Instability and motion of electron-hole drops

Yu. A. Bychkov, S. V. Iordanskiĭ, and É. I. Rashba

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences

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A theory of recombination instability of electron-hole drops due to the existence of a condensation flow from the gas (i.e.,exciton) phase to the surface of the drops and the recombination flow inside the drop is constructed. When the spherical shape of the drop is distorted these flows have a tendency to enhance the initial deformation; if their influence prevails over the surface tension, the drops become unstable. An instability criterion is derived and the singularities of the instability evolution are investigated for small droplets in the course of their rapid growth and for drops close to their stationary size. Drop motion in an inhomogeneous exciton gas is also considered, both in the diffusion and in the Knudsen regime. It is shown that, depending on the parameter values, the drops can move both with and against the exciton-concentration gradient. The interaction of the drops via the exciton gas can cause the excitions to move both toward and away from one another.

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1. INTRODUCTION

The existence of electron-hole drops, to which an extensive literature has been devoted during the last decade, is by now a well established fact. The research results and the noted prospects were expounded in a number of reviews.[1-4] It appears that the least explained aspect of the problem is at present the kinetics of the drops—their formation, evolution, motion, and splitting. Notwithstanding the many recent papers in this direction (see, e.g., Refs. 5–7), the difficult problem of drop kinetics is apparently still far from solved.

We consider in this article several questions connected with the kinetics of drops: new possible mechanisms of their instability, motion of drops in the gradient of the exciton concentration, and the interaction of the drops with one another. It should be noted that certain parameters of the excitons and drops are still insufficiently well known; in addition, they depend on the temperature and may be structure-sensitive (i.e., they may vary from sample to sample) and differ substantially for different semiconducting materials. It is therefore not our purpose to develop a quantitative theory for some fully defined system. On the contrary, starting from a relatively realistic model, we consider related phenomena in various ranges of parameters and determine which of them are sufficiently general and which are peculiar only to definite region or behave differently in different regions.

2. RECOMBINATION INSTABILITY

The drop-instability mechanisms considered in the present papers are connected with the anisotropy of the flows in the gaseous exciton phase and the liquid electron-hole phase at a small deviation of the drop shape from spherical. The quantitative theory is presented in Sec. 3. Since it is quite complicated, we precede it with a quantitative picture to make clear the principal considered physical mechanisms.

We consider an electron-hole drop surrounded by an exciton gas. We assume that the exciton mean free path in the gas phase Λ_g and the electron and hole mean free path in the liquid phase Λ_l are small compared with the drop radius $R(\Lambda_g, \Lambda_l \ll R)$. The continuous-medium approximation is valid for the description of either phase. We start with the instability mechanism connected exclusively with the condensation flow of the excitons in the gas phase into the drop. At $\Lambda_g \ll R$ the condensation flow is diffuse and the exciton concentration near the drop surface is lower (and possibly much lower) than their concentration n_∞ at large distances from the drop; we denote the concentration difference by Δn.

Imagine now that the fluctuation caused the drop to lose its spherical shape and assume, for example, the shape of an egg, as in Fig. 1. Then the narrower end, being farther from the drop center, is located in a region with higher exciton density; the condensation flow to this end is therefore stronger. On the contrary, the flatter part turns out to be in a region with a lower exciton concentration, and therefore the condensation flow to it is weaker. Thus, the nonuniformity of the condensation flow of the excitons to the drop enhances the initial fluctuation and can make the spherical drop unstable and cause it to break up into smaller droplets. We now derive an instability criterion from qualitative considerations.

We characterize the deviation of the drop from a

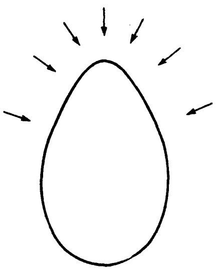

FIG. 1. Drop-instability mechanism connected with the anisotropy of the condensation flows from the gas phase.

sphere by the dimensionless paramter ε. The change of ε consists of an increase due to the condensation flow and a decrease due to restoration of the spherical shape under the influence of the surface-tension forces:

$$d\varepsilon/dt=(d\varepsilon/dt)_{\text{cond}}+(d\varepsilon/dt)_{\text{surf. tens.}} \tag{2.1}$$

The density of the diffusion flow on the surface drop $j\sim D\Delta n/R$ where D is the exciton diffusion coefficient. The rate of change of the radius drop due to this flow is $R/R\sim D\Delta n/NR^2$, where N is the concentration of the electron-hole pairs in the liquid phase. It is convenient to rewrite this expression in terms of R_{st}, the radius of the drop in a stationary gas having the same density. In such a drop the total diffusion flow to its surface is equal to the recombination flow in it: $jR_{st}^2 \sim R_{st}^3 N/\tau_0$; here τ_0 is the lifetime in the drop. Therefore

$$D\Delta n/N\sim R_{st}^2/\tau_0 \tag{2.2}$$

and $R/R\sim(R_{st}/R^2\tau_0^{-1}$. If we assume that the change of the condensation-flow density at low nonsphericity ε is proportional to ε, then

$$\left(\frac{d\varepsilon}{dt}\right)_{\text{cond}}=\frac{\alpha_1}{\tau_0}\left(\frac{R_{st}}{R}\right)^2\varepsilon,\quad \alpha_1\sim1. \tag{2.3}$$

To estimate the second term in (2.3), we note that the rate of restoration of the shape of the drop by the surface tension is limited by the friction between the electron-hole liquid and the lattice. We shall describe this friction by the force $\rho\mathrm{u}/\tau_l$ per unit volume, where $\rho=mN$, N is the density of the liquid, m is the exciton mass, u is the velocity of the liquid, and $\tau_l\sim\Lambda_l/v_F$, v_F is the Fermi velocity in the liquid. It is possible, however, to combine the surface tension σ, the radius R, the parameter ρ/τ_l, i.e., the three quantities on which the second term of (2.1) depends, into a single combination with the dimension of time. As a result we have

$$\left(\frac{d\varepsilon}{dt}\right)_{\text{surf. tens.}}=-\alpha_2\frac{\sigma\tau_l}{\rho R^3}\varepsilon,\quad \alpha_2\sim1. \tag{2.4}$$

Instability sets in when the small ε and its derivative $d\varepsilon/dt$ are of the same sign. From (2.1)–(2.4) we get directly the instability criterion

$$R>R_3^{is}\sim\frac{\sigma\tau_0\tau_l}{\rho R_{st}^2}\sim\frac{\sigma\tau_l N}{D\rho\Delta n}. \tag{2.5}$$

We emphasize that the recombination time ε does not enter in the instability criterion. This is a reflection of the fact that the instability is due entirely to the condensation flow to the drop, and not to the recombination flow inside the drop. It is therefore natural to call it the condensation instability. A similar instability can occur in a system of atoms adsorbed on a surface when "two-dimensional" gas condenses into drops of a "two-dimensional liquid."

We proceed now to consider the second instability mechanism, due to the presence of recombination flows in the liquid phase. In the presence of friction between the liquid and the lattice, the recombination flow can be maintained inside the drop only on account of an excess pressure that increases towards the periphery. It is easy to verify that this pressure is $p(r)\sim\rho r^2/\tau_0\tau_l$, where r is the distance from the center of the drop. When the drop differs from a sphere both the distribution of the recombination and the distribution of the pressure become anisotropic. Disregarding for the time being the presence of gaskinetic and capillary contributions to the pressure, the total pressure drop between the center of the drop and the surface is the same. But at fixed r the pressure at different points will differ by an amount $\delta p\sim\varepsilon\rho R^2/\tau_0\tau_l$. This statement is illustrated by Fig. 2, which shows the pressure patterns for a drop with elongated (cigar) shape. It is seen that for such a drop the flows in the liquid are predominantly meridional, from the equitorial plane to the symmetry axis, i.e., they enhance the nonsphericity.

Since the meridional flows produced by the pressure δp are limited by the friction force characterized by the coefficient ρ/τ_l, it follows from dimensionality considerations that the corresponding contribution to $d\varepsilon/dt$ is of the order of

$$(d\varepsilon/dt)_{\text{pres}}=\alpha_3\varepsilon/\tau_0,\quad \alpha_3\sim1. \tag{2.6}$$

It replaces the first term in (2.1); the previous expression (2.4) remains in force for the second term. As a result we obtain the instability criterion

$$R>R_2^{is}\sim(\sigma\tau_0\tau_l/\rho)^{1/4}. \tag{2.7}$$

It will be shown in Sec. 3 below that this instability develops at all values $L\geqslant2$, and the critical radius is minimal for the quadrupole instability $L=2$; this in fact is the meaning of subscript in R_2^{is}. Condensation instability, on the contrary, exists within the framework of the model of Sec. 3 only at $L\geqslant3$; this is indicated by the subscript of R_3^{is} in (2.5).

It is important that R_2^{is} contains τ_0 explicitly. Therefore this instability is directly connected with the recombination in the drop; in this sense it is natural to call it recombination instability. The recombination-instability criterion (2.7) was obtained earlier[8] within the framework of the hydrodynamic model.[9] The theory of condensation and recombination instabilities will be developed in the next section. Since they are interrelated and cannot be separated in all cases, we shall frequently use the common term "recombination instability" for both.

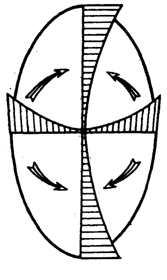

FIG. 2. Instability mechanism connected with the presence of recombination flows inside the drop. These flows are due to anisotropic distribution of the pressure, and have a meridional direction from the equator if the drop is cigar-shaped. The pressure distribution patterns are shown along the symmetry axis and in the equatorial plane.

Condensation instability can appear in the course of the growth of the drops only if $R_3^{is} < R_{st}$ or

$$\frac{\Delta n}{N} \gtrsim \frac{1}{D\tau_0}\left(\frac{\sigma\tau_0\tau_l}{\rho}\right)^{2/3}, \tag{2.8}$$

i.e., at sufficiently large supersaturations. Since $R_3^{is} \sim (R_2^{is})^3/R_{st}^2$, the recombination-instability criterion is more stringent. However, even in the case when the criterion (2.8) is satisfied, if the drop has a small initial nonsphericity ε the time $\sim\tau_0$ corresponding to the drop growth may not be sufficient for the instability to advance enough to cause splitting of the drop. It is therefore important to know whether this instability can develop during the stage when R is close to R_{st}. Assuming $R \approx R_{st}$ in (2.3) and comparing with (2.4), we obtain for the instability of a drop with a stationary radius a criterion that coincides exactly with (2.7). Consequently, both instabilities develop simultaneously in the region $R \approx R_{st}$.

It is obvious that R_2^{is} sets the upper bound of the drop sizes in the stationary regime, for in that case n_∞ does not exceed a value n_∞^0 such that $R_{st}(n_\infty^0) \approx R_2^{is}$. The determination of the drop distribution in size in the region of smaller radii is a difficult problem of kinetics.

3. THEORY OF RECOMBINATION INSTABILITY

To construct a theory for recombination instability we start with a simplified isotropic scheme, assuming that the mean free path is small compared with R in both the gas and in the liquid phase ($\Lambda_g, \Lambda_l \ll R$) and that we can neglect both the drop heating due to the recombination processes and the influence of the electric fields due to the exciton dissociation. The principal mechanism of drop instability is presently taken to be the phonon wind proposed by Keldysh.[6] The shape instability can be due also to electron-phonon interaction in the drop[10]; the electric charge of the drops[3,11] can lead to phenomena of the type of electrocapillary instability. We do not consider these effects here. Some additional limitations will be formulated below.

The problem is divided into solution of the equations in the gas and liquid phases and the matching of the solutions on their boundary.

A. Gas phase. The exciton motion can be described by the differential equation

$$\partial n/\partial t = D\nabla^2 n - n/\tau_r + G. \tag{3.1}$$

If $R \ll (D\tau_r)^{1/2}$, which holds true at not too short lifetimes τ_r in the gas phase, then recombination can be neglected in the actual region at scales $\sim R$. In the same region we can also leave out the exciton photogeneration ($G = 0$). Assuming also that the characteristic reciprocal times $\lambda \ll D/R^2$ (it will be clear later on that this is equivalent to $R^2 \ll D\tau_0$), we can omit also $\partial n/\partial t$ from (3.1), which reduces then to $\nabla^2 n = 0$ with solutions

$$n(\mathbf{r}) = \left(n_\infty - \frac{A}{r}\right) + \sum_{LM} n_{LM}, \quad n_{LM} = -\frac{B_{LM}}{r^{L+1}} Y_{LM}, \tag{3.2}$$

where Y_{LM} are spherical harmonics. The spherically symmetric part of the solution corresponds to a stationary distribution of the concentration and satisfies the boundary condition $n(r \to \infty) = n_\infty$, with n_∞ determined only by the pump strength, and n_{LM} corresponding to the quasistationary perturbations due to the change of the drop shape.

The constants A and B_{LM} are determined by the boundary conditions on the surface of the drop. The difficulty is that Eq. (3.1) may no longer be valid near the surface. In fact, assume by way of estimate that at $r = R$ the concentration $n(R) \approx n_T$, which is the equilibrium exciton concentration at the surface of the liquid phase[1] at the temperature T. Determining A and estimating the concentration change δn in a layer $\sim\Lambda_g$ at the drop surface, we get

$$\frac{\delta n}{n_T} \sim \frac{\Lambda_g}{R}\frac{n_\infty - n_T}{n_T}.$$

At low temperatures $n_T \ll n_\infty$ and the condition $\delta n \ll n_T$ may be violated even at $\Lambda_g \ll R$. At the surface layer we must therefore replace the diffusion equation by the kinetic equation, but the latter, owing to the condition $R \gg \Lambda_g$, can be solved in the quasi-planar approximation.

We consider now the kinetic equation in the approximation of the effective collision time τ_g:

$$v_z \partial f/\partial z + (f - \bar{f})/\tau_g = 0. \tag{3.3}$$

Its solution in the half-space $z > 0$ is of the form

$$f(z, \mathbf{v}) = f_+ = f_0(v)\exp\left(-\frac{z}{v_z\tau_g}\right) + \frac{1}{\tau_g v_z}\int_0^z \exp\left(-\frac{z-z'}{\tau_g v_z}\right)$$

$$\times \bar{f}(z', v)\,dz', \qquad v_z > 0, \tag{3.4a}$$

$$f(z, \mathbf{v}) = f_- = \frac{1}{\tau_g v_z}\int_\infty^z \exp\left(\frac{z'-z}{\tau_g v_z}\right)\bar{f}(z', v)\,dz', \quad v_z < 0. \tag{3.4b}$$

Here $\bar{f}$ is obtained by averaging f over the angles:

$$\bar{f}(z, v) = \frac{1}{4\pi}\int d\Omega\, f(z, \mathbf{v}); \tag{3.5}$$

the free-path time τ_g depends generally speaking on the velocity v. The solution (3.4) satisfies the boundary conditions.

On the drop surface ($z = 0$) the distribution function of the departing particles ($v_z > 0$) is at equilibrium:

$$f_+(z=0, v, v_z>0) = f_0(v) = n_T\varphi_0(v),$$

where $\varphi_0(v)$ is a normalized Maxwellian function. This condition corresponds to the assumption that the sticking coefficient of the excitons on the surface is equal to unity ($\gamma = 1$). The validity of this assumption is not obvious, but it simplifies the calculations greatly.

Substitution of (3.4) in (3.5) leads after simple transformations to the self-consistency condition

$$\bar{f}(z, v) = f_0(v) + \frac{1}{2\Lambda_g}\int_0^\infty E_1\left(\frac{|z-z'|}{\Lambda_g}\right)(\bar{f}(z', v) - f_0(v))\,dz', \tag{3.6}$$

where

$$E_1(u) = \int_1^\infty e^{-ut}\frac{dt}{t}. \tag{3.7}$$

Finally,

$$\bar f(z,\ v)=f_0(v)+[\bar f(z=0,v)-f_0(v)]M(z/\Lambda_g), \tag{3.8}$$

where $M(\zeta)$ is the solution of the Milne equation[12]

$$M(\zeta)=\frac{1}{2}\int\limits_0^\infty E_1(|\zeta-\zeta'|)M(\zeta')d\zeta', \quad M(0)=1. \tag{3.9}$$

This equation has been well investigated, so that with the aid of (3.4) and (3.8) we can express the complete solution in terms of the function $\bar f(v)\equiv\bar f(z=0,v)$. The form of this function is determined from the condition that in the region $\Lambda_g \ll z \ll R$ the solution of the Milne equation must be joined to the solution of the diffusion equation:

$$\partial\bar f(z,\ v)/\partial z\approx(dn/dz)_{z\to 0}\varphi_0(v)\equiv n_s'\varphi_0(v).$$

Here n_s' is the limiting value of the derivative of the solution of the diffusion equation as $z\to 0$, and finally does not coincide with the exact value of the derivative of $\bar f$ at $z=0$ integrated over the velocities. Using the known asymptotic form $M(\zeta)\approx\sqrt{3}(\zeta+q_\infty)$, $q_\infty\approx 0.710$, which is valid at $\zeta\gg 1$, we obtain in lieu of (3.8)

$$\begin{gathered}\bar f(z,\ v)=\{n_T+3^{-1/2}n_s'\Lambda_g(v)M(z/\Lambda_g)\}\varphi_0(v)\\ \approx\{n_T+n_s'(z+q_\infty\Lambda_g(v))\}\varphi_0(v), \quad z\gg\Lambda_g.\end{gathered} \tag{3.10}$$

Calculating with the aid of (3.4) the density j_s of the exciton flow to the drop and the pressure on it as the momentum transfer, and taking into account the properties of the function $M(\zeta)$, we obtain

$$\begin{gathered}j=-Dn_s',\\ p=n_s^*T,\ n_s^*=n_T+q_\infty\langle v^2\Lambda_g\rangle n_s'/\langle v^2\rangle\equiv n_T+\Lambda^* n_s',\end{gathered} \tag{3.11}$$

where the angle brackets denote averaging over the Maxwellian distribution.

B. Liquid phase. In the description of the hydrodynamics of the liquid phase, we confine ourselves to the incompressible-liquid approximation. Then, the linearized Euler equation supplemented with the friction against the lattice,[2)] becomes

$$\partial\mathbf{u}/\partial t+\mathbf{u}/\tau_l=-\nabla p/\rho, \tag{3.12}$$

and the continuity equation becomes

$$\operatorname{div}\mathbf{u}=-\tau_0^{-1}. \tag{3.13}$$

From these we get an equation for the pressure

$$\nabla^2 p=\rho/\tau_0\tau_l, \tag{3.14}$$

whose solution is

$$p=p_0+\frac{\rho r^2}{6\tau_0\tau_l}+\sum_{LM}\pi_{LM}r^L Y_{LM}. \tag{3.15}$$

In the quasistationary approximation $\mathbf{u}\approx-\tau_l\nabla p/\rho$ or

$$u_r^0=-\frac{r}{3\tau_0}, \quad u_r^{LM}=-L\frac{\tau_l}{\rho}\pi_{LM}r^{L-1}Y_{LM}; \tag{3.16}$$

here u_r is the radial velocity component.

The linearized equation (3.12) is valid at $|(\mathbf{u}\nabla)\mathbf{u}| \ll |\mathbf{u}/\tau_l|$ or $\tau_l\ll\tau_0$. Allowance for the viscosity, i.e., the term $\eta\nabla^2\mathbf{u}/\rho$ in (3.12), leads to corrections of the order of

$$\eta\tau_l/\rho R^2\sim\Lambda_{ee}\Lambda_l/R^2\ll 1,$$

where Λ_{ee} is the mean free path with respect to electron-electron collisions; usually $\Lambda_{ee}\ll\Lambda_l$.

C. Boundary conditions. The boundary conditions reduce to continuity of the flows and to mechanical equilibrium of the surface. For the spherically symmetrical part of the flow with account taken of $\Lambda_g\ll R$ it follows from (3.2) and (3.11) that

$$n_s'=A/R^2\approx(n_\infty-n_T)/R. \tag{3.17}$$

It is of interest to note that this formula corresponds to retention of only the first term in n_s^* [see (3.11)]. The second term can be omitted even if it is of the order of or larger than n_T, since it competes in the equations not with n_T, but with the term $\sim Rn_s'$, which exceeds it in the ratio $R/\Lambda_g\gg 1$.

The flow-continuity condition yields, when account is taken of (3.16) and (3.17),

$$\dot R=\frac{D}{R}\frac{n_\infty-n_T}{N}-\frac{R}{3\tau_0}. \tag{3.18}$$

From this we get the known equation for the stationary drop radius[7]

$$R_{st}^2=3D\tau_0(n_\infty-n_T)/N; \tag{3.19}$$

its characteristic growth time is $3\tau_0$.

Let now the drop deviate somewhat from a sphere, so that its radius in the direction $\Omega=(\vartheta,\varphi)$ is

$$R(\Omega)=R+\xi Y_{LM}(\Omega), \quad |\xi|\ll R. \tag{3.20}$$

Replacing in (3.2) the radius $r=R(\Omega)+z$, expanding up to terms $\sim\xi z$, and matching the obtained expansion to the quasi-planar solution, we obtain, allowing for (3.11), in the lowest nonvanishing order in Λ_g/R, the limiting values of the parameters on the gas-phase side

$$\begin{gathered}(n_s')_{LM}=(L-1)\frac{n_\infty-n_T}{R^2}\xi,\\ (j_g)_{LM}=-D(n_s')_{LM}, \quad (p_g)_{LM}=T\Lambda^*(n_s')_{LM}.\end{gathered} \tag{3.21}$$

Writing down the equations for the pressure inside the liquid and for the capillary motion (see Ref. 13, Sec. 61), we get from (3.15), (3.16), and (3.20) the expressions for the corresponding contributions:

$$\begin{gathered}(p_l)_{LM}=\rho R\xi/3\tau_0\tau_l+\pi_{LM}R^L,\\ (p_c)_{LM}=(L-1)(L+2)\frac{\sigma}{R^2}\xi.\end{gathered} \tag{3.22}$$

Analogously, for the velocity of the normal liquid flow we have

$$u_{LM}=-\frac{\xi}{3\tau_0}-L\frac{\tau_l}{\rho}\pi_{LM}R^{L-1}. \tag{3.23}$$

The boundary conditions are

$$\dot\xi=\frac{D}{N}(n_s')_{LM}+u_{LM}, \quad (p_l)_{LM}=(p_c+p_g)_{LM}. \tag{3.24}$$

The second of these equations allows us to determine π_{LM}. Using also (3.21) and (3.23), we get

$$\frac{\dot\xi}{\xi}=\frac{L-1}{3\tau_0}\left(\frac{R_{st}}{R}\right)^2+\frac{L-1}{3\tau_0}\left\{1-3L(L+2)\frac{\sigma\tau_0\tau_l}{\rho R^3}-L\frac{\Lambda^*T\tau_l}{mDR}\left(\frac{R_{st}}{R}\right)^2\right\}. \tag{3.25}$$

The presence of the factor $L-1$ ensures a neutral equilibrium of the drop with respect to translation of its center.

For the instability condition at $L \geqslant 2$ we choose the growth of the relative deformation ξ/R; this quantity is analogous to the nonsphericity parameter ε used in Sec. 2. We then get from (3.18) and (3.25) an equation for the increment:

$$\lambda = \frac{d}{dt}\ln\frac{\xi}{R} = \frac{L-2}{3\tau_0}\left(\frac{R_{st}}{R}\right)^2 + \frac{L}{3\tau_0}\left\{1-3(L-1)(L+2)\frac{\sigma\tau_0\tau_l}{\rho R^3}-(L-1)\frac{\Lambda\cdot T\tau_l}{mDR}\left(\frac{R_{st}}{R}\right)^2\right\}. \tag{3.26}$$

This equation is our principal result, and the remainder is essentially its analysis. It is useful therefore to clarify the physical meaning of the individual terms. The first term is connected with the condensation flow from the gas phase. The second term is connected with various components of the pressure and its three members are respectively the anisotropic part of the pressure due to the recombination flow in the liquid, the capillary pressure, and the gaskinetic pressure on the liquid surface. It is seen that the two first contributions are positive and give rise therefore to instability, while the last two are negative and therefore suppress the instability.

It is easy to verify by taking (3.19) into account that the ratio of the gaskinetic and capillary pressures is of the order of $\Lambda_g \Delta p/\sigma$, where $\Delta p = T(n_\infty - n_T)$. It appears that this ratio is usually small, and to simplify the calculations we shall omit the gaskinetic pressure.

It is seen from (3.26) that since the first term vanishes at $L=2$, the conditions for the development of instabilities with $L=2$ and $L>2$ can differ substantially.

We begin with the case $L=2$. The competing terms are here the volume and capillary pressures, and instability exists at $R > R_2^{is}$:

$$R_2^{is} = (12\sigma\tau_0\tau_l/\rho)^{1/3}. \tag{3.27}$$

We called this the recombination instability mechanism in Sec. 2; Eq. (3.27) for the critical radius was obtained earlier.[8] It follows also from (3.26) that the increment $\lambda \sim \tau_0^{-1}$. i.e., the instability evolves within a lifetime on the order of the time required for the drop to grow to the size $R \sim R_{st}$.

We proceed now to instabilities with $L>2$, when the first term of (3.26) differs from zero. The difference from the case $L=2$ is particularly large at high concentrations in the gas phase, when $R_{st} \gg R_2^{is}$; the condensation contribution to the instability is predominant here. The instability occurs at $R > R_L^{is}$:

$$R_L^{is} = \frac{3(L-1)L(L+2)}{(L-2)}\frac{\sigma\tau_0\tau_l}{\rho R_{st}^2} = \frac{(L-1)L(L+2)}{4(L-2)}\frac{(R_2^{is})^3}{R_{st}^2}. \tag{3.28}$$

The coefficient is minimal at $L=3$, when

$$R_3^{is} = 7.5(R_2^{is})^3/R_{st}^2. \tag{3.28'}$$

This quantity may turn out to be much smaller than R_2^{is}.

The kinetics of the development of instability with $L>2$ is of interest. According to (3.26), its characteristic growth time is

$$\tau_L \sim \tau_0 (R_L^{is}/R_{st})^2 \ll \tau_0. \tag{3.29}$$

It is therefore natural to assume that the breakup of the drops is in a scale $\sim R_L^{is}$ with a time interval $\sim\tau_L$. We note also that $R_L^{is} \propto (n_\infty - n_T)^{-1}$, i.e., it decreases with increasing exciton concentration. On the other hand if $R_{st} \sim R_2^{is}$, then $R_2^{is} < R_L^{is}$ with $L>2$ because of the fast increase of the coefficient of the capillary pressure in (3.26) with increasing L. Thus, the character of the instability depends strongly on the exciton concentration: at relatively low concentrations there should develop an instability with $L=2$, which should give way at high concentration to instability with $L=3$.

We have compared above the gaskinetic pressure, i.e., the last term of (3.26), only with the capillary pressure. It is instructive, however, to compare it also with the first two terms, since instability is possible only if it is smaller than their sum. The necessary condition for this, in order of magnitude, of the form $v_T\tau_l/R < 1$. It restricts rather strongly the radii at which instability is possible. The hybrid parameter $v_T\tau_l/R$, which includes the velocity in the gas v_T and the relaxation time τ_l, will be encountered in Sec. 4 below; it characterizes the ratio of the effects connected with asymmetric momentum and mass flows when the exciton gas condenses.

The feasible numerical estimates are quite crude even in the case of germanium, on which most research was done. If we assume $D \sim 10^3$ cm^2/sec, $\tau_0 \sim 10^{-4}$ sec, $\tau_l \sim 10^{-9}$ sec, $\sigma \sim 10^{-4}$ dyn/cm, $\rho \sim 10^{-10}$ g/cm^3, $n_\infty \sim 3\cdot 10^{14}$ cm^{-3}, $N \sim 10^{17}$ cm^{-3}, then we get $R_{st} \sim 3\cdot 10^{-2}$ cm and $R_2^{is} \sim R_3^{is} \sim 10^{-2}$ cm. Since the results are close, this estimate permits no choice between the mechanisms for the cases $L=2$ and $L=3$. Both values, however, are larger by approximately a decade than the experimental one. This allows us to give preference to the mechanism with $L=3$, inasmuch as decreasing R_2^{is} by one-half ensures a decrease of R_3^{is} by a decade and reconciles the theoretical estimates with the experimental data.

Of course, the foregoing estimate is in no way proof that the mechanism of recombination instability of the drops has actually been observed experimentally in germanium. It does make it meaningful to bear this mechanism in mind together with phonon wind.[6]

We note in conclusion the following circumstance. The condensation mechanism, which corresponds to the first term in (3.26), is essentially connected with the anisotropy of the diffusion flows in the gas phase and should vanish at $\Lambda_g \gg R$, i.e., in a Knudsen gas. On the contrary, the instability with $L=2$ is determined predominantly by processes inside the drop and would be preserved at $\Lambda_g \gg R$ if the regime inside the drop were to remain hydrodynamic, i.e., $\Lambda_l \ll R$. As a result of Fermi degeneracy, however, we have inside the drop $\Lambda_l \gg \Lambda_g$ (Ref. 14); such a situation is therefore not realized.

4. MOTION OF DROPS IN A CONCENTRATION GRADIENT AND DROP INTERACTION VIA THE GAS PHASE

The question of the behavior of drops in an exciton-concentration gradient was discussed in a number of papers. It was proposed, in particular, that it is precisely the inhomogeneous distribution of the exciton concentration in the gas phase which is responsible for the observed motions in the drop cloud. It will be shown below that both the drop velocity and the dominant mechanism of their displacement depend very strongly on the relations between a number of parameters. In particular, we shall find it convenient to consider separately the cases $R \gg \Lambda_g$ and $R \ll \Lambda_g$, which correspond to the diffusion and to the Knudsen regimes in the vapor phase.

In all cases, however, regardless of the ratios of the parameters, we can separate two principal physical mechanisms that lead to displacement of the drop. The first is the transfer of momentum to the drop in account of the asymmetry of the flows and of the concentrations in the gas phase, as a result of which the liquid acquires a hydrodynamic velocity, and the stationary regime is reached via friction. The second mechanism is connected with the asymmetric condensation of the excitons because of the supersaturated vapor on the drop; it leads to motion of the center of gravity of the drop Z without motion of the liquid in the drop relative to the lattice. We shall call these the hydrodynamic and condensation mechanisms, respectively. They lead to opposite directions of particle motion: with and against the gas flow in the first and second cases, respectively. The equations that describe these processes are

$$\frac{d}{dt}(M\mathbf{V}_h) = -\oint \hat{\Pi}\, d\mathbf{S} + \mathbf{F}, \tag{4.1}$$

$$M\frac{d\mathbf{Z}}{dt} = -\oint (\mathbf{R}-\mathbf{Z})(\mathbf{j}_M\, d\mathbf{S}), \quad \mathbf{V}_c = \frac{d\mathbf{Z}}{dt}, \tag{4.2}$$

where $\hat{\Pi}$ and $\mathbf{j}_M$ are the momentum and mass flows from the gas to the surface of the drop, $\mathbf{V}_h$ and $\mathbf{V}_c$ are the hydrodynamic and condensation contributions to the total drop velocity: $\mathbf{V} = \mathbf{V}_h + \mathbf{V}_c$, $\mathbf{F}$ is the effective friction force that describes the momentum transfer from the drop to the lattice, $\mathbf{R}$ is a vector that runs over the surface of the drop, and $d\mathbf{S}$ is oriented along the outward normal to the liquid.

We assume that in the absence of the drop the concentration in the gas phase has a linear variation

$$n(\mathbf{r}) = n_\infty + \mathbf{r}\nabla n_\infty, \tag{4.3}$$

with

$$|\nabla n_\infty|/n_\infty \ll \min\{\Lambda_g^{-1}, R^{-1}\}.$$

A. Drop motion in the diffusion regime ($R \gg \Lambda_g$). Solving the diffusion equation $\nabla^2 n = 0$ (see Sec. 3A) with the boundary conditions on the drop surface $n_s = n_t$ and with a condition at infinity compatible with (4.3), we obtain for the concentration around a drop with center at the point $\mathbf{Z} = 0$

$$n(\mathbf{r}) = n_\infty - (n_\infty - n_T)R/r + (1 - R^3/r^3)\mathbf{r}\nabla n_\infty. \tag{4.4}$$

The asymmetric part of the exciton flux density on the drop surface, determined by the last term in (4.4), is equal to $3D\mathbf{R}\nabla n_\infty/R$. It is seen from it that the condensation flux preserves the spherical shape of the drop, and displaces the latter as a unit with a velocity

$$\mathbf{V}_c = 3D\nabla n_\infty/N; \tag{4.5}$$

the very same formula follows also from (4.2).

As to the hydrodynamic velocity, in the lowest order of the diffusion theory we have $\Pi_{ij} = p\delta_{ij}$, and since $p = Tn_T = \text{const}$, on the surface, the integral in (4.1) vanishes and the equation is satisfied at $\mathbf{V}_h = 0$. A nonzero $\mathbf{V}_h$ results only from various correction terms. For example, a difference between n_s^* and n_T [cf.(3.11)] leads to a pressure difference between opposite sides of the drop $\sim T\Lambda_g|\nabla n_\infty|$ and gives rise to $\mathbf{V}_h \sim -(v_T\tau_l/R)\mathbf{V}_c$ (v_T is the thermal velocity of the excitons in the gas). The contribution of the nonlinear terms mnu_iu_j in the momentum flux for the gas, as well as the contribution of the Stokes viscous force in the gas flow never exceed the foregoing estimates. Thus, the direction of the drop motion is determined by the parameter $v_T\tau_l/R$; if it is small, then $\mathbf{V} \approx \mathbf{V}_c$. We have already encountered this parameter above, namely in the problem of the drop instability it determined the ratio of the anisotropic gaskinetic and hydrodynamic contributions to the pressure at $R \sim R_{st}$ [cf. the third and first terms in the curly bracket of Eq. (3.26)].

We have assumed throughout a sticking coefficient $\gamma = 1$. In the opposite limiting case when $\gamma \to 0$, we obtain for the asymmetrical part of the concentration

$$n_{as}(\mathbf{r}) = (1 + R^3/2r^3)\mathbf{r}\nabla n_\infty, \tag{4.6}$$

and for the drift velocity[3)]

$$\mathbf{V}_h = -\frac{3}{2}\frac{T\tau_l}{mN}\nabla n_\infty. \tag{4.7}$$

It is seen from a comparison of (4.5) and (4.7) that in the two cases the velocities differ by the symbol parameter $V_c/V_h \sim \tau_g/\tau_l$. Owing to the Fermi degeneracy in the liquid, τ_l is relatively long,[14] so that $\tau_g/\tau_l \ll 1$. This raises the question whether a regime in which V_c predominates is possible at all at reasonable values of γ. The case $\gamma \neq 1$ does not admit of an analytic solution, for in this case the equation analogous to (3.6) acquires a term with a kernel that depends on $z + z'$. It is easy, however, to join together approximately the flow in the diffusion region and the wall current, in analogy with the procedure used in Ref. 7. We arrive then at the following conclusion. The condensation term should prevail in the velocity at $\gamma R/\Lambda_g \gg 1, \gamma \gtrsim (v_T\tau_l/R)^{1/2}$. Physical considerations connected with the structure of the drops create an impression that γ is close to unity. The decisive role is therefore played by the value of the parameter $v_T\tau_l/R$. If it is less than unity then V_c can prevail, and if it is larger than unity then the prevailing quantity is always

$$V_h \sim -\frac{v_T\tau_l}{\gamma R}\frac{D}{N}\nabla n_\infty, \quad \frac{\gamma R}{\Lambda_g} \gg 1. \tag{4.7'}$$

B. Drop interaction in the diffusion regime. Because of the inhomogeneous distribution of the exciton concentration around the drops, an interaction between them should exist—either attraction or repulsion. We start with the case $\gamma=1$. Then it follows from (4.4) and (4.5) that for drops of radius R and separated by a distance $\mathbf{r}$

$$\mathbf{V}_c=\frac{3DR}{N}(n_\infty-n_T)\frac{\mathbf{r}}{r^3}\,. \tag{4.8}$$

We see therefore that the sign of the velocity depends on the regime: the drops repel one another in the course of condensation $(n_\infty>n_T)$ and attract in the course of evaporation $(n_\infty<n_T)$. This is the qualitative difference between our mechanism and phonon wind,[8] where there is always repulsion. Yet the dependence on r is the same—the inverse square law.

At $\gamma\neq 1$, different regions of the parameter values are possible. If $\gamma R/\Lambda_g\ll 1$, then

$$\mathbf{V}_h\sim-\gamma v_T\frac{\tau_l}{\tau_g}\frac{n_\infty-n_T}{N}\frac{R^2\mathbf{r}}{r^3}. \tag{4.9}$$

If $\gamma R/\Lambda_g\gg 1$, then

$$\begin{gathered}\mathbf{V}_c\sim v_T\frac{n_\infty-n_T}{N}\frac{\Lambda_g R\mathbf{r}}{r^3},\\ \mathbf{V}_h\sim-\frac{v_T}{\gamma}\frac{n_\infty-n_T}{N}v_T\tau_l\frac{\Lambda_g\mathbf{r}}{r^3};\end{gathered} \tag{4.10}$$

the relation between these quantities was discussed at the end of the preceding subsection.

C. *Drop motion in the Knudsen regime*. Because of the small drop dimensions, the perturbations of the exciton concentration in the drop vicinity, over scales $\sim\Lambda_g$, are small relative to parameter $(R/\Lambda_g)^2\ll 1$. We can therefore disregard the perturbation produced by the drop itself in the distribution function of the excitons incident on the drop from the gas phase. In the reference frame connected with the drop, this distribution function is

$$\varphi(\mathbf{v})=\varphi_0(v)\{1-\mathbf{v}(\tau_g\nabla n_\infty+m\mathbf{V}/T)\}; \tag{4.11}$$

φ_0 is the Maxwellian function. Here $\mathbf{V}\approx\mathbf{V}_h$, since V_c is small $(V_c\ll V_h)$; actually, for an immobile drop $\mathbf{V}_c=(D/2N)\nabla n_\infty$, and allowance for the drift of the drop with velocity V_h does not change the order of magnitude of V_c. The total momentum transfer from the gas to the drop, which can be easily calculated using (4.11), is

$$-\oint\hat{\Pi}\,d\mathbf{S}=-{}^1/_3\pi R^2m(\langle\tau_g v^3\rangle\nabla n_\infty+m\langle v^3\rangle n_\infty\mathbf{V}/T+3\langle v\rangle n_T\mathbf{V}). \tag{4.12}$$

The last term describes here the momentum loss when the moving drop evaporates; when substituted in (4.1), this term cancels out the term $\mathbf{V}_h dM/dt$, on the left-hand side.

Two motion conditions are possible here, depending on which of the frictions prevails, with the lattice or with the gas [the second term in (4.12)]. If friction with the lattice prevails, namely

$$\frac{n_\infty}{N}\frac{v_T\tau_l}{R}\sim\frac{n_\infty}{N}\frac{\tau_l}{\tau_g}\frac{\Lambda_g}{R}\ll 1, \tag{4.13}$$

then the stationary drop velocity, obtained by equating the right-hand side of (4.1) to zero, is

$$\mathbf{V}=-\frac{\tau_l\langle\tau_g v^3\rangle}{4RN}\nabla n_\infty. \tag{4.14}$$

In this case $V_c/V_h\sim R/v_T\tau_l<R/\Lambda_g\ll 1$, since $\tau_l>\tau_g$.

If friction with the gas predominates, i.e., a criterion opposite to (4.13) is satisfied, then

$$\mathbf{V}=-\frac{T}{m}\frac{\langle\tau_g v^3\rangle}{\langle v^3\rangle}\nabla\ln n_\infty; \tag{4.15}$$

this velocity is of the same order as the exciton-gas velocity produced by the concentration gradient[4)] ∇n_∞. In this case $V_c/V_h\sim n_\infty/N\ll 1$.

The ratio of the velocities (4.14) and (4.15) is determined by the parameter contained in (4.13).

D. *Drop interaction in the Knudsen regime*. In the Knudsen case the interaction between the drops can be substantial only at short distances, $r\lesssim\Lambda_g$, and reduces to partial screening of the gaskinetic flow. We present the estimates, but do not write out the numerical coefficients.

If $n_\infty>n_T$ (condensation regime), the drops attract, with

$$\mathbf{V}\sim-\frac{n_\infty}{N}\frac{\tau_l v_T^2R}{r^2}\frac{\mathbf{r}}{r},\quad \mathbf{V}\sim-v_T\left(\frac{R}{r}\right)^2\frac{\mathbf{r}}{r} \tag{4.16}$$

for the respective cases of friction with the lattice and with the exciton gas (it is assumed that $n_\infty\gg n_T$).

If $n_\infty<n_T$ (evaporation regime), the drops repel, and for the same two cases we have

$$\mathbf{V}\sim\frac{n_T}{N}\frac{\tau_l v_T^2R}{r^2}\frac{\mathbf{r}}{r},\quad \mathbf{V}\sim v_T\frac{n_T}{n_{eff}}\frac{R^2}{r^2}\frac{\mathbf{r}}{r}, \tag{4.17}$$

where $n_{eff}=\text{mas}\{n_\infty, n_T(R/\Lambda_g)^2\}$; we assume that $n_T\gg n_\infty$. Of course, at low temperatures, when n_T is exponentially small, the decisive contribution should be made by evaporation of the nonthermalized particles produced via Auger recombination in the drop.

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[1)] Confining ourselves to relatively large drops greatly exceeding the critical nucleus, we neglect the capillary correction[5] to n_T.

[2)] Recombination in a moving liquid can be also accompanied by a momentum transfer that depends on the recombination mechanism. But the corrections for this momentum-loss mechanism are small compared with friction in a ratio $\tau_l/\tau_0\ll 1$.

[3)] It is easily seen that neither (4.5) nor (4.7) agrees with the result given in Ref. 4 [see Eq. (7.10) there], but our Eq. (4.7) differs only by the numerical factor 3.

[4)] Both (4.14) and (4.15) differ from the equation proposed earlier for the drop velocity in the Knudsen regime [see Eq. (12) of Ref. 12]. In particular, Eq. (4.15) differs from the cited equation by the factor Λ_g/R.

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Change in the local geometry of the Fermi surface and the anomaly of the electronic sound absorption coefficient

M. I. Kaganov and T. L. Lobanova

M. V. Lomonosov Moscow State University

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A break in a bridge is accompanied by the appearance or disappearance of parabolic-point lines on the Fermi surface. As a result, the change in the connectivity of the Fermi surface leads to an anomaly in the angular dependence of the sound absorption coefficient due to electrons, Γ_e. The closeness of the parabolic-point line to the conical line increases the anomaly. The concept of a phase transition of 2 1/2 order is generalized to include changes in the local geometry of a Fermi surface not accompanied by a change in its connectivity. It is shown that the formation of a "waist" or a "crater" is accompanied by the appearance of angular singularities in Γ_e.

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1. INTRODUCTION

After the prediction of a phase transition of $2\frac{1}{2}$ order[1] due to a change in the connectivity of the Fermi surface, the study of the electron energy spectrum of metals and degenerate semiconductors by means of external action on their Fermi surface has become widespread. Change in the connectivity of the Fermi surface, as is known, takes place because of the rearrangement of the structure of the surface1)

$$\varepsilon(\mathbf{p})=\varepsilon_F \tag{1}$$

close to isolated points of p space ($\mathbf{p}=\mathbf{p}_k$), at which either a new cavity of the Fermi surface is created or a bridge is broken.[1,2] The significant effect of such a small change in the spectrum on the thermodynamic characteristics is due to the fact that, because of degeneracy, the latter are determined not by all the electrons, but only by those located on the Fermi surface, and the role of slow electrons is anomalously large, since the density of states at $\varepsilon=\varepsilon_F$ is

$$\nu(\varepsilon)=\frac{2}{(2\pi\hbar)^3}\oint\limits_{(\varepsilon_F)}\frac{dS}{v},\qquad \mathbf{v}=\nabla_p\varepsilon. \tag{2}$$

We recall that $\mathbf{v}(\mathbf{p}_k)=0$.

The kinetic properties of a metal are especially sensitive to the structure of the Fermi surface, and, in certain cases, the change in the connectivity of the Fermi surface can lead to radical changes. For example, if, as a result of the breaking of a bridge, the Fermi surface changes from open to closed, then the conductivity in a strong magnetic field along the direction of the openness can increase by factors of tens or even hundreds (see Ref. 2, Sec. 28).

The sound absorption coefficient Γ is especially sensitive to a change in the local structure of the Fermi surface.[3] The fact is that, first, the role of slow electrons in the interaction with the sound is even more important than its role in the thermodynamic characteristics, since the formula that determines the sound absorption coefficient Γ in the form of an integral over the Fermi surface contains v^2 in the denominator [cf. with formula (2)], and second, electrons on the "belt" participate in the absorption of shortwave sound ($kl\gg 1, k=\omega/s$ is the wave vector of the sound wave of frequency ω, s is velocity, l is the mean free path of the electrons)2)

$$\mathbf{vk}=0,\qquad \varepsilon(\mathbf{p})=\varepsilon_F, \tag{3}$$

the structure of which can change upon change in the

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