Electron-hole drops on a germanium surface and a kinetic phase transition

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The dependence of the lifetime of an electron-hole liquid on its average concentration on a crystal surface has been investigated. It is shown that in the transition from individual drops to a continuous liquid layer the lifetime increases suddenly by more than an order of magnitude; this is attributed to the appearance of a thin, gaslike film between the drops and the surface. The observed effect can be called a kinetic phase transition.

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We have investigated the recombination radiation of electron-hole drops that have been pressed against a crystal surface. It is known that the lifetime of drops produced within a massive sample is $\tau_0 \approx 40$ μsec.[1] If the drops are forced to emerge at the surface (due to the action of a phonon wind[2] on nonuniform deformation,[3] or simply as a result of excitation of a sufficiently thin sample[4]), then their lifetime will decrease; this is normally attributed to a high rate s of surface recombination. If $s \gtrsim 10^2$ cm/sec, then the vanishing time of a drop of radius $R = 1$ μm at the surface will be $\tau_s \approx R/s \leqslant 10^{-6}$ sec. It was found, however, that the recombination time can vary by more than an order of magnitude in different experiments for similar s values. For example, in the experiments described in Ref. 2 the drop radiation was quenched in a time $\leqslant 1$ μsec. In other experiments[4] involving high-power pulsed pumping of thin samples ($\leqslant 10$ μm thick), when the time for drops to move to the surface was very short, the lifetime of the liquid measured from the luminescence decay was equal to (10–15) μsec, i.e., an order of magnitude greater than τ_s.

These contradictory results can be explained if we assume that a thin film of a comparatively low-density n' electron-hole plasma is formed between the drops and the crystal surface. The surface-recombination time τ_s' in this case is determined by the ratio of n' to the particle density n_0 in the liquid: $\tau_s' = \tau_0\, n_0/n' \gg \tau_s$. We assume that the drop wets the surface; if the drop radius is $R \sim (10^{-4}$–$10^{-5})$ cm, then the pressure in the film will be equal to the capillary pressure $p \approx 2\sigma/R \sim 5$–$50$ dyne/cm^2. For a drop of larger radius (in the limit—for the layer of liquid that appears on the surface at a high excitation level) the capillary pressure almost vanishes, but, as a rule, the external pressure caused by a phonon wind or by nonuniform deformation remains (the force that acts on a particle pair in the layer is 10^{-14}–10^{-15} dyne, which gives a pressure $p \sim 0.1$ dyne/cm^2 for a 1-μm-thick layer). If $n' \sim p$, i.e., the film is "gaslike", then the recombination time must increase sharply in the transition from an individual drop to a layer.

The concept of the surface film was used in an experiment in which the dependence of the surface lifetime on the drop generation rate and on the force that presses the drop to the surface was investigated. The wide face of a $1 \times 5 \times 10$-mm pure Ge sample was illuminated with a pulsed GaAs laser; the diameter of the light spot was ≈ 4 mm, and the pulse duration of the light was ≈ 3 μsec. We sputtered a thin metal layer (thermal contact) to the opposite face of the sample, through which a 1- to 3-μsec current pulse, which was locked-in with the laser pulse, was passed. The thermal pulse propagated through the crystal and ejected drops to the sample surface.[2] The dependence of the integrated (with respect to time) radiation intensity I of the electron-hole liquid on the excitation intensity G was recorded for different thermal pulse parameters.

Typical results are shown in Fig. 1. Curve 1 represents the $I(G)$ dependence at $T = 4.2$ K and defines the scale of the characteristic excitation levels: The threshold of drop appearance corresponds to an average exciton density $\bar{n} \approx 10^{14}$ cm^{-3}. At $T = 1.65$ K the condensation threshold decreases by more than two orders of magnitude,[1] so that the $I(G)$ dependence is almost linear (curve 2). An increase of the slope dI/dG means that the drop lifetime increases with decreasing temperature. If the thermal pulse is initiated simultaneously with the light pulse, then, starting with a certain critical value of the thermal pulse, the slope of the $I(G)$ dependence decreases; in this case the threshold G^* of radiation increases sharply (curves 3, 4, and 5). At $G < G^*$ there is almost no radiation, although the threshold for the appearance of drops at this temperature is $G_+ \ll G^*$ (the fact that the sample had not been overheated by the thermal pulse was checked by analogous experiments at $T = 4.2$ K). The drops produced at $G_+ < G < G^*$ reach the surface quickly and recombine in a time close to τ_s. Assuming that the radiation signal of the drops at $G < G^*$ is close to the noise (Fig. 1), we can estimate the maximum lifetime of individual drops

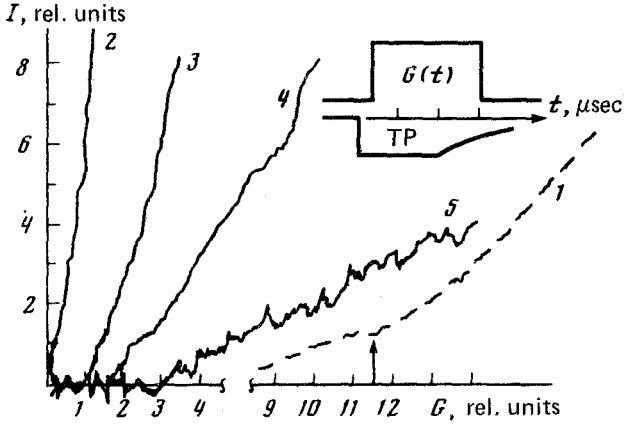

FIG. 1. Dependence of the recombination-radiation intensity I of electron-hole liquid on the excitation intensity G. 1, $T = 4.2$ K. Condensation threshold is indicated by the arrow. 2–5, $T = 1.65$ K. The duration of the thermal pulse is $\theta = 1.2$, 1.6, and 2 μsec for curves 3, 4, and 5, respectively. For a given amplitude of the thermal pulse (TP) the variation in the drop velocity is caused by the diffusion "tail" of phonons (see inset). A result similar to that in Fig. 1, curves 3–5, is obtained at a constant duration of the thermal pulse $\theta = 3$ μsec and at a varying amplitude.

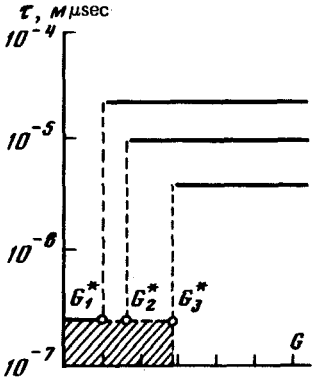

FIG. 2. Dependence of the lifetime of the electron-hole liquid on the pumping G; the values G_1^*-G_3^* correspond to Fig. 1.

τ_s': $\tau_s'/\tau_0 \approx 10^{-2}$. Consequently, $\tau_s' \leqslant 4 \times 10^{-7}$ sec. At $G > G^*$ the slope of the curves 3, 4, and 5, which determines the lifetime of the liquid, depends on the amplitude of the thermal pulse; it can be seen that the lifetime changes abruptly (Fig. 2) and that the lifetime and threshold pumping G^* are related to the intensity of the photon stream that presses the liquid to the surface.

By using the experimental values of τ_s' and G^* we can estimate the particle density in the surface film. We shall examine a liquid layer near the surface (Fig. 3). The layer of thickness d recombines in a time $\tau_s' = dn_0/sn'$. Assuming that $d \approx 10^{-4}$ cm, $s \approx 10^2$ cm/sec, $n_0 = 2 \times 10^{17}$ cm^{-3}, and $\tau_s' = 3$-20 μsec (Fig. 2), we obtain $n' \approx (1$-$5) \times 10^{16}$ cm^{-3}. On the other hand, the condition $n''v > n's$, where v is the velocity at which the drop moves toward the surface and n'' is the average liquid density to the right of the layer (Fig. 3), must be satisfied in order for the layer to be excited. Assuming that $n'' \approx 10^{13}$ cm^{-3} (Fig. 1) and $v \sim 10^5$ cm/sec, we obtain $n' \sim 10^{16}$ cm^{-3}.

The nature of the gas film is not clear at present. We can assume that the surface states act on the drop the same way as the impurities that are located in the bulk of the crystal, i.e., the drop is drawn to the surface and its density decreases.[1] At the surface there is normally a large density of recombination centers, 10^{11}-10^{12} cm^{-3} (Ref. 5); this corresponds to a bulk density of impurities equal to 10^{16}-10^{18} cm^{-3}. It has been shown in several papers (see, for example, Ref. 6) that the liquid phase generally does not appear in heavily doped Ge samples. If it is assumed that the particles at the surface enter a region of "heavy doping," then they must leave the

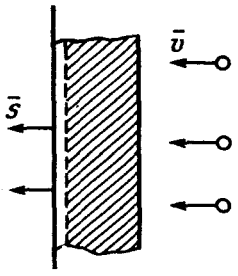

FIG. 3. Diagram of growth and annihilation of a liquid layer at the sample surface. V is the rate of rise of the drops to the surface and s is the surface recombination rate. The gaslike layer is located between the dashed line and the surface.

liquid phase and pass into surface states. A rough estimate of the density n' can be obtained by equating the minimum external pressure p to the intrinsic Fermi gas pressure: $p \sim \hbar^2/m\,(n')^{5/3}$; at $p = 10^{-1}$ dyne/cm^2 $n' \sim 10^{15}$ cm^{-3}.

The dependence of the lifetime of the surface on the drop size should influence the formation kinetics of the liquid layer. A small drop that falls to the surface normally recombines before another drop is able to fall. The recombination rate of a large drop is slower than that for a small drop; consequently, for a given stream of drops to the surface there exists a liquid "puddle" of critical size, for which the gowth rate is equal to the recombination rate. The formation kinetics for a drop of critical size are analogous to the kinetics of a critical seed for the vapor-liquid transition.[7] However, the decomposition rate of a drop of critical size is determined by surface recombination rather than by evaporation. The foregoing comments make it possible to call the transition from individual drops to a layer a kinetic phase transition.

Using the obtained results, we can explain the radiation threshold in thin Ge samples.[8] This threshold was found to be very high ($\gtrsim 10^{14}$ cm^{-3}) and only slightly temperature dependent. The fact is that the true threshold and the condensation thresholds in a thin sample coincide with these in a massive sample and at a temperature $T \approx 1.7$ K they correspond to an average exciton density of $\approx 10^{12}$ cm^{-3}. In a thin sample, however, the drops rise to the surface rapidly and have a very short lifetime until a layer is formed. The radiation threshold observed at low temperatures is attributed to the kinetic phase transition, i.e., to the formation of a surface layer of liquid. This also apparently applies to experiments in which threshold clearing of thin Ge samples has been observed.[9]

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