# Characteristics of the gas-liquid transition in a system of particles with finite lifetime

S. G. Tikhodeev
*P. N. Lebedev Physics Institute, Academy of Sciences of the USSR*

(Submitted 20 January 1983)
Pis'ma Zh. Eksp. Teor. Fiz. **37**, No. 5, 216–218 (5 March 1983)

The temperature range $0 < T < T_1$, in which the gas-liquid transition in a system of particles with finite lifetime occurs without the hysteresis characteristic of first-order phase transitions, is estimated.

PACS numbers: 64.70.Fx, 51.90. + r

The electron-hole (EH) liquid, which is formed in a number of semiconductors at low temperatures and high excitation levels (see, for example, Ref. 1), differs from the usual liquids, aside from other characteristics, by the finiteness of the lifetime of its constituent particles (due to recombination of EH pairs). This leads to the fact that at the lowest temperatures the exciton-gas–EH-liquid transition occurs without the hysteresis characteristic of first-order phase transitions and metastable transformations of states.[2–6]

The reason for the appearance of metastable states and hysteresis with the usual gas-liquid transition is well known (see, for example, Ref. 7). In order for a viable nucleus of the liquid phase to form, the energy barrier stemming from the positive-surface contribution to the thermodynamic potential for small nuclei must be overcome. It was shown in Refs. 8, 2, and 4 that allowance for the finiteness of the particle lifetimes leads to the appearance of an additional term in the effective thermodynamic potential. This term includes the new (aside from evaporation) possibility for particles to leave the nucleus: They can cease to exist. At the lowest temperatures, this annihilation dominates over evaporation. Numerical analysis of the effective potential shows[2,4] that in some temperature range $0 < T < T_1$ the energy barrier disappears, and the possibility for the appearance of metastable states and hysteresis disappears with it. The disappearance of hysteresis with the formation of EH liquids in Ge and Si has been observed experimentally.[5,6]

In this paper, we present a simple proof of the fact that the gas-liquid transition in any system of particles with finite lifetime must occur without hysteresis at sufficiently low temperatures, and we also obtain an equation for the $T_1$ temperature range of such behavior.

The equation for the growth rate of the nucleus of the liquid phase with radius $R$, including the finiteness of the particle lifetime $\tau_0$, has the form[9]

$$\frac{dR}{dt} = \gamma v_T \frac{n - n(R)}{n_0}, \qquad n(R) = n_T(R) + \frac{n_0 R}{3\gamma v_T \tau_0}. \tag{1}$$

Here $n$ and $n_0$ are the particle densities in the gas and liquid; $v_T \simeq \sqrt{kT/2\pi M}$ is the thermal velocity of particles in the gas and $M$ is their effective mass; $0 < \gamma < 1$ is the

  

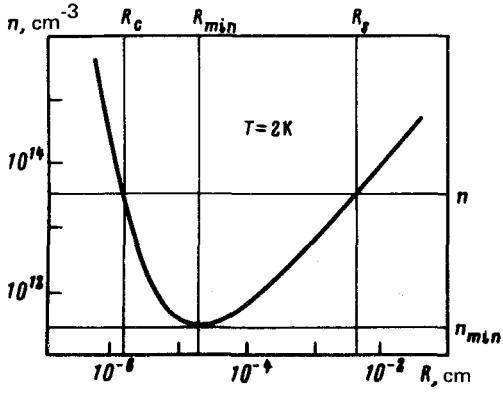


FIG. 1. The function $n(R)$ for an EH liquid in Ge at $T = 2$ K.

attachment coefficient for particles to stick to the surface of the drop;

$$n_T(R) = \nu\left(\frac{MkT}{2\pi\hbar^2}\right)^{3/2} \exp\left(-\frac{\varphi}{kT} + \frac{2\sigma}{n_0 RkT}\right) \tag{2}$$

is the gas density in thermodynamic equilibrium at the surface of the drop with radius $R$; $\nu$ is the degeneracy of the ground state of the gas; $\varphi$ and $\sigma$ are the binding energy and surface tension of the liquid.

The condition $n = n(R)$ for fixed gas concentration is the equation for the stationary size of the drop. Figure 1 shows a typical function $n(R)$ (for an EH liquid in Ge, when[9] $\varphi = 16, \gamma = 1, M = 4\times 10^{-28}$g, $\varphi = 2.1$ meV, $\sigma = 2\times 10^{-4}$erg/cm$^2$). For $n < n_{\min}$, stationary drops do not exist and for $n > n_{\min}$, the equation $n = n(R)$ has two solutions: $R_c$ and $R_s$. $R_c$ lies to the left of the unstable branch $n(R)$; this is the radius of the unstable critical nucleus. $R_s$ lies on the right stable branch $n(R)$; this is the stable stationary size of the drop for given gas density $n$. Using an approach analogous to that used in the usual theory of nucleation,[7] it can be shown that the effective thermodynamic potential has an extremum at these points: the maximum at $R_c$ (this corresponds to the instability of this point) and a minimum at $R_s$.[8,2]

The value $R_{\min}$, which separates the left unstable branch of the function $n(R)$ from the right stable branch, is the minimum possible radius at a given temperature of the drop. The equation for $R_{\min}$ has the form[9]

$$R_{min}^2 = 6\gamma \frac{\sigma v_T \tau_0}{n_0^2 kT} n_T(R_{min}). \tag{3}$$

The function $R_{\min}(T)$, obtained by solving Eq. (3) numerically for an EH liquid in Ge, is shown in Fig. 2 (curve 1). $R_{\min}$ decreases with decreasing temperature. Curves 2 and 3 were calculated for the same $\nu, \gamma M, \varphi, \sigma$, but for different values of $\tau_0$. At a fixed temperature, $R_{\min}$ decreases with decreasing lifetime. All curves at the lowest temperatures converge to a single point. This is easy to verify by passing to the limit $T \to 0$ in (3). Thus the solution of (3)

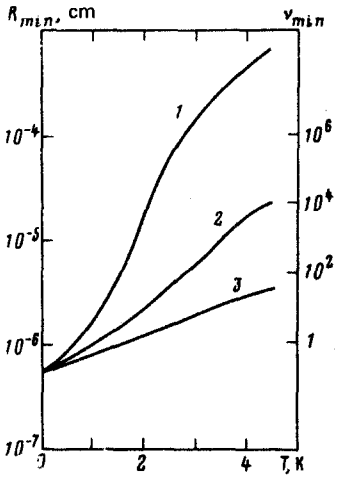


FIG. 2. Minimum drop size $R_{\min}(T)$. The values of the parameters $\nu,\gamma,M,\varphi,\sigma$ are presented in the text; $\tau_0 = 40$ $\mu$s (curve 1); $\tau_0 = 40$ ns (curve 2); $\tau_0 = 0.4$ ns (curve 3).

$$R_0 \equiv R_{min}(0) = \frac{2\sigma}{n_0\varphi} \tag{4}$$

depends only on the equilibrium parameters of the liquid.

Let us use Langmuir's equation[7] $\sigma \cong (1/5)\varphi n_0^{2/3}$ to estimate the number of particles $\nu_0$ in a drop of radius $R_0$: $\nu_0 = (4/3)\pi R_0^3 n_0 \cong (4\pi/3)(2/5)^3 \sim 0.3$. Thus $\nu_0$ is a number identical in order of magnitude for any liquid with finite lifetime. In addition, $\nu_0 \lesssim 1$. For this reason, the entire unstable branch of the function $n(R)$ in any system of particles with finite lifetime for $T \to 0$ falls into a region of such small values of $R$ that these values correspond to nuclei with a physically meaningless number of particles amounting to less than unity. Since the thermodynamic potential is maximum in $R_c$, this indicates that the potential barrier disappears with alls the ensuing consequences.

It is easy to estimate the dimensions of the temperature range $0 < T < T_1$, in which there is no potential barier. As the temperature is increased the number of particles in the smallest nucleus increases, and at a temperature $T_1$ it becomes quite large. Let us assume, for simplicity, that this occurs for $\nu_{\min}(T_1) \sim 10$ (the estimate of $T_1$, as we shall see, is insensitive to the choice of this value). Then, using Langmuir's equation, the estimate $\varphi \sim \hbar^2 n_0^{2/3}/M$, and ignoring the numerical factors $\sim 1$, we obtain the following equation for $x = \varphi/kT_1$:

$$e^x/x = \gamma\frac{\tau_0\varphi}{\hbar} \tag{5}$$

The right side of (5) is much greater than unity ($\sim 10^8$ for Ge and $\sim 10^2$–$10^3$ for rectilinear band semiconductors with $\tau_0 \sim 1$ ns). For this reason, to logarithmic accuracy, we obtain

$$T_1 \cong \frac{\varphi}{k \ln\left(\gamma \frac{\tau_0 \varphi}{\hbar}\right)} \tag{6}$$

For an EH liquid in Ge, Eq. (6) gives $T_1 \sim 1.3$ K, a value that coincides with the value known from experiment[5] and from numerical calculations.[2] For Si, setting $\gamma = 1$, $\varphi = 8.2$ meV, $\tau_0 = 150$ ns,[1] we obtain $T_1 \sim 6$ K. Agreement with the value[6] $T_1 \sim 10$ K measured experimentally is obtained if we set $\gamma \sim 10^{-2}$. This corresponds, in order of magnitude, to the values obtained in Refs. 6 and 10 by adjusting the condensation theory to experiment.

I am grateful to L. V. Keldysh for discussing the problems considered in this paper.

[1]T. M. Rice, J. C. Hansel, T. G. Phillips, and G. Thomas, Electron-Hole Liquid in Semiconductors [Russian translation], Mir, Moscow, 1980.
[2]R. M. Westervelt, Phys. Status Solidi B **74**, 727 (1976); *ibid.* **76**, 31 (1976).
[3]M. Combescot and R. Combescot, Phys. Lett. A **56**, 228 (1976).
[4]S. Koch and H. Haug, Phys. Lett. A **74**, 250 (1979).
[5]R. M. Westervelt, J. L. Staehli, and E. E. Haller, Phys. Status Solidi B **90**, 557 (1978).
[6]P. Voisin, B. Etienne, and M. Voos, Phys. Rev. Lett. **42**, 526 (1979).
[7]Ya. I. Frenkel, Kineticheskaya teoriya zhidkosteĭ (Kinetic Theory of Liquids), Nauka, Leningrad, 1975.
[8]R. N. Silver, Phys. Rev. B **11**, 1569 (1975); **12**, 5689 (1975).
[9]V. S. Bagaev, N. V. Zamkovets, L. V. Keldysh, N. N. Sibel'din, and V. A. Tsvetkov, Zh. Eksp. Teor. Fiz. **70**, 1501 (1976) [Sov. Phys. JETP **43**, 783 (1976)].
[10]R. B. Hammond and R. N. Silver, Phys. Rev. Lett. **42**, 523 (1979).

Translated by M. E. Alferieff
Edited by S. J. Amoretty