Recombination of nonequilibrium carriers in thin samples of strongly excited germanium

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The kinetics of recombination of carriers bound into electron-hole droplets (EHD) is investigated theoretically and experimentally under conditions when the EHD are dragged to the sample surface by phonon wind. It is shown that the form of the dependences of the liquid phase on the time elapsed after the excitation is turned off, and the character of the variation of these dependences when the pump level is increased are determined (other conditions being equal) by the mechanism that generates the nonequilibrium phonons responsible for the EHD dragging. The experimental results are discussed on the basis of familiar notions concerning the evolution of an EHD cloud [V. S. Bagaev *et al.*, Problems in Condensed Matter Sciences, 1983, Vol. 6, Chap. 3, p. 187].

INTRODUCTION

Under typical experimental conditions[1)] the spatial distribution and kinetics of EHD are determined by the dragging of the droplets by phonon wind, i.e., by a stream of nonequilibrium long-wave acoustic phonons with quasimomenta smaller than double the Fermi quasimomentum of the carriers in the liquid phase (see the review by Bagaev *et al.*[2]). If the phonon wind is intense enough, the droplets moving from the excitation region can reach the sample surface. Since the lifetimes on the crystal surface and volume are different, the EHD motion to the sample surface can alter substantially the recombination rate of the nonequilibrium carriers. The magnitude of this effect depends on the ratio of the surface and volume recombination rates and on the particle velocity, the latter being dependent on the phonon-wind that increases with increasing crystal excitation level. It is clear therefore that the influence of the sample surface on the recombination should be most pronounced at high pump intensities.

The shortening of the EHD lifetime with increase of pump level was observed in Refs. 3–5 and in a number of other studies. In Refs. 6 and 7 was observed an abrupt decrease of the liquid-phase volume when the droplets were carried to the surface by the phonon wind generated when the nonequilibrium carriers surrounding the droplet were heated by electromagnetic radiation. A similar effect was observed also when the nonequilibrium-carrier current dragging the droplets was produced with a thermal generator.[8,9] At high excitation intensities the rate at which the energy initially stored in the EHD was released to the sample surface may turn out to be so large that the surface recombination of the carriers bound into droplets is accompanied by boiling of liquid helium on the crystal surface.

To analyze the recombination kinetics of the liquid-phase particles under conditions when the EHD are dragged to the sample surface by phonon wind, information is needed on the dynamics of the droplet cloud. It is known[2] that the evolution, in space and time, of an EHD produced by a short light pulse proceeds in three stages that follow one another, in which the phonon wind is generated by different mechanisms.[2)] In the first stage,[11] the cloud of equilibrium carriers expands under the influence of the stream of primary long-wave acoustic phonons released during the concluding stage of the thermalization of the photoexcited carriers. An EHD layer begins to form in the course of the expansion. The second stage, [12,13] consists of motion of an EHD layer due to dragging of the droplets by phonons emitted upon relaxation of the "hot spot," [14] which is a relatively slowly expanding cloud of nonequilibrium short-wave phonons produced by thermalization of the carriers[3)].[15,16] In the third and final stage, the EHD cloud is expanded by a mutual droplet repulsion produced because each EHD absorbs phonons emitted by other droplets,[12,17,18]

The influence of the mutual repulsion of the EHD on the recombination kinetics in thin samples was analyzed in Ref. 5 under the assumption that the surface recombination is infinitely fast. Here we consider the EHD annihilation kinetics during the second and third stages of the droplet-cloud evolution and present the results of a numerical calculation of the dependences of the liquid-phase volume in germanium on the time elapsed after the excitation pulse, at various pump levels and ratios of the surface and bulk recombination rates. The recombination kinetics during the first stage of EHD cloud expansion was not calculated. The role of this stage, however, is qualitatively clear even without calculations. Carrier dragging by primary phonons becomes noticeable only under sufficiently strong excitation, when the nonequilibrium carriers (or the EHD) move with near-sonic velocities.[11] Because of the last circumstance, almost all the carriers produced by excitation of the sample are ejected to its surface within a very short time, provided the sample dimensions are not too large.[11,19] The carrier recombination rate will therefore be determined by their lifetime on the sample surface. It will be shown below that at high pump intensities a similar influence is exerted on the recombination kinetics by droplet dragging by phonons emitted from the hot spot. At the same time, the recombination ki-

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netics in the third stage of the EHD cloud expansion has an entirely different character. This difference in the recombination kinetics is of fundamental importance. The point is that after the nonequilibrium currents have become thermalized, the electron subsystem has practically no effect on the intensity and damping of the phonon wind produced as a result of this process, i.e., the electron and phonon subsystems are, so to speak, detached from each other. Whereas the mutual repulsion of the EHD is due to phonons emitted by the droplets themselves, the phonon-wind intensity is determined by the amount of liquid phase in the volume of the crystal, which decreases in the course of time on account of transport of the droplets to the sample surface and recombination of the liquid particles in its volume.

Recombination kinetics in thin germanium samples with various surface-recombination rates was investigated by studying the damping of the EHD luminescence. The results can be satisfactorily explained qualitatively on the basis of the considered model of recombination under conditions of EHD dragging to the sample surface by phonon wind.

THEORY

We consider recombination kinetics in a plane-parallel plate of thickness a, bounded by two infinite planes. Assume that absorption of G photons per unit area produces at the initial instant of time ($t=0$) EHD that are uniformly distributed through the volume. We confine ourselves to the low-temperature regime ($T \lesssim 2$ K in Ge), when droplet evaporation can be neglected.[2,21] Electron-hole recombination will leave in the plate at the instant t a total of $N_\Sigma(t)$ carriers per unit area, with $N_\Sigma^V(t)$ bound into EHD and located in the bulk of the plate, and the remainder $(N_\Sigma - N_\Sigma^V)$ in thin (thickness $\ll a$) subsurface layers near its faces. The equation that describes the kinetics of the recombination of the nonequilibrium carriers in the plate can then be written in the form

$$\frac{dN_\Sigma(t)}{dt} = -\frac{N_\Sigma{}^V(t)}{\tau_0} - \frac{N_\Sigma(t) - N_\Sigma{}^V(t)}{\tau_s}, \qquad (1)$$

where τ_0 is the lifetime of the particles bound in the droplets in the bulk of the crystal and τ_s the lifetime in the subsurface layer. We assume the following: 1) The EHD were dragged into the subsurface layer by phonons created in the bulk of the plate. 2) The flux of particles from the bulk of the sample through the subsurface-layer boundary is independent of the number of particles in this layer (i.e., the average density of the nonequilibrium carriers in the layer is much less than the density of the liquid phase). 3) There is no delay between the emission and absorption of the phonons,[11] so that in view of the symmetry of the plate the phonon fluxes reflected from opposite faces, and those released on recombination of carriers in the opposite subsurface layer, are cancelled out at each point of the plate. This last assumption means in essence that we neglect phonon reflection and the phonon wind produced by surface recombination of the carriers. By virtue of assumptions 1–3, to calculate $N_\Sigma^V(t)$ we must solve the problem of expansion of a planar EHD cloud of thickness a at ($t=0$) in an infinite crystal, by specifying the specific mechanism whereby the phonon wind is generated.

Since the stages, listed in the introduction, of the space and time evolution of a droplet cloud are separated in time,[12–18] we shall consider the dragging of droplets by phonons emitted by the hot spot separately from the expansion of the cloud by the mutual-repulsion forces, and regard the phonon propagation and the electron-phonon interaction as isotropic. Note that if no account is taken of the phonon reflection from the plate surfaces, the effect of the first cloud-expansion stage on the recombination kinetics can also be investigated by using the scheme described below. The equations that describe the cloud dynamics during this stage are given in Ref. 11. Since, however, the first stage is of short duration, the difference in the carrier-recombination kinetics in the first and second stages of the cloud evolution should manifest themselves most distinctly for short times, when allowance for the finite phonon propagation time becomes essential.[11] The last circumstance prevents us from ignoring phonon reflections from the sample walls, for in this case the phonon fluxes reflected from the opposite faces will not cancel each other. Restricting ourselves to these remarks concerning the first stage of EHD cloud dynamics, we proceed now to calculate $N_\Sigma^V(t)$ during the next two stages.

Let the hot spot produced at the instant $t=0$ as a result of thermalization of photoexcited carriers be homogeneous and localized in the region of thickness a where these carriers were created. The hot spot relaxes and emits ballistically propagating long-wave phonons—the phonon wind. In view of the symmetry of the problem, the droplet motion is one-dimensional and a particle moving inside the hot spot and located a distance x from its center has at the time t a velocity (Refs. 2, 13, 22)[5)]

$$v(x,t) = \frac{\Gamma\bar{G}}{\bar{n}_{max}a}\frac{x}{\tau_{ph}}\exp\left(-\frac{t}{\tau_{ph}}\right), \qquad (2a)$$

where τ_{ph} is the hot-spot relaxation time,

$$\bar{n}_{max} = \frac{Mn_0{}^2}{4\pi\rho_{ph}{}^2\tau_p\tau_0} \qquad (3)$$

is a certain characteristic density, averaged over the sample volume, of the carriers bound into droplets[2,5] (if a homogeneous EHD cloud with average density $n = n\max$ is produced at $t=0$ in an infinite crystal, expansion of this cloud by the forces of mutual repulsion doubles its volume at $t\to\infty$),

$$\rho_{ph} = \left[\frac{\beta E_g}{4\pi}\frac{n_0}{s\tau_0}\alpha(k_F)\right]^{1/2} \qquad (4)$$

is the EHD repulsive-interaction constant,[2,17] M is the effective mass of the electron-hole pair, n_0 is the density of the liquid phase, τ_p is the EHD momentum relaxation time, s is the speed of sound, $\alpha(k_F)$ is the coefficient of absorption by the EHD of phonons whose wave vector k_F is equal to the Fermi vector of the electrons bound into droplets, E_g is the semiconductor band gap, β is the energy fraction released in nonradiative collision recombination of the liquid particles, and finally the parameter $\Gamma = \Delta E/\beta E_g$ is the ratio of the energies going into phonon wind in the hot spot per excited electron-hole pair (ΔE) and into EHD upon recombination of a pair of liquid particles (βE_g). Note that $\bar{n}_{\max}/\Gamma$ is the limiting (maximum attainable) average density of particles

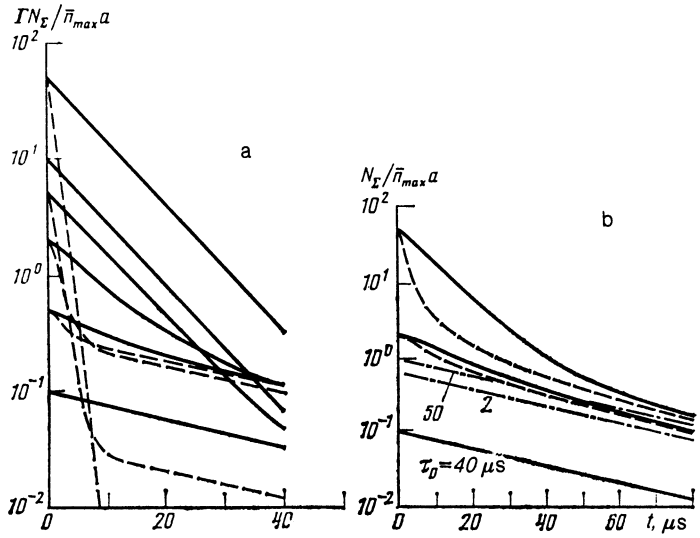

FIG. 1. Effect of phonon wind and of the sample surface on the recombination kinetics of carriers bound into droplets in Ge for various pump levels and for two values of the ratio τ_0/τ_s, viz., 5 (solid lines) and 40 (dashed). a) Phonon wind generated on relaxation of the hot spot. N_Σ (in units of $\bar{n}_{max}a/\Gamma$) was calculated from Eq. (7a). The curves are for different values of the excitation density $G = N_\Sigma$ at $t = 0$. b) Phonon wind emitted by the EHD themselves. N_Σ (in units of $\bar{n}_{max}a$) calculated from Eq. (7b). Dash-dot curves—calculated from (8b) at $G/\bar{n}_{max}a = 50$ and 2.

that can be bond into drops via stationary excitation of the sample, if the surface of the latter does not hinder the droplet motion[2,23] (for germanium we have $\bar{n}_{max} \approx 2\cdot 10^{14}$ cm^{-3} (Ref. 2), $\Gamma \approx 0.7$ for pulsed surface excitation,[2] and $\Gamma \approx 0.2$ for stationary excitation[22]).

In expansion of a planar EHD cloud by mutual repulsion forces, the equation for the droplet velocity can be written in the form

$$v(x, t) = N_\Sigma(x, t)/2\bar{n}_{max}\tau_0, \tag{2b}$$

where $N_\Sigma(x,t)$ is the total number of droplets bound into pairs per unit area of a layer of thickness $2x$ at the instant of time[6] t.[2,17] Integrating Eqs. (2a) and (2b), the latter with allowance for the relation

$$N_\Sigma(x(t), t) = N_\Sigma(x(0), 0)\exp(-t/\tau_0),$$

we obtain the expressions

$$\ln\frac{x(t)}{x(0)} = \frac{\Gamma\bar{G}}{\bar{n}_{max}a}(1-e^{-t/\tau_{ph}}), \tag{5a}$$

$$x(t) = x(0) + \frac{N_\Sigma(x(0),0)}{2\bar{n}_{max}}(1-e^{-t/\tau_0}), \tag{5b}$$

which describe the time dependence of the moving-droplet coordinate. Putting $x(t) = a/2$ in these expressions we can find the thickness $2x(0)$ of the region on the boundary in which were located at $t = 0$ the drops that left at a given instant of time the boundaries of the initially excited crystal region. The number of carriers bound into droplets in this region (in a region of thickness a) is given by (if the excitation region is uniformly filled with droplets at $t = 0$)

$$N_\Sigma^V(t) = \frac{2x(0)}{a}\bar{G}e^{-t/\tau_0}. \tag{6}$$

We used here the initial conditions $N_\Sigma(0) = N_\Sigma^V(0) = \bar{G}$ and took into account the recombination in EHD as they move through the excitation region.

Substituting the formulas obtained from (5) for $x(0)$ [at $x(t) = a/2$] in expression (6), and then $N_\Sigma^V(t)$ in Eq. (1) and solving the latter, we get

$$N_\Sigma(t) = \bar{G}e^{-t/\tau_s}\left\{1 + \frac{\tau_{ph}}{\tau_0}\left(\frac{\tau_0}{\tau_s} - 1\right)\exp\left(-\frac{\Gamma\bar{G}}{\bar{n}_{max}a}\right) \times \int_{e^{-t/\tau_{ph}}}^{1}\exp\left(\frac{\Gamma\bar{G}}{\bar{n}_{max}a}z\right)\frac{dz}{z^\theta}\right\}, \tag{7a}$$

$$N_\Sigma(t) = \bar{G}e^{-t/\tau_s}\left\{1 + \left(\frac{\tau_0}{\tau_s} - 1\right)\frac{\bar{n}_{max}a}{\bar{G}} \times \int_{e^{-t/\tau_0}}^{1}\frac{z^{-\tau_0/\tau_s}\,dz}{(1 + \bar{n}_{max}a/\bar{G}) - z}\right\}, \tag{7b}$$

where $\theta = 1 + \tau_{ph}(\tau_0 - \tau_s)/\tau_0\tau_s$. Expressions (7a) and (7b) describe the recombination kinetics of the carriers bound into droplets under conditions when the EHD are dragged to the sample surface by the phonon winds emitted respectively by the hot spot and by the EHD themselves. The $N_\Sigma(t)$ dependences calculated from these equations for germanium ($\tau_0 = 40\,\mu$s (Refs. 1 and 21) and $\tau_{ph} = 2\,\mu$s (Refs. 13 and 24) at different values of the pump level $\bar{G}$ and of the ratio τ_0/τ_s are shown in Fig. 1, while the dependences of N_Σ on the pump level at various instants of time t after the excitation pulse are shown in Fig. 2.

We discuss first the effect exerted on the recombination kinetics of the phonon wind generated on relaxation of the hot spot. As seen from Fig. 1a, at high excitation intensities practically all the EHD are ejected by the phonon wind into a subsurface region of the sample, and the recombination kinetics is described by the "surface lifetime" τ_s. The phonon wind attenuates quite rapidly (within a time $\sim\tau_{ph} \ll \tau_0$), and after the carrier recombination in subsurface layer is terminated, the EHD remaining in the bulk of the sample die out after a lifetime τ_0. This process is described by the

$$N_\Sigma(t) = \bar{G}\exp\left[-\frac{\Gamma\bar{G}}{\bar{n}_{max}a}\right]e^{-t/\tau_0}, \tag{8a}$$

which is valid at large t [$t \gg \tau_{ph}, t > (\Gamma G/\bar{n}_{max}a)\tau_s\tau_0/$

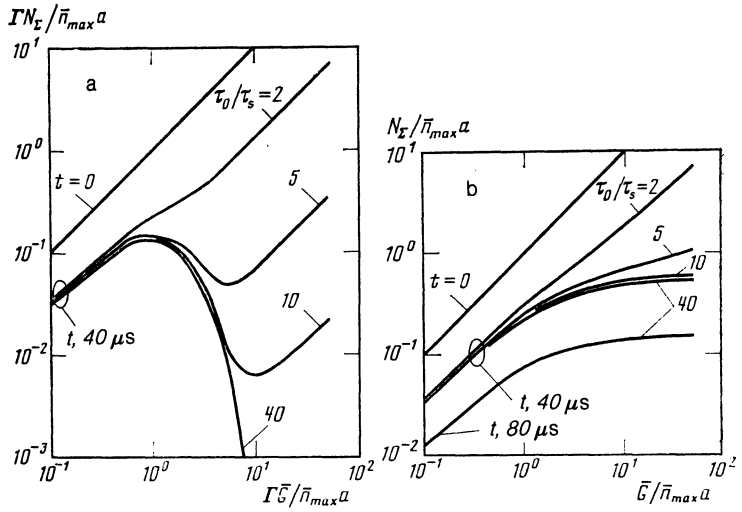

FIG. 2. Total number of nonequilibrium carriers in the sample vs the pump level at various instants of time t following the excitation pulse, for several values of the ratio τ_0/τ_s. a) Phonon wind emitted by hot spot. Calculation from Eq. (7a). b) Phonon wind emitted by the EHD themselves. Calculation from Eq. (7b).

$(\tau_0 - \tau_s)]$. The factor preceding the exponential that contains t varies nonmonotonically when the pump level is increased; it reaches a maximum at $\bar{G} = \bar{n}_{\max} a/\Gamma$ and decreases with further increase of $\bar{G}$. This means that at $t \gg \tau_{ph}$ the average density of the particles bound into droplets in the bulk of the plate is $\bar{n} = N_\Sigma^V/a$, or does not exceed $\bar{n}_{\max}/2.72\,\Gamma$ in the initially excited region of the infinite crystal at any pump intensity. At not too large t, so long as there are more carriers in the subsurface regions of the plate than in its bulk, the dependence of N_Σ on the pump level turns out to be more complicated (Fig. 2a).

A different picture should be observed when the EHD cloud is expanded by the mutual-repulsion forces. The plots of $N_\Sigma(t)$ corresponding to different pump intensities but to equal surface-recombination rates do not intersect (Fig. 1b), and for any instant of time the dependences of N_Σ on the pump level are monotonic (Fig. 2b). At large t

$$t \gg \tau_0, \quad t > \frac{\tau_0\tau_s}{\tau_0 - \tau_s} \ln\left(1 + \frac{\bar{G}}{\bar{n}_{max} a}\right),$$

when the greater part of the nonequilibrium carriers is in the bulk of the plate, we have

$$N_\Sigma(t) = \frac{\bar{G}}{1 + \bar{G}/\bar{n}_{max} a} e^{-t/\tau_0}. \qquad (8b)$$

The pre-exponential factor in this equation increases monotonically with increasing $\bar{G}$, and approaches $\bar{n}_{\max} a$ at large $\bar{G}$, i.e., the average carrier density in the bulk of the sample at high pump levels is $\bar{n} = \bar{n}_{\max} \exp(-t/\tau_0)$ independently of the excitation intensity. The causes of this behavior of N_Σ are quite clear. According to (2b), the velocity of the EHD that leave the crystal at a given instant of time is proportional to $N_\Sigma^V(t)$. Therefore, when N_Σ^V decreases in the course of time to such an extent that the particle flux on the sample surface becomes smaller than the particle recombination rate in the bulk of the plate, the kinetics at $N_\Sigma(t) \approx N_\Sigma^V(t)$ is determined mainly by the bulk recombination. It is easy to show that this recombination predominates at $N_\Sigma(t) < \bar{n}_{\max} a$.

Thus, the character of the dependences of N_Σ on the time and pump intensity is a reflection of the singularities of the dynamics of the droplet cloud and is determined by the mechanism that generates the nonequilibrium phonons responsible for the dragging of the EHD towards the sample surface.

EXPERIMENTAL RESULTS AND DISCUSSION

The experiments were performed on germanium samples measuring $(0.1 - 0.5) \times 5 \times 5$ mm with residual-impurity density less than 10^{12} cm^{-3}. After mechanical polishing the samples were etched in hydrogen peroxide for various time intervals, and were then freely suspended in the working volume of a helium cryostat, to prevent mechanical stresses due to cooling. All experiments were performed at a temperature $T = 1.7$ K.

The surface excitation of the germanium was by a copper-vapor laser (wavelength $\approx 0.51\,\mu$m, pulse duration ~ 10 ns, repetition frequency ~ 5 kHz, maximum pulse energy $J_{\max} \approx 120$ erg). The laser beam was focused on the sample surface into a spot of ≈ 4 mm diam. The recombination radiation emerging through the same surface was gathered on the entrance slit of a spectrometer tuned to the maximum of the LA luminescence line of the EHD. The dependences of the luminescence intensity on the time elapsed from the sample excitation were measured with a resolution $\sim 0.5\,\mu$s.

As expected in view of the foregoing, the luminescence kinetics of the EHD was strongly dependent on the excitation level, on the sample thickness, and on the surface-recombination rate. Figures 3a and 3b show the results for two samples of thickness $a = 0.1$ mm, etched for 1.5 and 1.0 min, respectively. At the lowest pump level the luminescence intensity of both samples decreased exponentially with time, with a time constant $\tau_0 \approx 36\,\mu$s corresponding to the EHD lifetime in the bulk. With increasing excitation level, the time constant of the exponential falloff decreases, and at the highest short-time pump intensities the luminescence is more rapidly damped. Whereas for the sample with the lower surface-recombination rate the dependence of the EHD recombination-radiation intensity on the pump level is monotonic for all t (Fig. 4a), for the other sample this dependence is N-shaped at sufficiently long t (Fig. 4b), i.e., at

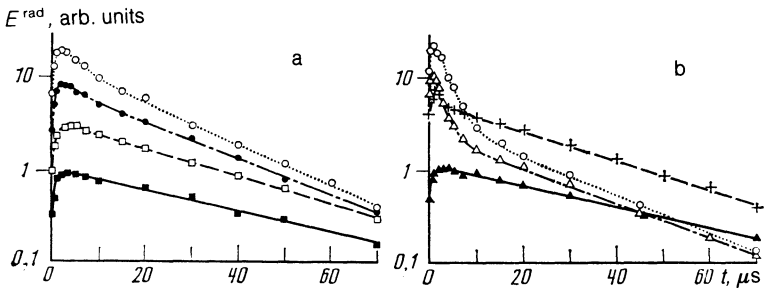

FIG. 3. EHD luminescence kinetics in Ge samples of thickness $a = 0.1$ mm at various pump levels $J/J_{max} = 1.0$ (○), 0.40 (●), 0.12 (□), 0.03 (■), 0.59 (△), 0.04 (▲), 0.41 (+) ($J_{max} = 120$ erg). The samples were etched for 1.5 min (a) and 1 min (b).

high excitation levels ($J \gtrsim 0.5\, J_{max}$) the luminescence intensity decreases within a time $t \lesssim 10\ \mu s$ so strongly that it becomes even smaller than for weaker pumping. At $t = 1$ to 2 μs the dependence of the luminescence intensity on the excitation level was linear for all investigated samples.

The character of the plots shown in Fig. 3a for samples of thickness $a = 0.1$ mm at $t \gtrsim 10\ \mu s$ remained unchanged when the etching time was increased to 7 min. However, there was no rapid kinetics at small t in this case even at the maximum pump level. In sufficiently thick ($a \approx 0.5$ mm) and well etched samples the lifetime ($\tau_0 \approx 36\ \mu s$) was independent of the excitation level, and the luminescence intensity was correspondingly proportional to the pump intensity at any instant of time.

One can hardly expect to describe all the experimental data by using the excessively idealized model considered above.[7)] It can nevertheless be used for a qualitative analysis of the observation results. Thus, the presence of an N-shaped dependence of the EHD recombination-radiation intensity on the excitation level (Fig. 4b) shows that the initial abrupt falloff of the radiation intensity at large pumps (Fig. 3b) cannot be due to expansion of the EHD cloud by the mutual repulsion forces (see Fig. 2). This conclusion agrees with the data of Ref. 18, in which it was shown, for experimental conditions close to ours, that the phonon wind emitted by the EHD themselves makes the main contribution to the drag force at $t \lesssim 2\ \mu s$. Within a time $t \gtrsim 2\ \mu s$ the drops are displaced ≈ 0.5 mm from the excited surface of the sample. Thus, as should be expected, the main singularities of the recombination kinetics should be due to one of the preceding stages of the EHD cloud dynamics. In that case, as noted above, if the pump intensity is high enough (the maximum pump intensity in our experiment corresponded to $\bar{G} \approx 50 \bar{n}_{max} a$ at $a = 0.1$ mm) almost all the nonequilibrium carriers are ejected to the sample surface, and the carrier-recombination kinetics at short t is described by the "surface lifetime" τ_s. An estimate of τ_s from the data shown in Fig. 3 yields for the maximum pump level $\tau_s \approx \mu s$ and $\approx 11\ \mu s$ at etching times 1 and 1.5 min, respectively. Although it is clear beforehand that the displacement of the EHD by a distance $a = 0.1$ mm is due to dragging of the droplets by primary phonons released during the concluding stage of carrier thermalization (this conclusion follows from the results of Refs. 11, 13, and 18), for lack of anything better we shall attempt to estimate the parameter Γ with the aid of Eq. (8a), which is valid for droplet dragging by phonons emitted by the hot spot. We shall assume roughly that at $t \approx 10\ \mu s$ and $J = J_{max}$ (Fig. 3b) the total number of carriers in the sample is equal to the numerical value of the factor preceding the time-containing exponential. Such an estimate yields $\Gamma \approx 0.05$. This value of Γ is noticeably lower than the $\Gamma = 0.7$ corresponding to conversion of the carrier energy into phonon wind in the hot spot following pulsed surface excitation.[2] At the same time, the obtained value of Γ does not differ greatly from the $\Gamma \approx 0.03$ obtainable from the data of Ref. 11, and corresponds to conversion of the carrier energy into a primary phonon wind (the same order of Γ is obtained for the primary phonon wind also by an estimate using the data of Ref. 19).

The phonon wind resulting from thermalization of the nonequilibrium carriers attenuates rapidly; were there no repulsive interaction between the EHD, an exponential decrease of the luminescence intensity would be observed at long t, with a rate corresponding to the bulk lifetime of the EHD. The shortening of the lifetime with increasing excitation level at $t > 10\ \mu s$ (Fig. 3) attests therefore to dragging of the droplets to the sample surface by phonons emitted by the EHD themselves. Note that since the expansion of the cloud by the mutual repulsion forces follows the two preceding cloud-dynamics stages, the spatial distribution of the EHD may turn out to be highly nonuniform already during the start of this stage, even if the sample was uniformly filled with droplets at the initial instant of time.

We call attention to one more conclusion that follows from the results shown in Fig. 3. At low pump levels, when

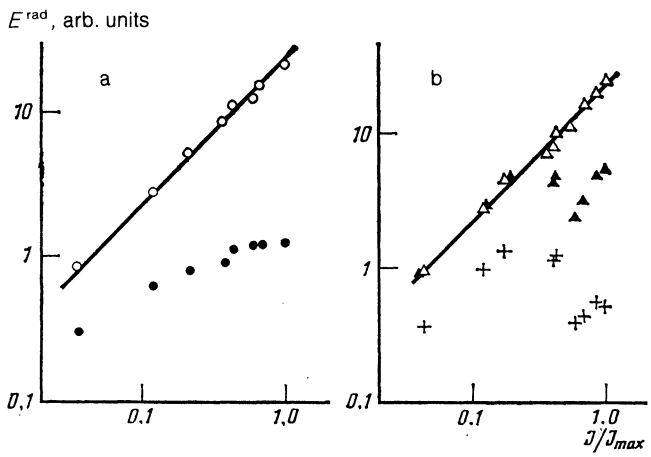

FIG. 4. EHD luminescence intensity vs pump level at various instants of time after the excitation pulse. $J_{max} = 120$ erg, $a = 0.1$ mm, etching times 1.5 min (a) and 1.0 min (b); $t(\mu s) = 1$ (△), 2 (○), 7 (▲), 45 (+) and 50 (●).

the phonon wind intensity is low, the EHD produced in the sample by surface excitation are concentrated near the excited surface in a layer not more than 100 μm thick.

Thus, at least two of the three EHD-cloud dynamic stages described in the Introduction were revealed in the experiments described above. One is connected with the phonons released by thermalization of the carriers, and the other with the mutual repulsion of the EHD. The strongest influence on the nonequilibrium-carrier recombination was exerted by primary long-wave phonons emitted by the thermalized carrier. In the experiments described in Refs. 4 and 5, however, the main contribution to the EHD dragging towards the surface was made by phonons emitted by the droplets themselves. Thus, the dependences of the EHD luminescence intensity on the pump level, observed in Ref. 4, were similar to those shown in Fig. 2b, while the luminescence-intensity time dependences corresponding to different pump levels were, in accordance with Eq. (8b), the same at $\bar{G} \gg \bar{n}_{\max} a$. This raises the question of identifying the conditions under which a particular stage of the EHD-cloud dynamics manifests itself. This question is important for the understanding of the totality of the experimental facts that have a bearing on droplet dragging by phonon wind and on the spatial distribution of the EHD. It is simplest to formulate the conditions under which observation of the first and third stage of cloud kinetics is possible. Dragging of EHD by primary thermalized phonons becomes noticeable at $\Gamma \bar{G} / 4a\bar{n}_{\max} \gtrsim 0.2$. When this condition is satisfied, more than half of the carriers produced in the sample leave the initially excited region of thickness a.[11,19] The expansion of the EHD cloud by the mutual-repulsion forces should be observed at average densities $\bar{n} \gtrsim \bar{n}_{\max}$ of the carriers bound into droplets.

The greatest difficulties arise when an attempt is made to formulate a similar criterion for the second stage of the EHD-cloud dynamics, even though this stage is more frequently observed in experiment than the others.[2] The phonon relaxation processes in a hot spot have by now become much better understood.[16] Numerical estimates (to be sure, quite crude) seem to show, however, that at distances on the order of the size of the EHD cloud the short-wave phonons have no time to decay into long-wave ones that drag the EHD.[22] An impression is gained that effective conversion of the hot-spot phonons into phonon wind requires reflection of the low-frequency phonons from the sample surface. Let us list the experimental facts that support this viewpoint. The most pronounced droplet dragging by phonons emitted from the hot spot is observed for samples whose excited surface is in a vacuum (is not in contact with liquid helium),[11,13] in which case the front of the EHD cloud moves several mm away from the excited surface. If, however, the sample is placed in liquid helium (with the remaining experimental conditions unchanged), this stage of EHD motion is substantially suppressed—the front of the cloud is moved only several tenths of a millimeter.[18] Analysis of the experimental conditions under which this stage of the EHD-cloud kinetics is observed for samples placed in liquid helium[25–29] shows that dragging of the EHD over considerable distances is apparently made possible by formation of a film of gaseous helium on the excited surface. This film prevents the nonequilibrium phonons from moving from the sample into the liquid helium. At pump intensities typical of such experiments, the vapor film is produced within a rather short time.[10] It is possible that the increase of the hot-spot lifetime with increasing excitation level, observed in Ref. 28, is also due to the boiling of the helium. A similar conclusion can be drawn also from the results obtained under stationary excitation. Formation of a vapor bubble in the laser focus[30] occurred in all experimental observations of the anisotropic spatial EHD-cloud structure that results mainly from droplet dragging by phonons emitted on relaxation of the hot spot.

CONCLUSION

We reported above the results of an investigation of recombination kinetics of carriers bound in EHD when the droplets are dragged towards the sample surface by phonon wind. An analysis, which is applicable under certain conditions also to excitons and free carriers, shows that the recombination kinetics in thin samples depends strongly on the phonon-wind generation mechanism and differs considerably from the kinetics in diffuse propagation of nonequilibrium carriers.

It must be emphasized that in a thin sample the electron-hole liquid is, so to speak, in two states with different lifetimes of its component particles. These two "phases" are separated in space—one is in the interior of the crystal and the other on its surface. The ratio of the volumes of these "phases" is determined by their lifetimes and by the intensity of the phonon wind. The question of the properties and recombination mechanism of the EHD in the subsurface layer of the sample calls, however, for a special study.

One more remark is essential for the interpretation of a number of experiments. The phonon wind can influence recombination processes in an exciton-droplet cloud and in thick samples, when the dragging to the surface plays no role. Thus, the recombination kinetics can be distorted by the EHD distribution in size,[31,32] a distribution formed by the phonon wind,[33] by expansion of the EHD cloud,[4] by dragging of the droplets by phonons outside the exciton cloud, etc.

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[1)]Exceptions are hysteresis-type experiments carried out at low pump intensities, only slightly higher than the liquid-phase formation threshold. These experiments are described in detail in Ref. 1.

[2)]Not necessarily all three stages of the cloud kinetics must be observed in each particular experiment. The conditions under which a particular stage can be observed are of great importance and will be dealt with in the discussion of the experimental results.

[3)]Since we are not interested here in the details of the processes in the "hot spot," we shall use this term in a somewhat broader sense than in Ref. 16.

[4)]Since the nonequilibrium carriers in Ge, Si, and a number of other semiconductors[20] are in the liquid phase at low temperatures and at sufficiently high pump intensities, we shall speak, for the sake of argument, of EHD motion. The reasoning that follows can in fact be used also in the treatment of recombination in a system of free carriers (or excitons), provided that the spatial distribution of the carrier is determined by

phonon-wind forces and not by diffusion.

[5)]The numbers of the equations pertaining to dragging of droplets by phonons emitted by the hot spot are labeled by the letter "*a*"; those describing the expansion of the cloud by the mutual repulsion forces are labeled "b."

[6)]If a cloud of free carriers or excitons is considered, the analog of the phonon wind emitted by the EHD can be, to some degree, the flux of phonons produced by nonradiative recombination of the carriers (excitons). The analogy is complete if part of the energy released in this process is converted into long-wave phonons within a time shorter than a/s and the carrier lifetime.

[7)]It appears that the assumptions made in the calculation do not conform to the experimental situation in at least two respects: first, the initial distribution of the nonequilibrium carrier over the sample volume can hardly be regarded as uniform. Second, at high pump level the average carrier density in the subsurface layer may turn out to be of the same order as the density of the liquid.

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