

Gas-liquid phase transition induced by light

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The changes in the properties characterizing the nonideal nature of a gas due to the influence of resonant radiation are investigated. It is shown that a change of the critical parameters and of the range of coexistence for a gas-liquid system may result from the influence of light. The conditions under which radiation leads to a condensation of unsaturated gas are clarified. The properties of the condensed state are investigated.

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The effect of external influences on the behavior of various systems near phase transition points has been widely investigated in recent years. The possibility of an effect due to resonant radiation on the critical parameters of a gas-liquid system was discussed in an article by Gudzenko and the authors.^[1] The thermodynamics of such systems are investigated in detail in the present article, and the conditions under which a noticeable change in the parameters characterizing the nonideal nature of the gas takes place and which lead in particular to the photocondensation effect, are clarified.

The change in the parameters of the equation of state for the gas-liquid system due to the effects of resonant radiation, which transfers particles (atoms or molecules) from the ground level to some excited level, is due to the fact that the attractive potential between excited particles and also between excited and nonexcited particles is, in a number of cases, markedly different from the interaction potential of particles located in their ground states. Rearrangement of the intermolecular interaction may lead to dependences of the shape of the coexistence curve and the values of the critical parameters on the intensity of the external light field.

Let us consider a gas consisting of N particles with a pair interaction potential between them. For these particles let us distinguish the ground electronic level and some excited level (the energy difference ε between the levels will later correspond to the frequency of the incoming field). Let the interaction of particles which are in their ground states be described by the potential $u_{ik} = u(|\mathbf{r}_i - \mathbf{r}_k|)$; let the interaction of particles in excited states be described by the potential $g_{ik} = g(|\mathbf{r}_i - \mathbf{r}_k|)$; and let the interaction of particles in different states be described by the potential $q_{ik} = q(|\mathbf{r}_i - \mathbf{r}_k|)$. For the systems under consideration the level broadening due to the nonideal nature of the gas is small and cannot lead to overlapping of the levels, that is,

$$|u_{ik}|, |g_{ik}|, |q_{ik}| \ll \varepsilon. \quad (1)$$

in the region of attraction. We shall also assume that $\beta^{-1} = kT \ll \varepsilon$ (T denotes the temperature of the system).

Let the gas be under the influence of a quasicontinuous laser pulse. As a consequence of inequalities (1), the average value s of the difference in populations between the upper (N_2) and lower (N_1) levels, calculated per particle ($s = (N_2 - N_1)/(N_2 + N_1)$), is determined solely by the

effect of the external field on the system. Therefore, averaging over the states of internal energy of the particles and neglecting correlations of the internal states of the particles due to the interaction with the field, we may write down the Hamiltonian of the system in the form (we do not write down terms corresponding to internal degrees of freedom not directly connected to the interaction with the field, such terms being unimportant for thermodynamic calculations)

$$H = \frac{1}{2} \varepsilon s (N_1 + N_2) + \sum_i \frac{\mathbf{p}_i^2}{2m} + \frac{1}{8} \sum_{i \neq k} [g_{ik}(1+s)^2 + u_{ik}(1-s)^2 + 2q_{ik}(1-s^2)],$$

where m and \mathbf{p}_i denote the mass and momentum of the i -th particle. The value of s should be sought from the self-consistent system of Maxwell's equations in the medium and the equations of motion for the density matrix (see, for example, [2]). For each specific system s is determined by the totality of kinetic processes which transfer particles to various excited levels and to the continuous spectrum. Without limiting the applicability of the investigation, for the sake of simplicity we shall neglect the dependence of the degree of excitation s on the spatial coordinates, assuming that a thin layer of gas is irradiated uniformly at each point.

The system under consideration exists in a state of partial equilibrium, since the distribution of particles in the levels is not an equilibrium distribution, but is specified independently with the aid of the external field's influence. The possibility of a thermodynamic treatment follows from the fact that the time to establish thermal equilibrium with respect to the translational motion of the particles is many orders of magnitude smaller than the time required to transfer excitation energy to the energy of their translational motion. The state of the system will be stationary in time if the incoming intensity varies weakly throughout the duration of the illumination and if the temperature of the gas doesn't change significantly as a consequence of the illumination (the latter condition may impose a restriction on the intensity and duration of the incoming radiation). Upon fulfillment of these conditions, the possibilities for observation of the effect are optimal.

Let us calculate the free energy of the system, F , by the standard method in the molecular field approximation, having chosen a specific form of the interaction potentials in the following way: $u(r) = q(r) = g(r) = 0$ for $r \geq r_1$; $u(r) = -u_0$, $g(r) = -g_0$, $q(r) = -q_0$ for $r_0 < r < r_1$;

$u(r)=g(r)=q(r)=+\infty$ for $r \leq r_0$. Such a specific model then allows us to utilize for estimates the data of Gurevich *et al.*^[3] with regard to the depths of the attractive potentials of atoms found in various states. These data indicate, at least for the examples considered below, that one can introduce identical values of the attractive and repulsive radii for $u(r)$, $g(r)$, and $q(r)$. Here it should be noted that nothing in the way of more accurate data concerning the r dependence of the interaction potentials is available; at the same time it is well known that the basic properties characterizing the behavior of a system near critical points weakly depend on the detailed shape of the individual-particle interaction potentials. For F we have

$$-\beta F = \frac{3\bar{N}}{2} \ln \frac{mkT}{2\pi\hbar^2} + \bar{N} - \frac{1}{2} \beta \epsilon s \bar{N} + \frac{1}{2} \beta \bar{N} \bar{v}_1 (\varphi + 2sf + s^2\psi) + \bar{N} \ln \frac{1-\bar{v}_0}{\bar{v}} \quad (2)$$

where

$$f = \frac{1}{2} (g_0 - u_0), \quad \psi = \frac{1}{2} (g_0 + u_0 - 2q_0), \quad \varphi = \frac{1}{2} (g_0 + u_0 + 2q_0), \quad \bar{N} = N_1 + N_2,$$

$\bar{v} = \bar{N}/V$ denotes the density of the active particles, $v_0 = (16/3)\pi r_0^3$ and $v_1 = (4/3)\pi(r_1^3 - r_0^3)$ denote the effective volumes of the spheres of repulsion and attraction, respectively. According to Eq. (2), we obtain the following expression for the pressure P

$$\beta P = -\frac{1}{2} \beta \bar{v}_1 \bar{v}^2 (\varphi + 2sf + \psi s^2) + \bar{v} / (1 - \bar{v} v_0). \quad (3)$$

For arbitrary intensity of illumination, the equation of state has the form of van der Waals' equation. According to Eq. (3), the ratio of the critical parameters of the system in the presence of the radiation field to their values in the absence of the field ($s = -1$) is given by (see Figs. 1a, b)

$$\alpha = \frac{T_c}{T_c^{vdW}} = \frac{P_c}{P_c^{vdW}} = \frac{1}{4} \left[1 + \frac{g_0 + 2q_0}{u_0} - 2s \left(1 - \frac{g_0}{u_0} \right) + s^2 \left(1 + \frac{g_0 - 2q_0}{u_0} \right) \right]$$

(here and in what follows we assume $\bar{v} \approx N/V$, which is valid for the systems investigated below). Near T_c the behavior of the coexistence curve is described by the formula

$$|V - V_{cr}| = \left[\frac{36}{17} \frac{V_c^2}{T_c} (T_c - T) \right]^{1/2}. \quad (4)$$

In the case of strong (saturating) fields, one can easily show with the aid of Maxwell's equations that $s \approx 0$, and

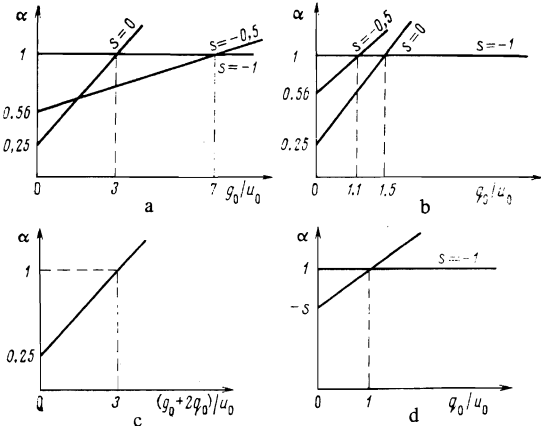


FIG. 1. The dependence of the ratio α of the critical parameters on the interaction potentials; a) $q_0 = 0$, b) $g_0 = 0$; c) the case of saturating fields; d) the case of weak fields.

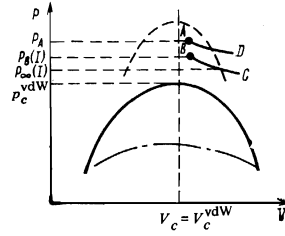


FIG. 2. Variation of the region of coexistence under the influence of radiation.

then $\alpha = \varphi/u_0$ (Fig. 1c). In the case of weak fields, when $s + 1 \ll 1$, the effect is determined by just the interaction of excited particles with unexcited particles. In fact, in this case (Fig. 1d) we have

$$\alpha = 1 + (q_0/u_0 - 1)(s + 1). \quad (5)$$

For $q_0 > u_0$ (for $g_0 + 2q_0 > 3u_0$ in the case of saturating fields) a rise ($T_c > T_c^{vdW}$) takes place on the $P-V$ diagram and, according to Eq. (4), there is a narrowing of the region of coexistence (the dotted line in Fig. 2), that is, a phase transition may occur for certain values of the thermodynamic parameters, values at which the usual condensation does not occur (the solid line in Fig. 2 is the coexistence curve in the absence of radiation). For $q_0 < u_0$ ($g_0 + 2q_0 < 3u_0$ in the case of saturating fields), a reduction of the region of coexistence takes place along with its simultaneous broadening (the dash-dot line in Fig. 2), i. e., the opposite effect is realized, when the gas-liquid transition is suppressed by radiation. The case of weak fields is of the greatest practical interest. In this connection it is much easier to satisfy the conditions for optimal observation of the effect, under which the system is in stationary equilibrium during the total duration of the illumination.

Let us cite the explicit dependence of the degree of excitation s on the radiation intensity I in the absence of additional relaxation channels, except for those which directly connect the ground level to the excited levels, i. e., neglecting transitions to other excited states and to the continuous spectrum. Such a two-level model of relaxation is valid for excitation of low-lying atomic or molecular levels. In this case, from the equations for polarization of the medium and from Maxwell's equations with inequalities (1) taken into consideration, one can easily obtain the following result for a quasistatic resonant pulse^[2]

$$s = -(1 + I/I_s)^{-1},$$

where $I_s = [2\sigma(\omega)\tau_1]^{-1}$, $\sigma(\omega) = 4\pi\tau_2^{-1}\epsilon d^2 / \{\hbar^2 c [(\omega - \epsilon/\hbar)^2 + \tau_2^{-2}]\}$ denotes the photoabsorption cross section, τ_1 and τ_2 denote the longitudinal and transverse relaxation times, d is the dipole moment of the transition, and $(\omega - \epsilon/\hbar)$ denotes the detuning from resonance. From here it follows that in weak fields according to Eq. (5) we will have

$$\alpha = 1 + (q_0/u_0 - 1)I/I_s.$$

The corresponding dependences are shown in Fig. 3.

Now let us show that a change in the properties of the condensed state takes place under the influence of the radiation. Let us find, for example, the dependence of the surface tension coefficient η of the condensate on

the intensity. In the general case the dependence of η on T is given by the expression^[4]

$$\eta(T) = \frac{kT_c}{v_c^{1/3}} G\left(\frac{T}{T_c}\right),$$

where $G(T/T_c)$ is a universal function, and v_c is the specific volume at the critical point. The form of the function G is established from phenomenological considerations, where the most satisfactory expression is given by the Katayama-Guggenheim formula: $G(T) \sim (1 - T/T_c)^{11/9}$ for $T < T_c$. Since $T_c = T_c(I)$ according to Eq. (5), then $\eta = \eta(T, I)$. In particular, for $I/I_s \ll 1$

$$\eta(T, I) = \text{const} \frac{T_c^{\text{vdW}}}{v_c^{2/3}} \left[1 + \left(\frac{q_0}{u_0} - 1 \right) (s+1) \right] \left(1 - \frac{T}{T_c} \right)^{11/9}. \quad (6)$$

For $q_0 > u_0$ the liquid phase, arising under the influence of the radiation, possesses a larger coefficient of surface tension than the usual liquid state of this substance. Formula (6) also shows that upon a change of the intensity the drops of liquid phase which appear should change their dimensions, increasing their radius with increasing intensity.

Let us consider the transition into the liquid state. Prior to illumination let the system be found in the gaseous phase at temperature T , pressure P_A , and volume V (point A in Fig. 2). Upon illumination the system is found in a new state (point B), which corresponds to the same temperature but a different pressure $P_B(I)$:

$$P_B(I) - P_A = -1/2 \rho^2 v_1 [\varphi + 2sf + s^2 \psi - u_0]. \quad (7)$$

A displacement of the isotherms takes place under the influence of light: on Fig. 2, corresponding to the case $q_0 > u_0$, $s+1 \ll 1$, the isotherm AD changes into BC . We shall now assume that point A is located near (P_c^{vdW}, V_c) and point B falls inside the new region of coexistence (the dotted line in Fig. 2), the raising of which is calculated according to Eqs. (4) and (5). Then point B corresponds to finding the system in a state of supersaturated vapor, which corresponds to a certain pressure $P_\infty(I) < P_B(I)$ above the planar interface:

$$P_\infty(I) = P_c \left[1 - 48 \sqrt{\frac{36}{17}} \left(1 - \frac{T}{T_c} \right)^{11/9} \right]. \quad (8)$$

The rate of condensation is completely determined by the degree of supersaturation $\zeta = P_B(I)/P_\infty(I)$ and by the size of the critical nucleation center $r^* = 2\eta[P_B(I) - P_\infty(I)]^{-1}$. We note that, according to Eqs. (6)–(8), for a given value of ζ the rate of photocondensation is decreased in comparison with the case of ordinary condensation. This difference, however, is unimportant for heterogeneous nucleation.

Above, in the thermodynamic calculations, the bound states—excited molecular complexes—were not taken into consideration. Such structures are unstable with respect to the emission of a photon accompanied by a

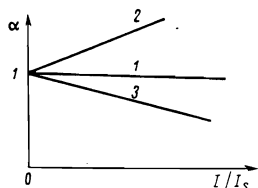


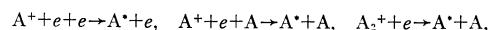
FIG. 3. Dependence of α on the intensity of the radiation (two-level model of relaxation): 1— $q_0 = u_0$, 2— $q_0 > u_0$, 3— $q_0 < u_0$.

transition to a lower (disintegrating) state, and with respect to elementary processes of a different type: predissociation, the formation of molecular ions at large (electronic) energies of excitation, etc. If their rate of decomposition is sufficiently large, the bound states will turn out to have no significant influence on the thermodynamics. In the opposite case, an accumulation of excited molecular complexes will take place in a gas. The condensation of such a gas will be determined by their van der Waals' interaction among themselves and with unexcited particles. A photocondensation process of this type is also of interest; however, in this case the effect is apparently weaker since the attractive potential of the complexes being formed is clearly smaller than the attractive energy of the initial particles, whose magnitude just guarantees the formation of bound states.

As was mentioned in^[1], the inert gases may be of great interest for observation of the effect. For xenon, for example, $q_0/u_0 \sim 10$ to 20, for argon the same ratio ~ 130 , and for helium ~ 800 .^[3] Now let us show for the example of the inert gases, that for a sufficiently weak intensity of the illumination, taking account of any additional channels of relaxation from excited states of the atoms, in addition to a direct transition to the ground state, has essentially no effect on the parameter s , which determines the magnitude of the effect. For specific estimates we shall direct our attention to the case of argon and helium,^[1] existing under conditions close to critical (density $\sim 10^{21}$ cm⁻³, the temperature for helium is ~ 5 K, for argon it is ~ 150 K). Among the processes taking place with the participation of excited atoms, it is necessary to isolate the formation and decay of bound states AA^* . In this connection, since the terms of the inert gas quasimolecule AA^* intersect the terms of the molecular ion A_2^+ , one can assume^[7] that the formation of a bound state is actually very similar to the appearance of an ion A_2^+ . In the case of inert gases, the collisions of excited atoms represent another important kinetic process



and lead to the ionization of one of the atoms and to formation of fast electrons having an energy equal to the difference between the doubled excitation energy and the ionization potential. Confining our attention in the equations for the density matrix of the medium to processes involving the formation of molecular and atomic ions, and also to the inverse recombination processes



we obtain

$$\begin{aligned} \partial N_+ / \partial t + N_+ / \tau_1 &= -\sigma I (N_+ - N_-) - 2\sigma_m v_T N_+^2 - \sigma_{mi} v_T N_+ N_- + \\ &+ K_{mr} N_2 N_+ N_e + K_r^{(1)} N_+ N_e^2 + K_r^{(2)} N_+ N_- N_e, \\ \partial N_- / \partial t - N_- / \tau_1 &= \sigma I (N_+ - N_-) + \sigma_m v_T N_-^2 - \\ &- \sigma_{mi} v_T N_- N_+ - K_{mr} N_2 N_- N_e, \\ \partial N_+ / \partial t &= \sigma_m v_T N_+^2 - K_r^{(1)} N_+ N_e^2 - K_r^{(2)} N_+ N_- N_e, \\ \partial N_2 / \partial t &= \sigma_{mi} v_T N_+ N_- - K_{mr} N_2 N_- N_e, \end{aligned} \quad (10)$$

where $N_+ = N_1/V$, $N_* = N_2/V$; N_+ , N_2 , and N_e denote the densities of atomic ions, molecular ions, and electrons; v_T denotes the mean thermal velocity of the heavy particles; σ_{mi} denotes the cross for the formation of a molecular ion; σ_i denotes the cross section for the process (9); $K_r^{(1)}$, $K_r^{(2)}$, and K_{mr} are the rate constants for

the processes of triple shock recombination, triple recombination in the presence of an atom, and dissociative recombination. Recognizing that, in the presence of quasicontinuous illumination, the system turns out to be under the conditions of "steady-state flow" ($\partial/\partial t=0$), from Eqs. (10) we obtain

$$(\gamma+1)n_+ + \nu n_-^2 - \gamma n_- = 0, \quad \nu n_-^2 - \beta_1 n_+ (n_- + n_{2+})^2 - \beta_2 n_+ n_- (n_- + n_{2+}) = 0, \\ \delta n_+ n_- - \chi n_{2+} (n_+ + n_{2+}) = 0, \quad n_- + n_+ + n_{2+} + 2n_{2+} = 1,$$

where

$$n_- = \frac{N_-}{\bar{\rho}}, \quad n_+ = \frac{N_+}{\bar{\rho}}, \quad n_{2+} = \frac{N_{2+}}{\bar{\rho}}, \quad n_{2+} = \frac{N_{2+}}{\bar{\rho}} \quad (\bar{\rho} = N_- + N_+ + N_{2+} + 2N_{2+}) \\ \gamma = \sigma I \tau_i = I/2I_s, \quad \nu = \sigma_i \nu_T \tau_i \bar{\rho}, \quad \delta = \sigma_{mi} \nu_T \tau_i \bar{\rho}, \quad \chi = K_{mr} \tau_i \bar{\rho}, \\ \beta_1 = K_r^{(1)} \tau_i \bar{\rho}^2, \quad \beta_2 = K_r^{(2)} \tau_i \bar{\rho}^2.$$

Now let $I \ll I_s$, i. e., $\gamma \ll 1$. Then

$$n_- \approx \gamma, \quad n_+ \approx 1 - \gamma^{1/2} \left(\frac{4\delta}{\chi} \right)^{1/2} - \gamma, \quad n_{2+} \approx \gamma^{1/2} \left(\frac{\delta}{\chi} \right)^{1/2}, \quad n_{2+} \approx \gamma^{1/2} \frac{\nu}{\beta_2} \left(\frac{\chi}{\delta} \right)^{1/2}, \quad (11)$$

whence it follows that

$$s = (n_+ - n_-) / (n_+ + n_-) \approx -1 + 2\gamma. \quad (12)$$

Formulas (11) and (12) are valid for $\gamma \ll 1$ such that

$$4\nu\gamma \ll 1, \quad 4\delta\gamma/\chi \ll 1, \quad \nu\chi/\delta\beta_2 \ll 1. \quad (13)$$

According to Eqs. (5) and (12) the change of the critical temperature is given by

$$\Delta T = T_c - T_c^{vdW} = 2T_c^{vdW}(q_0/u_0 - 1)\gamma. \quad (14)$$

We note that, for $I \ll I_s$ expression (12) agrees with the above obtained expression for the two-level model of relaxation, that is, for $I \ll I_s$ the value of s is slightly sensitive to the presence of additional relaxation channels, including the formation of bound states (molecular ions). Then by using the data^[7] concerning the cross sections for elementary events, it is easy to obtain the result that, for $\gamma \lesssim 10^{-4}$, conditions (13) are satisfied over wide ranges of variation of the quantities ν , β_2 , δ , and χ . It follows from expressions (11) and (13) that $n_+ \ll n_{2+}$, and the number of active particles $N_1 + N_2$ varies slightly. We note that, as a consequence of the smallness of N_* , the influence of other (in addition to the processes considered above) possible kinetic processes involving the participation of A^* on the parameters s and $N_1 + N_2$ will also be negligible.

For $\gamma \sim 10^{-4}$ (estimates for the value of I_s are cited in^[1]), the following estimates are obtained from (14) for the magnitude of the effect: $\Delta T(\text{Ar}) \approx 5$ K, $\Delta T(\text{He}) \approx 0.5$ K. However, it should be kept in mind that warming-up processes due to collisions of heavy particles with high-energy electrons impose restrictions on the duration τ_i of the illumination and on the feasibility of observing the effect under steady state conditions. The increase ΔT_n in the temperature of the system is primarily determined by process (9); it can be estimated according to the formula $\Delta T_n = \sigma_i(2\varepsilon - \varepsilon_0)\bar{\rho}\gamma^2 v_i/c_v$ (ε_0 denotes the ionization potential, c_v is the heat capacity per particle). Requiring that $\Delta T_n < \Delta T$ be satisfied for $\sigma_i \sim 10^{-15}$ cm², $(2\varepsilon - \varepsilon_0) \sim 10$ eV ($c_v \sim 10$ k near T_c ^[8]), we obtain $\tau_i \lesssim (10^{-6}$ to 10^{-5} sec). We note that the cited estimate of the warming up is an overestimate because the presence of thermal conductivity should lead to a re-

duction of ΔT_n , since the coefficient of thermal conductivity $\kappa \sim |T - T_c|^{-1/2}$ near T_c .

The requirements on the absence of radiative warming up are reduced in the case of atoms with low-lying energy levels. They should also not be essential for vibrational and rotational molecular transitions. However, the absence of data on the interaction potentials of excited molecules and on kinetic processes does not allow us to make the corresponding estimates. With a strong overestimate, without taking account of emergence of the system into the steady state regime of "flux" equilibrium, here ΔT_n can be estimated as the ratio of the total energy obtained from the laser to the heat capacity of the illuminated layer, $\Delta T_n \sim (\varepsilon \sigma \gamma I_s / c_v) \tau_i$.

The above investigation was based on simple, model concepts with regard to the shape of the interaction potentials and the rates of elementary processes. Refinements are hardly appropriate at the present time, due to the lack of a complete set of the requisite data. The simplifications adopted above should not have any influence on the principal conclusions; however, it should be kept in mind that the utilization of more realistic data may change the quantitative results. For example, if the potentials $u(r)$ and $q(r)$ are markedly shifted one with respect to the other, this should lead to an equation of state which differs from that of van der Waals. The coexistence region will be shifted with respect to the straight line $V = V_c^{vdW}$. We note that one can attempt to observe a radiation-induced change in the properties characterizing the nonideal nature of a gas by utilizing the Joule-Thomson effect (with regard to the change of the inversion temperature), condensation upon rapid adiabatic compression (Wilson cloud chamber), etc.

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¹As is shown in^[5,6] the molecular field approximation gives an equation of state which is in good agreement with the experimental data on condensation even in the case of helium.

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