\mathfrak{N}\,,$$

$r_{x\infty}$ being the stable size at $N_{x\infty}$. Using Eq. (3) and noting that $N_0 - N_{x\infty}$ is probably close to $N_0 - N_{0x}$, we obtain $r_{x\infty}$ and $N_{x\infty}$ as

$$r_{x\infty} \sim \left(\frac{\tau_d(N_0 - N_{0x})}{\tau_x \mathfrak{N}}\right)^{1/3}. \tag{34}$$

(b) *Let us first consider a very short laser rise time* $\tau_L$ so that for $t \sim t_M$, $N_0(t) \sim N_0$. In that case, the solution of Eq. (30)–(32), expressed in terms of the nucleation ratetime $\tau_M = N_M/J_M n_M^* n_M$, is

$$\tau_M = \tau_x\left(\frac{2^{10}}{3^6 5^4}\right)\left(\frac{\tau_x}{\tau_d}\right)^3 \frac{1}{n_M^{*4}}\left(\frac{N_M}{N_0}\right)^4,$$

$$N_M = \frac{N_0}{\tau_x}\,\frac{t_M}{1 + (1/9n_M^*)} \simeq \frac{N_0}{\tau_x}\,t_M\,, \tag{35}$$

$$\delta \sim (36\tau_d^3 \tau_M)^{1/4}\,.$$

The first relation is an implicit equation for $N_M$. Its solution is given in Table IV. It is interesting to note that the maximum $N_M$ varies very slowly with $N_0$. For that, one differentiates the logarithm of this equation, noting that the change with $N_M$ comes essentially from

$$\tau_M \sim \exp(+\,\tilde{s}^3/2\ln^2 R_M)\,.$$

This gives

$$\Delta N_M/N_M \sim (4/n_M^*)(\Delta N_0/N_0)\,, \tag{36}$$

which is much smaller than $\Delta N_0/N_0$ for the usual values of $n_M^*$, so that the maximum of $N_x$ does not change very much with the laser excitation $N_0$.

The second relation tells that the exciton density rises as if there were no drops up to $N_M$ where the drops appear. The larger $N_0$ is, the sooner the e-h liquid exists (see Fig. 6). The last equation gives the time $2\delta$ during which the nucleation takes place. It agrees with the results in the fast-nucleation regime of Sec. III. At that point, one

TABLE IV. Variations with the excitation $R_0$ of the maximum of the exciton supersaturation and of the nucleation rate time $\tau_M$, for short and long laser rise time.

| | | | | |
|---|---|---|---|---|
| $R_0$ if $\tau_L \ll \tau_x$ | 7.5 | 11 | 15 | 73 |
| $R_m$ | 4.8 | 5 | 5.2 | 6 |
| $n_M^*$ | 36 | 33 | 31 | 24 |
| $\tau_M$ (sec) | $5.8\times10^{-17}$ | $1.3\times10^{-17}$ | $3.6\times10^{-18}$ | $4.7\times10^{-20}$ |
| $R_0$ if $\tau_L = \tau_x$ | 12 | 24 | 43 | (890) |

has to check that the time $2\delta$ of the nucleation is larger than the minimum time to reach thermalization among the embryos, condition implicitly assumed when one uses the nucleation current. The collection time $\tau_c$ increases when $T$ decreases, but this effect is partly compensated by the fact that the supersaturation necessary to reach the fast-nucleation regime increases when $T$ decreases (for Ge $R_M \sim 5$ at 4.7 °K, $R_M \sim 500$ at 2 °K).

Table V shows that, for typical condition in the fast nucleation, the thermalization assumption is justified, but extremely large $N_0$ might change $R_M$ enough to produce much smaller $\tau_M$, so that $\delta$ becomes of the order $\tau_{\text{inf}}$ and the theory loses its meaning. Anyway, in the case of extremely large $N_0$, we will see at the end of this section that the number of drops is no longer controlled by the nucleation process exposed above.

Turning to the steady-state quantities, we find from Eq. (33) that the number of drops increases like $N_0^3$, in agreement with Keldysh's[7] result:

$$\mathfrak{N} \simeq R_0^3 \left[ 10 \frac{n_M^*}{R_M^2 (R_M - 1)^3} \left( \frac{3\tau_c}{\tau_x} \right)^3 \right] N_{0x} . \tag{37}$$

The term in the bracket, depending on $N_M$, varies very slowly with $N_0$ as $N_M$ does. From Eq. (34), we find that the radius of the drops decreases like $(R_0 - 1)^{1/3}/R_0$:

$$r_{x\infty} \simeq \frac{(R_0 - 1)^{1/3}}{R_0} \left[ \frac{(\tau_d \tau_x^2)^{1/3}}{3\tau_c} \frac{(R_M - 1) R_M^{2/3}}{n_M^{*2/3}} \right], \tag{38}$$

and the exciton density is very close to the thermodynamical one:

$$R_{x\infty} = 1 + \frac{(R_0 - 1)^{1/3}}{R_0} \left[ \left( \frac{\tau_x}{\tau_d} \right)^{2/3} \frac{1}{6} (R_M - 1) \frac{R_M^{2/3}}{n_M^{*2/3}} \right] \sim 1 . \tag{39}$$

TABLE V. Comparison between the time of the nucleation $2\delta$ and the thermalization time $\tau_{\text{inf}}$.

| $T$ (°K) | $R_0$ | $R_M$ | $\tau_M$ (sec) | $2\delta$ (sec) | $\tau_{\text{inf}}$ (sec) |
|---|---|---|---|---|---|
| 4.2 | 11 | 5 | $1.3\times10^{-17}$ | $1.5\times10^{-7}$ | $10^{-9}$ |
| 2 | 2900 | 500 | $4\times10^{-16}$ | $4\times10^{-7}$ | $10^{-8}$ |

(c) *If the laser is still rising during the nucleation*, the preceding dependences on $N_0$ of the number of drops and their size at steady-state equilibrium are no longer valid. In that case, one can take $N_0(t) \sim N_0 t/\tau_L$. One shows that the existence of a maximum for $N_x$ implies that [see Eq. (31)] the current $J_M$ is such that

$$4\alpha J_M N_M / n_M^* \gg (N_0/\tau_x \tau_L)^2$$

(which can be checked at the end).

From Eqs. (30)–(32), we extract that the nucleation ratetime $\tau_M$ varies like $N_0^{-2}$ instead of $N_0^{-4}$, that the density of the maximum still varies very slowly with $N_0$, that the nucleation lasts a little more than in Eq. (35), but still about the same as in Sec. III, and that the exciton density still rises to $N_M$ as if there were no drops:

$$\tau_M = \tau_x \left( \frac{2^{10}}{3^6 5^4} \right) \left( \frac{\tau_x}{\tau_d} \right)^3 \left( \frac{\tau_L}{\tau_x} \right)^2 \left( \frac{N_M}{N_0} \right)^2 ,$$

$$\delta \sim 10 (\tau_d^3 \tau_M)^{1/4} , \tag{40}$$

$$N_M = \frac{N_0}{\tau_x \tau_L} \frac{t_M^2}{2} \frac{1}{1 + (2/36 n_M^*)} \simeq \frac{N_0}{\tau_x \tau_L} \frac{t_M^2}{2} .$$

The relation between $N_M$ and $N_0$ is given in Table IV. One then deduces that the number of drops increases like $R_0^{3/2}$ in agreement with Keldysh[7] i.e., more slowly than for a very short laser rise time:

$$\mathfrak{N} = R_0^{3/2} \left[ \left( \frac{3\tau_c}{(\tau_x \tau_L)^{1/2}} \right)^3 \frac{n_M^*}{(R_M - 1)^3 (R_M)^{1/2}} 25^2 \right] N_{0x} \tag{41}$$

and that their size at equilibrium decreases with $R_0$ also more slowly:

$$r_{x\infty} = \frac{(R_0 - 1)^{1/3}}{R_0^{1/2}} \times \left[ \frac{(\tau_d \tau_x^2)^{1/3}}{3\tau_c} \left( \frac{\tau_L}{\tau_x} \right)^{1/2} \frac{(R_M - 1) R_M^{1/6}}{n_M^{*1/3} 5^{4/3}} \right], \tag{42}$$

the steady-state density being still very close to $N_{0x}$.

(d) In conclusion, in the fast-nucleation regime the exciton density increases up to a value $N_M$ almost as if there were no drops (see Fig. 6). The maximum

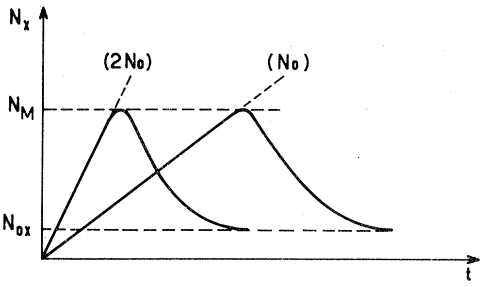


FIG. 6. Time dependence of the exciton density for two excitations in the fast-nucleation regime.

of the supersaturation $R_M$ is almost independent of the excitation, but it is very sensitive to the temperature. (Note that the exact numerical value of $R_M$ is affected by the choice of the prefactor $j_0$.) At $N_M$ baby drops are formed and their formation lasts a very short time, of the order $(\tau_d^3\tau_M)^{1/4}$. Then the exciton density starts to decrease. At steady state, after a time larger than $\tau_x$, $3\tau_d$, $\tau_L$, the supersaturation is very close to unity. Finally, one finds that the number of drops and their size depend on whether $N_M$ was reached during the laser rise or when its maximum was already reached.

One remarks that the larger the excitation, the smaller the drops (because the amount of baby drops formed during the nucleation is larger). So in order to get large drops, one should not use large excitation, but instead stay close to threshold. Similarly if one wants a large supersaturation at steady state, one should not use a very powerful laser, but instead stay very close to threshold.

(e) There remains one limit case that we have not considered in detail: what happens if too many drops are created during the nucleation, the excitation not being large enough to allow them to grow to a size larger than $r_{\min}$? From Eqs. (4) and (38) this corresponds, in the case of Ge at 4.2 °K, to an excitation $R_0$ larger than 20 (with our choice of $j_0$ hidden in $R_M$). There is obviously no possible steady state with that amount of drops.

From the conservation equation

$$N_0/\tau_x = \dot{N}_x + \mathfrak{N}\dot{n} + \mathfrak{N}n/\tau_d,$$

(where we have neglected the exciton decay), we see that the size of $\mathfrak{N}$ drops when $\dot{N}_x = \dot{n} = 0$ should be $n_\infty = N_0\tau_d/\mathfrak{N}\tau_x$. If $n_\infty < n_{\min}$, such a size is not an equilibrium one. The drops can continue to grow beyond $n_\infty$, as long as $\dot{N}_x$ can be negative. But this cannot last too long because $N_x$ will finally reach $N_{\min}$ where, due to Eq. (2) for the evolution on one drop, $\dot{n}$ becomes negative; then all the drops should shrink and disappear leading to an oscillatory situation. In fact, the drops size has a dispersion due in particular to the fact that they are created during a small but finite time $2\delta$. During the decrease of the exciton density, the smaller drops will shrink before the larger ones, producing an extra amount of excitons, which allow the larger ones to continue their growth. This leads to a final situation with drops at size $n_{\min}$, their number being

$$\mathfrak{N} \simeq N_0\tau_0/n_{\min}\tau_x. \tag{43}$$

But one can imagine an extreme situation when the nucleation time is so small that the drop-size dispersion is very narrow and the drops will become unstable almost altogether, leading to an oscillatory situation. This will happen very fast anyway and the system will finally relax to the preceding equilibrium state (43).

## VI. CONCLUSION

From the results on the slow- and fast-nucleation regimes, we deduce (Fig. 7) the dependences over the whole range of excitation $R_0 = N_0/N_{0x}$ at a

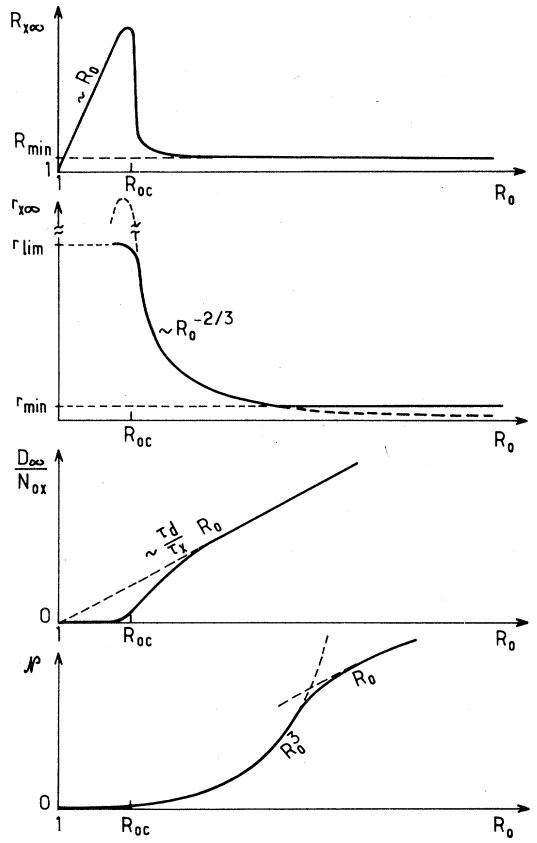


FIG. 7. Dependence over the whole range of excitation $R_0 = N_0/N_{0x}$ of the exciton density $N_{x\infty}$, drops radius $r_{x\infty}$, amount of e-h $D_\infty$ and density of drops $\mathfrak{N}$ at steady state for short laser rise time. For long laser rise time, the drops radius would decrease as $\sim R_0^{-1/6}$ and the density of drops would increase as $\sim R_0^{3/2}$.

given temperature of the exciton density $R_{x\infty}$, the amount of e-h liquid $D_\infty$, the density of drops $\mathfrak{N}$, and the radius $r_{x\infty}$ at steady state (i.e., after a reasonable amount of time).

As we have seen, the characteristics of the steady state are controlled by the nucleation process and have nothing to do with the minimum of the pseudofree energy,[3] which would lead to a pseudoequilibrium state eventually reached after an infinite time.

The "reasonable" amount of time after which a steady state is obtained is of the order sup $(\tau_L, \tau_x, 3\tau_d)$ in the fast-nucleation regime, but depends very much on the excitation in the slow-nucleation regime. More precisely, one needs to wait a time in the order of $\tau_0\tau_d/\tau_x$ which decreases close to threshold as the inverse of the nucleation current (calculated at $R_0$) in agreement with the Shah *et al.* results.[8]

The exciton supersaturation should increase in the case of homogeneous excitation, linearly with the excitation up to a maximum, where the nucleation starts to be fast (for $R_0 > R_{0c}$). Then, for higher excitation, the exciton density at steady state decreases to stay close to its minimum value.

The amount of e-h in the drops increases suddenly around $R_{0c}$. At higher excitation, the laser essentially compensates the e-h decay in drops $(N_0/\tau_x \sim D_\infty/\tau_d)$ so that $D_\infty$ increases linearly with $R_0$.

We have seen that in the slow-nucleation regime the drop size, controlled only by evaporation, collection, and decay, should be enormous. Including the phonons interaction, Keldysh[13] finds an upper limit for the drop size, which will be the size of the drops in that regime. Above $R_{0c}$, the drop radius decreases down to its minimum value; the way $r_{x\infty}$ decreases depends on whether or not the laser is still rising during the nucleation (because this affects the number of created drops). For a short rise time, $r_{x\infty}$ decreases as $(R_0-1)^{1/3}/R_0 \sim R_0^{-2/3}$ [from Eq. (39)], while for a long rise time the change is not as fast [Eq. (42)] giving $(R_0-1)^{1/3}/R_0^{1/2} \sim R_0^{-1/6}$] and the drops are larger. The comparison with the Bagaev *et al.*[7] experiments is not very easy because they measure $r_{x\infty}$ at constant excitation $N_0$ and various temperatures, while the analytical behavior of $r_{x\infty}$ in $T$ is not easy to extract from our theory. However, we note that an increase of $T$ corresponds to a decrease of $R_0$ if $N_0$ stays constant, because the thermodynamical density $N_{0x}$ would increase. We can then check (from their Fig. 10) that $r_{x\infty}(T)$ has qualitatively the inverse behavior of $r_{x\infty}(R_0)$. We can also see (in their Fig. 10) that an increase of $N_0$ produces a decrease of $r_{x\infty}$ as expected. Finally, we can check (in their Fig. 3) that, at a given $T$ or $R_0$, the drops radius is larger if the rise time is longer as predicted, but if $T$ increases (i.e., $R_0$ decreases) the radius tends to a unique value which does not depend on the laser rise time as expected.

Finally, we show on Fig. 7 the dependence of the density $\mathfrak{N}$ of drops with the excitation $R_0$ at a fixed temperature. The amount of drops increases first slowly as $R_0 \ln^3 R_0$ [see Eq. (22) of the slow-nucleation regime]. Above $R_{0c}$, it increases faster, but this increase depends on whether the laser excitation still increases during the nucleation or not. For a short rise time, $\mathfrak{N}$ increases as $R_0^3$, while for a long rise time $\mathfrak{N}$ changes only as $R_0^{3/2}$ [see Eqs. (37) and (41)], so that more drops are created if the rise time is short. At higher excitation, the size of the drops being fixed at its minimum value, the density of drops will finally tend to increase only linearly with $R_0$.

In Fig. 4 of Bagaev *et al.*[13] we check that more drops are created for a given excitation and temperature if the rise time is short. The dependences $\mathfrak{N}(1/T)$ (of their Figs. 6 and 9) look like our curve $\mathfrak{N}(R_0)$ with a clear change of curvature as expected ($R_0$ at constant $T$ changes as $1/T$ at constant $N_0$). In their Fig. 7,[7] we finally check that for $T = 3.6\,^\circ\mathrm{K}$ $\mathfrak{N}$ increases approximately as $R_0^3$ for short rise time and as $R_0^{3/2}$ for a long one, while the higher temperatures the increase is not as fast, although faster for short than for long rise time, probably because for the same $N_0$, $R_0$ is larger and we are closest to the linear region of the curve $\mathfrak{N}(R_0)$.

So the theory presented in this paper is in good agreement with the experiments, but we want to say that it does not cover the low-temperature region as outlined about silicon at $2\,^\circ\mathrm{K}$. For low temperature, we have already pointed out[4] that there is no more real phase separation between an exciton gas and e-h liquid in macroscopical drops but instead the number of embryos with $n$ e-h inside increases with $n$ from $n=1$ to a maximum for $n$ rather small. This is due to the fact that the temperature is so small that the evaporation is negligible and the decreasing part of the curve of Fig. 1 is all in the unphysical region $n<1$. The concept of critical embryo disappears and, by the way, all the nucleation theory based on the current of critical embryos passing the neck point. From Eq. (2) and Table II, we see that in order to have small embryos of e-h, one would need anyway an enormous supersaturation in agreement with recent experiments by Voisin *et al.*[14]

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$$(r_x - r)\left|\frac{r_x - r}{r - r^*}\right|^{r^*/r_x - r^*} = A e^{-t/3\tau_d},$$

where $A$ is constant obtained from the initial value of $r(t=0)$. For $r(t=0) > r^*$, and for $r_x \gg r^*$, this solution can be approximated by Eq. (5).
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