Kinetics of exciton condensation in germanium at high excitation levels

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The growth and decay kinetics of an electron-hole liquid, as well as the dependence of its volume on the excitation level, are considered theoretically and experimentally. It is shown that the repulsion between the droplets and intense recombination of the droplets on the surface of the crystal explain the results qualitatively. The experimentally observed deviations from the simple model of the interaction between the droplets is discussed.

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1. INTRODUCTION

The kinetics of exciton condensation in germanium crystals has been intensively studied in the past few years.[1] The main effects of metastability and supersaturation in an exciton system are determined, as is known, by the nucleation and growth of droplets from the exciton gas. At high excitation levels, when the volume of the electron-hole liquid (EHL) becomes appreciable, an analysis of exciton condensation must take into account also the repulsion between the droplets.[2]

Usually, when the crystals are surface-excited, high electron-hole pair densities are obtained by using thin samples, of thickness smaller than the exciton diffusion length. Under these conditions the surface state of the crystal can greatly influence the condensation kinetics. Gantmakher and Zverev[3] were the first to point out this circumstance, and proposed that intense recombination of the droplets on the surface prevented them from producing a high EHL density in thin pure-germanium crystals.

It is known that the lifetime of electron-hole pairs in thin germanium crystals at $T=300$ K decreases with increasing pumping, and amounts to 10^{-6}–10^{-7} sec at $n=10^{17}$–10^{18} cm^{-3} (Ref. 4). It can be assumed that at low temperatures, too, the emergence of the electron-hole droplets (EHD) with pair density $\approx 2\times 10^{17}$ cm^{-2} to the crystal surface will shorten their lifetime substantially. The emergence of the EHD to the surface can be due to mutual repulsion of the droplets, which was experimentally observed by Kavetskaya *et al.*[5]

Following Gantmakher and Zverev,[3] it was proposed that EHD can be possibly destroyed on the surface of a crystal by microwave breakdown of the excitons[6] and by their expulsion to the surface of the crystal by a flux of nonequilibrium phonons.[7] In the present paper we analyze, on the basis of a model proposed by Keldysh for the interaction between the droplets,[8] the growth and breakup of an EHL in the course of time, with allowance for the annihilation of the droplets on the crystal surface.

2. THEORETICAL TREATMENT

Keldysh[8] was the first to note that inasmuch as the EHD are sources of intense fluxes of nonequilibrium phonons, it follows that when the phonons are absorbed or scattered by the EHL they produce exactly the same kind of bulk forces that would be produced if this liquid were uniformly charged with charge density ρ. We use this idea in the present paper to analyze the kinetics of exciton condensation at sufficiently high excitation levels, when the influence of the phonon wind becomes decisive.

Following Keldysh,[8] we replace each EHD of volume V by a charge ρV. We assume further that EHD are uniformly produced in the volume of a thin infinite plate of germanium. When the light is applied, the EHD nuclei are produced mainly on the front of the exciting pulse, and their density remains subsequently practically constant.[9] In this case the growth of the volume of the EHL will be due to the growth of the volume of each droplet. The change of the droplet volume with time is described by a usual kinetic equation[10] from which, to simplify the analysis, we leave out the term that describes exciton evaporation from the EHD,

$$\frac{dV}{dt}=\frac{g}{n_0 N}-\frac{V}{\tau_0}, \tag{1}$$

where g is the rate of generation of electron-hole pairs per cm^3 and per second, τ_0 and N are the lifetime and density of the EHD, and $n_0=2\times 10^{17}$ cm^{-3} is the density of the electron-hole pairs in the EHD. The variation of the exciton density with time is not considered here, inasmuch as at high excitation levels only a small fraction of them is in the gas phase.

We determine the flux of the EHD to the sample surface as a result of mutual repulsion, assuming that the droplets recombine completely on the crystal surface. To this end we write down the continuity equation

$$\frac{\partial N}{\partial t}+\frac{\partial J}{\partial x}=0, \tag{2}$$

where the x axis is perpendicular to the crystal surface. The EHD flux to the surface is

$$J=\mu N E, \tag{3}$$

where $\mu=\rho\tau_p/Mn_0$ is the EHD mobility,[8] $\rho=10^2\, g^{1/2}$ cm$^{-3/2}$ sec^{-1} is the effective charge density of the droplet, $\tau_p=2\times 10^{-9}$ sec is the carrier-momentum relaxation in the EDC, and $M=4\times 10^{-28}\, g$ is the effective mass of the electron-hole pair. The effective field intensity E is determined from the Poisson equation

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$$\frac{\partial E}{\partial x}=4\pi N\rho V. \tag{4}$$

Since N is independent of x, we obtain

$$E=4\pi N\rho Vx, \tag{5}$$

$$J=4\pi\rho V\mu N^2x. \tag{6}$$

The change of the total EHL density $n=nVn_0$ with time is

$$\frac{\partial n}{\partial t}=Vn_0\frac{\partial N}{\partial t}+Nn_0\frac{\partial V}{\partial t}. \tag{7}$$

Taking (1), (2), and (6) into account we obtain ultimately

$$\frac{\partial n}{\partial t}+4\pi\mu n^2\frac{\rho}{n_0}+\frac{n}{\tau_0}=g. \tag{8}$$

Equation (8) differs from the usual kinetic equation by an added term that takes into account the EHD flux to the sample surface on account of the repulsion between the droplets. We examine now the solutions of Eq. (8) for several typical situations.

Stationary state. We introduce the notation

$$n'=g\tau_0,\quad n_1=g_1\tau_0=n_0/4\pi\mu\tau_0\rho.$$

In the stationary state, Eq. (8) has the solution

$$n_{st}=-\frac{1}{2}g_1+\left(\frac{1}{4}g_1^2+g_1g\right)^{1/2}. \tag{9}$$

It is seen from (8) and (9) that at $g\geqslant g_1$ or $n'\geqslant n_1$ the phonon wind comes into play. The average density of the electron-hole pairs that condense into EHD, at which the influence of the droplet repulsion becomes significant, is

$$n_1=\frac{Mn_0}{4\pi\rho^2\tau_0\tau_p}\approx 2\cdot 10^{15}\,\mathrm{cm}^{-3}. \tag{10}$$

We note that n_1 is determined only by an aggregate of constants of the EHL. At low crystal-excitation levels $g<g_1$ the EHL density varies linearly with the pump, $n_{st}\propto g$; at $g>g_1$ the density $n_{st}\propto g^{1/2}$. At very weak pumps $g\ll g_1$, however, when the supersaturation of the exciton gas becomes significant, Eq. (8) is not suitable, and the $n_{st}(g)$ dependence should be superlinear, as had been demonstrated theoretically and experimentally.[9,11]

Nonstationary state. The solution of (8) describing the kinetics of the growth of the EHL density when the excitation is turned on is

$$n(t)=n_{st}\left\{1-\left[1+\frac{1}{2}\left(\frac{\tau^*}{\tau_0}+1\right)\left(\exp\frac{t}{\tau^*}-1\right)\right]^{-1}\right\}. \tag{11}$$

When the excitation is turned off, the density of the condensed phase decreases with time as follows:

$$n(t)=n_{st}\left[\frac{1}{2}\left(\frac{\tau_0}{\tau^*}+1\right)\exp\frac{t}{\tau_0}-\frac{1}{2}\left(\frac{\tau_0}{\tau^*}-1\right)\right]^{-1}. \tag{12}$$

In (11) and (12)

$$\tau^*=\tau_0/(1+4g/g_1)^{1/2}.$$

When comparing the theoretical conclusions with the experimental results it must be borne in mind that the proposed model does not take into account the adhesion of the droplets to the impurity centers[12] and the possibility of their evaporation upon recombination on the surface.

3. EXPERIMENTAL TECHNIQUE

To verify experimentally the premises advanced above, we investigated the dependence of the EHL volume on the level of the quasistationary excitation of the crystal by sufficiently long light pulses $\tau_{pul}>\tau_0$. The use of long rectangular pulses has made it possible, in addition, to investigate the kinetics of the growth of the EHL and kinetics of its relaxation. To determine the total volume of the EHL we used in the present study the fact that the drops have a strong absorption band in the far IR region 200–75 μm.[13]

The experimental setup is similar to the one previously described.[14] A high-sensitivity Ge:B receiver (spectral sensitivity range 100–50 μm) was constantly exposed to background radiation at room temperature. The investigated germanium crystal was placed in a light pipe in the path of the background radiation. Illumination of the crystal produced EHD that absorbed a fraction $\alpha d<1$ of the background radiation, and a signal was produced by the receiver and was proportional to the volume of the condensed phase. We registered simultaneously also microwave absorption at $\lambda=2$ mm.

Notice must be taken of a number of shortcomings of the technique used to register the total volume of the EHL by using the absorption of electromagnetic waves near the plasma-resonance frequency. First, the absorption signal depends on the EHD size, especially if the measurements are made at the long-wave edge of the plasma-resonance line.[15] To attenuate this effect we registered a modulation signal from the short-wave tail of the absorption line (100–50 μm), i.e., in fact the intraband absorption by the holes bound in the EHD.[16] Second, weak parasitic absorption by the exciton may have been registered. The influence of this absorption should manifest itself at high temperature, when the exiton density increases noticeably. For this reason, the investigations were carried out at $T<2.5$ K. Finally, the appearance of nonlinear effects at $\alpha d>1$ forced us to limit the stationary-excitation level to a value ~1 W/cm^2.[15] We monitored αd by measuring the absorption in a thick sample ($d=2$ mm). The signal was then approximately twice as strong as from the investigated thin samples at maximum pumping. The samples were mounted on a carousel and they could be easily compared during the time of the experiment.

We investigated pure p-type germanium crystals with $N_A-N_D\approx 10^{12}$ cm^{-3} and dislocation density $\lesssim 10$ cm^{-2}. The samples were cut in the (111) plane in the form of 4×4 mm squares 105, 220, and 370 μm thick, polished mechanically, and then etched in CP-4A and in H_2O_2 + NaOH.

To ensure that the crystals with different thicknesses had identical surface finishes, we measured the rates of surface recombination prior to placing the samples in the cryostat. The surface recombination rate was determined from the usual formula $S=d/2\tau_s$, where τ_s is the carrier lifetime in a sample of thickness d; the lifetime was determined from the kinetics of the microwave absorption at $T=300$ K and at a pump ~10 W/cm^2. All samples had $S=100$ cm/sec.

The EHD were excited by uniformly illuminating the crystals with rectangular light pulses whose durations ranged from 10^{-5} to 5×10^{-4} sec. The rise and fall time of the pulses was 0.5 μsec. The repetition frequency was usually ~310 Hz. The light pulses were shaped by an acousto-optical modulator.[17] The continuous light source was an YAG:Nd laser or an argon laser. The light pulse power could be smoothly varied up to 0.4 W. In the Raman-Nath regime, the modulator made it possible to obtain two light beams of equal intensity.[18] This regime was used in control experiments to illuminate the germanium plate from both sides. In some experiments we used a GaAs laser with up to 5 W power and 10 μsec pulse duration. In all the experiments the intensity of the exciting light did not exceed the values at which film boiling of the superfluid helium, and correspondingly considerable superheat of the crystal, takes place.[19] The density n_{ex} of the electron-hole pairs produced by the light was estimated from the rate of generation, with account taken of the coefficient of reflection of the light from the crystal and from the dewar windows. The quantum yield was assumed to be unity.

The investigated crystal and the receivers were placed in a helium bath. Most experiments were performed at 1.9 ± 0.01 K, a temperature that limited the rate at which the helium vapor was pumped off. We used in the measurements a signal-accumulation system based on a multichannel pulse analyzer; this system made possible a 100-fold improvement in the signal/noise ratio. The measurement results were fed to a printout unit.

4. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

Figure 1 shows the dependence of the signal, which is proportional to αd, on the quasistationary excitation level for three identical samples of different thickness. On curves B and C one can separate three sections: $\alpha d \propto g^3$, $\alpha d \propto g$, and $\alpha d \propto g^{1/2}$. The first two were observed, for example, by Pokrovskiĭ and Svistunova.[11] The new feature, the sublinear dependence close to

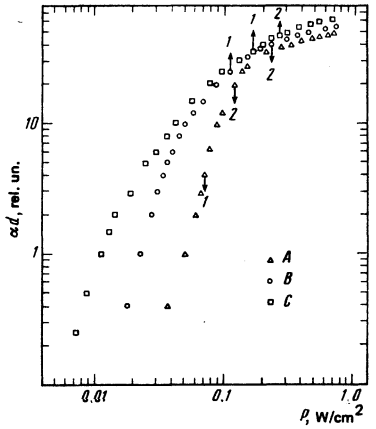

FIG. 1. EHD absorption signal vs. intensity of exciting light for samples of different thickness (A—105μm, B—220 μm, C—370 μm). T = 1.9 K. The arrows mark: 1) the start of the steepening of the leading front, 2) the appearance of fluctuations in the microwave absorption.

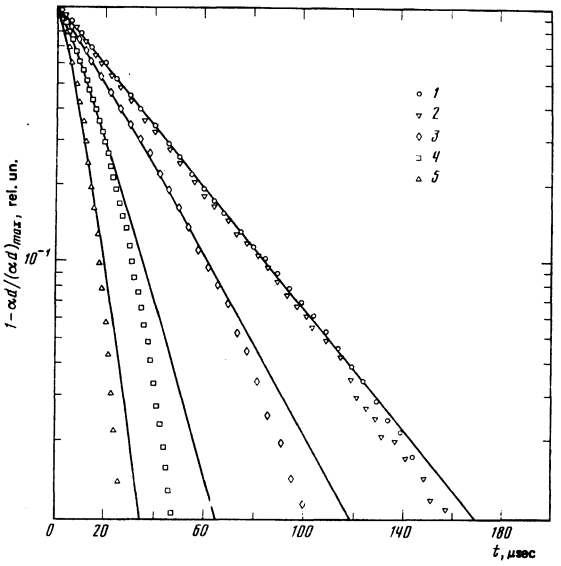

FIG. 2. Growth of EHD absorption at various exciting-light intensities. Pulse duration 250 μsec, T = 1.9 K. 1) 0.038 W cm^2, $n_{ex}<n_1 = 2\times10^{15}$ cm^{-3}, $\tau_0 = 37$ μsec; 2) 0.076 W/cm^2, $n_{ex} < 2\times10_{15}$ cm^{-3}, $\tau_0 = 37$ μsec; 3) 0.15 W/cm^2, $n_{ex} = 1\times1015$ cm^{-3}, $n = 6\times1014$ cm^{-}'; 4) 0.3 W/cm^2, $n_{ex} = 3\times1015$ cm^{-3}, $n = 3.2\times1015$ cm^{-3}; 5) 0.6 W/cm^2, $n_{ex}6\times1015$ cm^{-3}, $n = 1.2\times10^{16}$ cm^{-3}.

$g^{1/2}$, is typical of high excitation levels and is satisfactorily described by Eq. (9). At the same excitation levels, according to (11) and (12), one should expect a decrease in the EHL lifetime, which should manifest itself in faster EHL growth and fall-off kinetics.

The EHL growth kinetics for pulsed excitation of a sample 220 μm thick is illustrated in Fig. 2. At low exciting-light intensity (curves 1 and 2) the EHL are immobile and from the leading front it is possible to determine exactly the droplet lifetime $\tau_0 = 37 \pm 2$ μsec, including the influence of the exciton evaporation. Starting with a pump ~0.1 W/cm^2, however, the EHL lifetime decreases (curves 3–5). It is known that the optical hysteresis vanishes at these same excitation levels.[20] These phenomena are due to the start of the EHD motion and to their emergence to the crystal surface. The solid curves of Fig. 2 are the theoretical plots (11) corresponding to different densities n of the electron-hole pairs bound into EHD.

Comparison of the experimental curves 1–5 (Fig. 2) with the theoretical relation (11) shows a large discrepancy. Its cause is that the transition of the EHD system to the stationary state proceeds in an anomalous manner (see Fig. 3, curves 4 and 5). It is seen from the figure that the amplitude of the transient overshoot increases with increasing temperature (curve 6). This can be explained qualitatively as follows. Under surface excitation, the EHD first emerge to the illuminated surface of the sample, where they recombine intensively and generate additional phonon fluxes that can carry the EHD into the interior of the crystal, increasing in final analysis the flux of the EHD to the sample surface. It should be noted that the strong dependence of the transient overshoot (curve 6) seems to indicate an enhancement of the role of the phonon wind

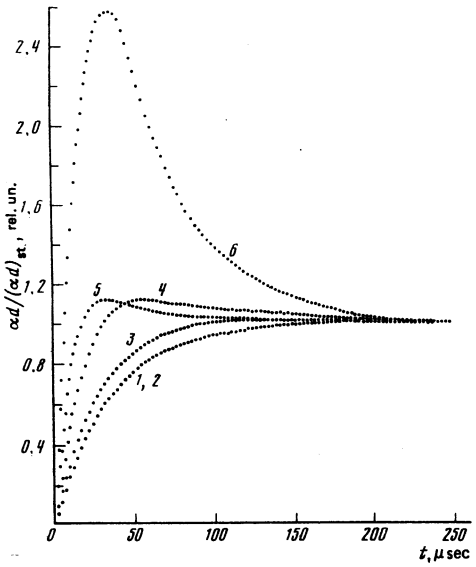

FIG. 3. Kinetics of transition of EHD system to the stationary state following surfact excitation. Light intensity, W/cm^2: 1—0.038, 2—0.076, 3—0.152, 4—0.30, 5—0.56, 6—0.152. Pulse duration 250 μsec, T = 1.9. Curve 6 corresponds to T = 2.3 K.

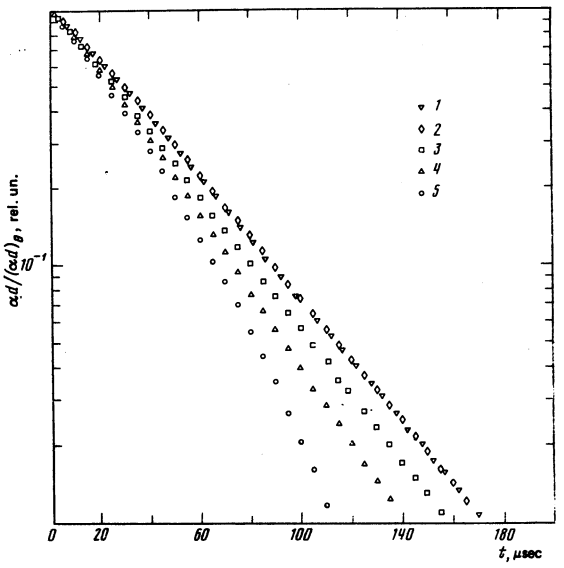

FIG. 5. Decrease of EHD absorption upon excitation of crystal by light pulses of varying durations (μ sec): 1—10, 2—25, 3—50, 4—100, 5—250. Light intensity 0.56 W/cm^2, T = 1.9 K.

with increasing temperature, compared with the forces that keep the EHD on the impurities.[12]

In addition to these singularities, when the pump values marked by the arrows numbered 2 on Fig. 1 are reached, an abrupt time-fluctuating growth of microwave absorption is observed. This phenomenon was observed by Ashkinadze and Sultanov,[21] but has not found an unambiguous explanation to this day.[22] We assume that this anomaly of the microwave absorption is due to evaporation of the droplets, or to their heating to critical temperature when in contact with the sample surface. In both cases, the rise in the carrier temperature and the decrease of their density lead to a sharp decrease of the carrier momentum relaxation time and accordingly to an absorption burst in the microwave region.[23]

We note that for samples 220 and 370 μm thick the steepening of the leading front comes into play at densities $n_{ex} \approx 10^{15}$ cm^{-3} (0.1 – 0.2 W/cm^3) (see Fig. 1), which is close to the estimate (10). For the 105-μm sample, the steepening of the leading front occurs at lower densities. The reasons for this are not clear. It is possible that additional mechanisms come into play at small thicknesses and carry the EHC to the surface, such as strain-induced attraction to the surface[24] or the influence of the exciton-density gradient,[25] which is particularly appreciable in thin samples.

We turn now to the kinetics of the EHL decay (Figs. 4–6). It is seen from Fig. 4 that when the EHL was

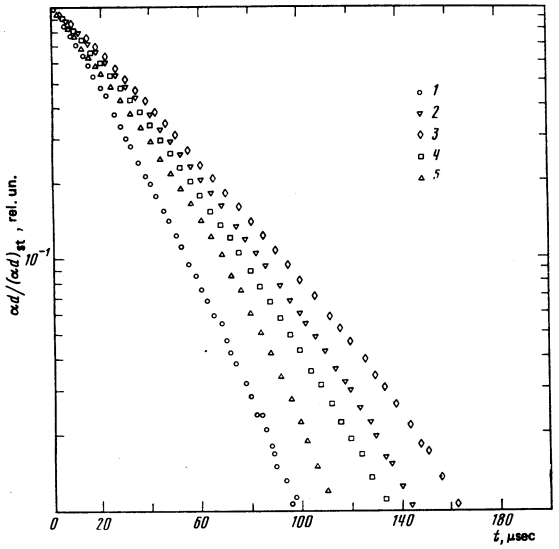

FIG. 4.

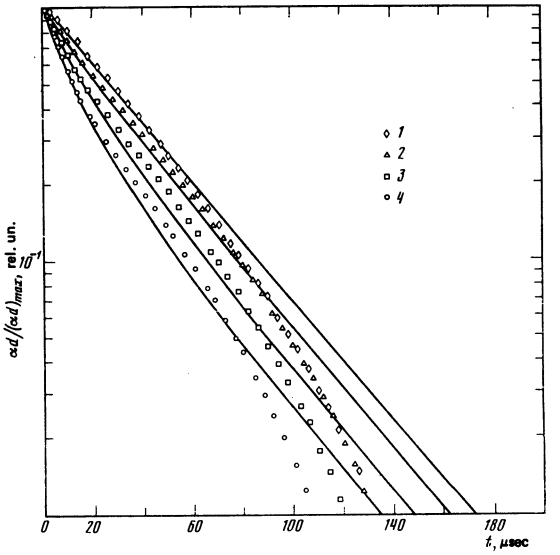

FIG. 6. Decrease of EHD absorption at various intensities of the exciting light. Pulse duration 10 μ sec, T = 1.9 K. 1) 0.6 W/cm^3, n_{ex} ($n_1 = 2\times10^{15}$ cm^{-3}; 2) 1.3 W/cm^2, $n_{ex} = 3\times10^{15}$ cm^{-3}; $n = 8\times10^{14}$ cm^{-3}; 3 - 2.0 W/cm^2, $n_{ex} = 4\times10^{15}$ cm^{-3}, $n = 3.2\times10^{15}$ cm^{-3}; 4) 3.7 W/cm^2, $n_{ex} = 8\times10^{15}$ cm^{-3}, $n = 9.6\times10^{15}$ cm^{-3}.

excited by 250-μsec pulses we were unable to obtain a significant acceleration of the decay kinetics, as might be predicted by (12), although a noticeable shortening of the rise time of the EHL is observed at the same pump values (Fig. 2, curves 3–5). One might assume that the shortening of the growth time is due to the influence of the phonon flux produced when the hot photoelectrons become thermalized. To check on this assumption, control experiments were made in which two each of the EHL growth kinetics were recorded under different crystal excitation conditions.

In the first experiment one kinetics was recorded with the crystal excited by light from a YAG : Nd laser ($P = 0.15$ W/cm^2, $h\nu = 1.16$ eV), and the other with argon-laser excitation ($P = 0.31$ W/cm^2, $h\nu = 2.43$ eV), with the total number of photons conserved.[26] The kinetics were identical.

In the second experiment, the kinetics was first recorded using an YAG : Nd laser ($P = 0.15$ W/cm^2) to illuminate first one surface of the crystal, and then both sides, again with the excitation level maintained constant. The results were likewise identical.

These experiments forced us to exclude the influence of the external phonon flux on the EHD when the crystal is excited with light of intensity up to 1 W/cm^2. We propose that the reason why the decay does not speed up when the crystal is excited by a long pulse is that the EHD adhere to impurities. The point is that the force that holds the EHD to the impurity ($F_0 \approx 10^{-9}$ dyne) does not depend on the EHD radius.[27] At the same time, the force exerted on the EHD by the remaining droplets is proportional to R^3 and is equal to $F = \rho VE$ or, with allowance for (5),

$$F = \frac{(4\pi)^2}{3} \rho^2 R^3 \frac{n}{n_0} x.$$

For example, at $n = 2\times10^{15}$ cm^{-3}, $R = 2$ μm, $x = 10^{-2}$ cm, $\rho = 102\, g^{1/2}$ cm$^{-3/2}$ sec^{-1} we have $F = 4\times10^{-10}$ dyne, i.e., $F \approx F_0$. However, $F \approx F_0$ already at $R = 5$ μm. It is therefore possible to compare the theory (13) with experiment of the EHD are large enough. This condition can be satisfied, since it has been observed that the drop radius depends on the duration of the exciting-light pulse.

It is usually assumed that at low temperatures ($T \lesssim 2$ K) the capture of the excitons by droplets can be neglected, and the total droplet evaporation time should be determined by the initial radius of the EHD.[17] On the basis of this, it is possible to determine roughly from Fig. 5 that the EHD have their maximum radius at 10–30 μsec after the light is turned on. With increasing excitation-pulse duration the total EHD evaporation time (Fig. 5, curves 3–5) and the EHL volume (Fig. 3, curve 5) decrease. The apparent reason is the emergence of the large EHD to the surface of the crystal. Similar peculiarities of the transient process, upon excitation of the droplets by long light pulses, were observed also by Shaklee[28] by using a light-scattering procedure.

Figure 6 shows the relaxation kinetics of EHD excited by a short light pulse. The experimental plots agree qualitatively with the theoretical (12). The discrepancy between the curves at $t = 20$–30 μsec indicates that the EHD are stopped at 20–30 μsec after the termination of the light pulse, inasmuch as the fluctuations of the microwave absorption stop at the same time. The deviation at ~80 μsec is due to evaporation of excitons from the EHD, a process not taken into account in the derivation of (12).

Returning to Fig. 4, it should be noted that the determination of the EHD radius from the decay kinetics when the droplets are excited by long light pulses is difficult for the following reason. The shape of the curves of Fig. 4 points to a significant role of the capture of excitons by the droplets. Under these condition, the determination of the radius of the EHD from the known formula for the evaporation time[10] is impossible. Numerical solution of the kinetic equations[10] is likewise not effective, since it is seen from Fig. 4 (curves 2 and 3) that the EHD lifetime determined from the trailing edge is substantially larger than 37 μsec (cf. curve 2 of Fig. 2). The reason for this is still unclear.

We note in conclusion that on the basis of a simple electrostatic model of the interaction between the droplets, an explanation was found not only for the kinetics of the EHD relaxation at high excitation levels,[29] but also for the dependence of the volume and of the lifetime of the EHL on the exitation level.

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