High-temperature phase transition in the coupled atom-light system in the presence of optical collisions

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The problem of photonic phase transition for the system of a two-level atomic ensemble interacting with a quantized single-mode electromagnetic field in the presence of optical collisions (OCs) is considered. We have shown that for large and negative atom-field detuning a photonic field exhibits high-temperature second-order phase transition to superradiant state under thermalization condition for coupled atom-light states. Such a transition can be connected with superfluid (coherent) properties of photonlike low branch (LB) polaritons. We discuss the application of metallic cylindrical waveguide for observing predicted effects.

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I. INTRODUCTION

Nowadays the investigation of phase transitions in atomic gases represents a huge area of experimental and theoretical research where condensed matter and statistical physics are closely connected to the application problems of quantum and atom optics, for example, in the field of quantum information science (see, e.g., [1]). Although Bose-Einstein condensation (BEC) of the atoms has been observed in many labs, the requirement to use extremely low (up to micro-Kelvins) temperatures strictly limits the utilization of such an effect for practical purposes. It provides an important reason for studying relatively high-temperature phase transitions. Usually such transitions take place in coupled matter-field systems; polaritons introduced many years ago for describing the interaction of quantized field with quantum excitations in the medium (see, e.g., [2]). At present the evidence of phase transitions with LB polaritons and their superfluid properties have been observed in solid state physics with semiconductor microstructures (see, e.g., [3–5]). In such systems polaritons can be treated as 2D gas of bosonic particles, so-called exciton polaritons appearing in the sample of quantum wells inserted in a semiconductor (CdTe/CdMgTe or GaAs) microcavity and having effective mass which is many orders smaller than a free mass of electrons. However, high- (room) temperature phase transition for current *narrow-band* semiconductors seems to be hard to reach due to exciton ionization. At the same time the time of thermalization for polaritons in solid state structures mentioned above is short enough and is in the picoseconds regime [3]. In this sense polaritons in atomic physics could be preferable for observing high-temperature phase transitions. Such polaritons potentially possess a longer coherence time. In particular, atomic polaritons represent superposition of photon and polarization of two- (or multi) level atoms and can be observed in various problems of atom-field interaction where long-lived coherence of quantized optical field strongly coupled with macroscopic atomic ensemble plays an essential role (cf. [6,7]).

The primary step for the experimental observation of phase transitions and BEC for atomic polaritons is connected with achieving thermal equilibrium in the coupled atom-light system. The so-called optical collisions (OCs) have been proposed recently for this purpose. The process of OC represents nonresonant interaction of a quantized light field with an atom in the presence of buffer gas particle (see, e.g., [8]). Although the main features of OCs have been investigated both in theory and in experiment for a long time (see, e.g., [9–11]), the thermodynamic properties of a coupled atom-light system have been studied quite recently [12–16]. In particular, in [12] it has been demonstrated that OCs lead to the thermalization of coupled (dressed) atom-light states under the interaction of the optical field with rubidium atoms in an ultrahigh-pressure buffer gas cell at high (530 K) temperature. Later in [13] we have proposed a theoretical approach of a dressed-state thermalization that accounts for the evolution of pseudospin Bloch vector components and characterizes the essential role of the spontaneous emission rate in the thermalization process. Although we have used a two-level model for describing OC process with buffer gas particles (see, e.g., [9–11]) the dependences theoretically obtained in [13] are qualitatively in good agreement with experimentally observed results for rubidium atoms. The predicted time of thermalization was in a nanosecond domain at full optical power 300 mW and negative atom-light detuning $\delta/2\pi = -11$ THz. The main features of such a thermalization are connected with the observed asymmetry of a saturated line shape that depends on the temperature of atomic gas and detuning $\delta = \omega_L - \omega_{\mathrm{at}}$; ω_L and ω_{at} are frequencies of optical field and atomic transition, respectively.

In a number of papers [15,16] authors discussed thermodynamic properties of a two-level sodium atomic system being under OCs with buffer gas particles in another limit of positive atom-light detuning, that is, for $\delta > 0$. In particular, there has been shown a strong laser generation in the atomic ensemble interacting with cw-laser field in the domain of a far blue wing of spectral line under the frequent collision with buffer gas particles. In this sense both problems, that is, lasing that occurs in atomic system in the presence of OC and phase transition for coupled atom-light states, evoke great interest and should be clarified.

Physically the problems under discussion are very close to another intriguing and long-stated problem of phase transition for a light field (or BEC of photons) (see, e.g., [17–20]). In

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fact, the comparison of lasing and condensation phenomena for photonic, atomic, and solid state systems as two possible ways for achieving macroscopic coherence has long been discussed (see, e.g., [21–23]). The first attempts to consider some analogy between lasing under the threshold region and second-order phase transition in ferromagnets have been made in [21]. Later a simple Dicke model that describes an ensemble of two-level atoms interacting with quantized electromagnetic field in standard laser quantum theories has been proposed for observing phase transition of photons [17]. In quantum optics phase transition in such a system has been interpreted as a transition to some superradiant (coherent) photonic state with zero chemical potential [18]. At the same time it is pointed out in [19] that in this case the establishment of spontaneous static field (a field with zero frequency) in the medium takes place. In fact such a phase transition creates some ferroelectric state in the medium.

From our point of view there exist important circumstances that must be taken into account while considering phase transitions mentioned above. In many cases including usual lasers, we deal in practice with *nonequilibrium* states of the system (cf. [22,24]). On the other hand, Bose-Einstein condensation phenomenon and phase transition under discussion have some meaning only in the limit of thermodynamically equilibrium state of a coupled atom-light system for which chemical potential is nonzero (cf. [25]). In this sense the applicability of a convenient Dicke model for the phase transition problem should be justified in each physical case.

Noticing that in previous experiments [12,13] the region of high and uniform laser intensity was about 70 μm. Physically it means that the medium is thin and the lifetime of photonlike polaritons discussed in this paper seems to be short in comparison with the thermalization time of a coupled atom-light system. However, the implementation of special metallic waveguides of various configurations with the length up to a few millimeters in the OCs experiment performed recently (see [14]) allows us to increase the time of atom-field interaction by many orders due to photon trapping and confinement. In fact, such waveguides pave the way to the investigation of the high-temperature phase transition problem with polaritons in atomic optics.

In the present paper we continue our theoretical investigation of thermodynamic and critical properties of coupled atom-light states appearing due to the interaction of two-level atoms with a single-mode optical field in the presence of buffer gas particles. In Sec. II we examine critical properties of photonic field under the thermalization of coupled atom-light states. In Sec. III we study the problem of a second-order phase transition in the system under discussion. In particular, we suggest a simple model of polaritons describing atom-field interaction at equilibrium. Relying on a thermodynamical approach we consider a mean-field Bardeen-Cooper-Schrieffer (BCS)-like gap equation for order parameter, normalized amplitude of photonic field. In the Appendix we discuss the properties of a cylindrical waveguide for realizing appropriate strong atom-field coupling in a single-mode regime. In conclusion, we summarize the results obtained.

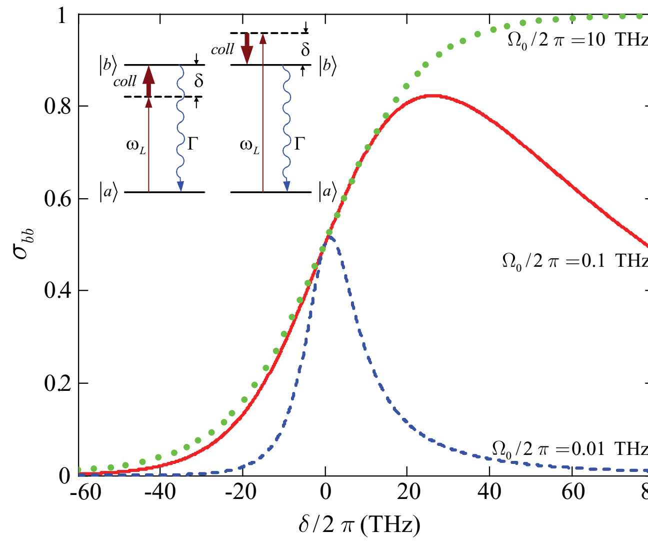

FIG. 1. (Color online) Population of the upper state $\sigma_{\rm bb}$ as a function of atom-field detuning $\delta/2\pi$ for 500-bar argon buffer gas at different values of the resonant Rabi frequency $\Omega_0/2\pi$. The parameters are $\gamma/2\pi = 3.6$ THz and $\Gamma \simeq 2\pi \cdot 6$ MHz. The inset includes the scheme of collisionally aided absorption in a two-level atom for $\delta < 0$ (left panel) and for $\delta > 0$ (right panel).

II. THERMODYNAMIC APPROACH FOR PHOTONIC FIELD UNDER THE OC PROCESS

An optical collision (OC) is an elementary process of a collision between an isolated atom of sort A and a foreign (buffer) gas particle of sort B resulting in the emission or absorption of nonresonant photons (see, e.g., [8]). The inset of Fig. 1 includes a two-level model for characterizing the OC process under consideration. Actually, in recent experiments [12–14] Rabi splitting frequency, atom-field detuning, and collisional broadening are at least three and more orders larger than splitting frequency between hyperfine levels for $5S$-$5P$ transition in rubidium D lines (cf. [26]). As it is demonstrated in [12,13] that the usual pressure-broadened rubidium D_1 and D_2 lines are visible at a moderate optical power $P = 25$ mW independently. In this limit collisional broadening can be treated for D_1 and D_2 lines of rubidium atoms separately. Contrary, at full optical power 300 mW for which the thermalization of coupled atom-light states occurs the essential asymmetry of D_1 and D_2 spectral lines and a common shape of pressure broadening curves are clearly seen in the experiment (cf. [12–14]). Hence the assumption of a two-level approach for the OC problem neglecting hyperfine structure of rubidium D lines seems to be justified (cf. [16]).

We suppose that an optical field with frequency ω_L interacts nonresonantly with two-level atoms having frequency spacing $\omega_{\rm at}$ in the presence of buffer gas particles. Physically OC means that excitation of level $|b\rangle$ is impossible due to atomic collisions only. A theoretical description of coupled atom-light state thermalization can be given by the density matrix approach (see, e.g., [13]). In general, the interaction of two-level atoms with a quantized optical field in the presence of collisions with buffer gas particles is characterized by the following physical parameters. First, there is Rabi frequency $\tilde{\Omega}_R = \Omega_R - \delta\eta/\Omega_R$ that describes the splitting of energy levels in the presence of collisions with buffer gas particles, $\Omega_R = \sqrt{\Omega_0^2 + \delta^2}$ is a definition of the same quantity without collisions when $\eta = 0$; $\Omega_0 = 2g\sqrt{N_{\rm ph}}$ is resonant Rabi splitting taken at detuning $\delta = 0$; $g = (\frac{|d_{\rm ab}|^2\omega_L}{2\hbar\varepsilon_0 V})^{1/2}$ is the atom-field interaction constant, which we assume to be identical for all atoms; $d_{\rm ab}$ is the atomic dipole matrix element; V is the interaction volume; $N_{\rm ph}$ is a total average number of photons involved in the OC process.

The collisions with buffer gas particles are determined by two parameters η and γ which can be expressed phenomenologically via the average phase shift that is accumulated during the collision (cf. [8]). Physically parameters η and γ can be also connected with molecular potentials of a compound $A + B$ system. Parameter η depends on the difference ΔU of diagonal matrix elements for interacting atoms A and B due to their collisions (scattering). According to the quantum mechanical approach to the OC problem, ΔU can be represented as $\Delta U = C_n R^{-n}$ for power law of atomic interaction, C_n is a constant that depends on a character of such interaction, R is a distance between the atom and perturber (buffer gas particle) for a given time, and for more details see [11].

Second, OC process is described by collisional rate (collisional broadening) γ that plays an important role in the thermalization process. In a general case parameter γ is characterized by the density of buffer gas particles, molecular potentials for a compound system, and depends on the value of atom-field detuning δ (see [11]). In the experiment described in [12] parameters γ and η are of the order of terahertz. Skipping some details of our calculations presented in [13], we are starting here with important results related to thermalization of coupled (dressed) atom-field states.

Figure 1 demonstrates a calculated population of the upper state $\sigma_{\rm bb}$ that is proportional to the total intensity of spectral components as a function of atom-field detuning δ taken for the interaction of a quantized field with two-level rubidium atoms characterized by mean resonance frequency of transition 382 THz corresponding to a weighted mean of rubidium D lines (cf. [26]). In the paper we focus on the so-called perturbative limit when atom-field detuning δ is large enough, that is, when inequalities $|\delta| > \gamma$, $\eta > \Omega_0$ are held. In this limit one can neglect phase shift introduced by OC, assuming that $\tilde{\Omega}_R \approx \Omega_R$. At the same time the scaled atomic population of the upper level $\sigma_{\rm bb}$ in this case yields

$$\sigma_{\rm bb} \simeq \frac{1}{2\left(1 + \frac{\Gamma\delta^2}{\gamma\Omega_0^2}\right)}\left[1 + \frac{\delta}{|\delta|}\tanh\left(\frac{\hbar|\delta|}{2k_B T}\right)\right], \tag{1}$$

where Γ is a spontaneous emission rate.

First, we examine the role of atomic collisions in the thermalization process assuming that $\Gamma\delta^2/\gamma\Omega_0^2 \gg 1$; a blue dashed curve in Fig. 1 corresponding to this limit. For negative detuning $\delta < 0$ from (1) we get

$$\sigma_{\rm bb} \simeq \frac{\gamma\Omega_0^2}{\Gamma\delta^2} e^{-\hbar|\delta|/k_B T}, \tag{2}$$

where we also suppose that inequality

$$\hbar|\delta| \gg k_B T \tag{3}$$

is satisfied.

The relation (3) can be recognized as a "low-temperature" limit in the framework of a convenient approach to existing theories for phase transitions and BEC problems (cf. [27]).

The result obtained from (2) is in good agreement with theoretical predictions for the spectral line obtained for a far red wing (so-called adiabatic wing) (see, e.g., [11]). In particular, the curve of spectral intensity decays exponentially in this case.

Now let us switch over to a positive-valued large atom-field detuning, that is, to $\delta > 0$. In this case we get from (1)

$$\sigma_{\rm bb} \approx \frac{\gamma\Omega_0^2}{\Gamma\delta^2}. \tag{4}$$

Equation (4) reproduces a physical result for a far blue wing (so-called, static wing of spectral line) well known from the theory of OCs (cf. [11]). The result is that the population of the upper level is inversely proportional to square of atom-light detuning.

However, according to our theoretical approach important features of population of the upper state are connected with the dependence of $\sigma_{\rm bb}$ on temperature T of atomic gas.

The thermalization of coupled atom-light (dressed) states occurs if condition

$$\Gamma/\gamma \ll \Omega_0^2/\delta^2 \ll 1 \tag{5}$$

is fulfilled. In particular, for large Ω_0 the $\sigma_{\rm bb}$ approaches Dirac-Fermi distribution function

$$\sigma_{\rm bb} \approx \frac{1}{1 + e^{-\hbar\delta/k_B T}}. \tag{6}$$

In [12,13] the best result concerning coupled atom-light state thermalization has been experimentally observed for maximally accessible atom-field detuning $\delta/2\pi = -11$ THz and resonant Rabi frequency $\Omega_0/2\pi = 0.1$ THz (indicated by solid curve in Fig. 1). The obtained time of thermalization was 10 times shorter than the natural lifetime for rubidium D lines.

For further processing it is important to determine a total atom-field excitation (polariton) density ρ as

$$\rho = \lambda^2 + \sigma_{\rm bb}, \tag{7}$$

that we consider to be constant at thermal equilibrium. In (7) $\lambda^2 = \langle f^\dagger f\rangle/N$ is a normalized average photon number, f ($f^\dagger$) is annihilation (creation) operator for the photons absorbed (or emitted) due to atomic collisions at the equilibrium, and N is a number of atoms.

The thermodynamic property of a coupled atom-light system depends on the contribution of photonic and atomic parts in Eq. (7) which is usually considered in some specific limits.

The so-called low density limit $\rho \ll 0.5$ for atom-field excitations (polaritons) implies that

$$\lambda^2 \ll 1, \tag{8a}$$

$$\sigma_{\rm bb} \ll \sigma_{aa} \simeq 1, \tag{8b}$$

which is obtained at negative atom-field detuning $\delta < 0$ at "low-temperature" limit (3), when atoms mostly populate their ground state $|a\rangle$.

Relation (8a) represents the necessary prerequisite for observing a second-order phase transition in various physical systems being at low temperatures (cf. [7,25,27]). As for the problem of OCs a low density limit (8) can be achieved at high temperatures but for a very large atom-field detuning $|\delta|$. An experimentally accessible relative photon number involved in the atom-field interaction and estimated for Rabi frequency $\Omega_0/2\pi = 0.1$ THz is $N_{ph}/N \simeq 6.24 \times 10^{-3}$ (see [12,13]). We assume that inequality (8a) is fulfilled for the system under discussion.

The saturation of atomic population is achieved at excitation density $\rho \approx 0.5$ for $\hbar|\delta| \ll k_B T$. This is the so-called secular approximation (see [8]) for which OCs tend to equalize dressed-state populations which is not the case of our study here.

For positive detuning ($\delta > 0$) from (7) and (8) we obtain $\rho > 0.5$ ($\sigma_{bb} > 0.5$) that corresponds to the limit of population inversion in a two-level atomic ensemble (cf. [15]). Physically, such a system is unstable because of spontaneous emission processes from the upper level. In particular, relevant population σ_{bb} diminishes at a large positive detuning for any fixed laser intensity (see Fig. 1).

To find photonic field properties under OC process at equilibrium we consider (7) as an equation for order parameter λ at the given density ρ. Putting in (7) $\lambda = 0$ for critical value α_c of vital parameter $\alpha \equiv \hbar\delta/k_B T$ that determines the phase boundary between normal and "superradiant" states one can obtain

$$\alpha_C = -\ln\left[(1-\rho)/\rho\right]. \tag{9}$$

Furthermore, one can get from (6)–(8) an expression for order parameter $\lambda(\alpha)$ as a function of atom-field detuning δ or temperature T

$$\lambda(\alpha) = \lambda_\infty \left\{1 - \frac{1}{\rho[1 + (1/\rho - 1)^{\alpha/\alpha_C}]}\right\}^{1/2}, \tag{10}$$

where $\lambda_\infty \equiv \sqrt{\rho}$ is an order parameter at "zero temperature" limit (3).

Equation (10) gives an opportunity to interpret critical properties of the photonic field occurring due to OCs as a result of atom-field thermalization. To be more specific, we suppose atom-light detuning δ to be negative. While modulus $|\delta|$ is small enough (such that $\hbar|\delta| \ll k_B T, \gamma$), the elementary processes of emission (or absorption) of photons by atoms happen independently on the atomic collisions with buffer gas particles during their free motion. The atomic collisions lead to the dephasing of emitted radiation. In this sense they are worse and we deal with a "normal" (incoherent) state for photons.

A physical picture is changed significantly by increasing atom-field detuning δ at high buffer gas pressures. For large $|\delta|$ under the condition of $\hbar|\delta| \gtrsim k_B T$ it is no longer possible to ignore correlations between elementary acts of atomic collisions with buffer gas particles and photon emission (or absorption). This is a case of OCs. Each collision happens in a very short time period compared with the time interval separating two collisions. Watching OCs for a long time when many collisions happen and frequent transitions of atomic population between dressed states occur it is possible to create some population of coupled atom-light states in thermodynamic equilibrium due to the thermalization process. This population can be established for "usual" (bare) atomic levels only for large values of atom-field detuning. In this case Eq. (10) represents transition to some ordering ("superradiant") state as a result of forming macroscopic "spontaneous" polarization for a coupled atom-light system. It should be noticed that the existence of some small residual polarization of the atomic system has been predicted in [13] as a result of the atom-light (dressed) states thermalization.

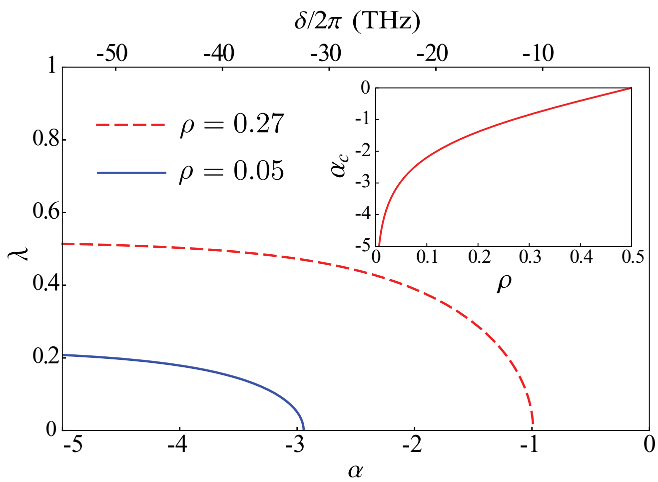

FIG. 2. (Color online) Order parameter λ vs vital parameter α (normalized atom-light detuning δ) for $T = 530$ K temperature of atomic gas. In the inset a phase boundary for critical parameter α_C vs LB polariton density ρ is presented (see the text).

Figure 2 demonstrates phase transition behavior for order parameter λ for the system under discussion. From a practical point of view it is much easier to vary atom-field detuning δ instead of atomic gas temperature T. The dashed curve corresponds to density $\rho = 0.27$ in accordance with experimentally achieved detuning $\delta/2\pi = -11$ THz. The solid curve is relevant to low density limit (8). The critical value $|\alpha_C|$ of parameter α is increased by decreasing excitation density ρ (see inset in Fig. 2). In the low density limit (8) Eq. (10) is simplified and looks like $\lambda(\alpha) \simeq \lambda_\infty[1 - (\rho)^{\alpha/\alpha_C - 1}]^{1/2}$. Phase transition occurs for large enough detuning (or at lower temperatures).

For positive detuning ($\delta > 0$) the problem of phase transition under discussion is sophisticated. Formally, full thermalization of coupled atom-light states and relevant phase transition can be achieved at infinite resonant Rabi splitting frequency Ω_0 and atom-field detuning δ, when the population of the upper level is $\sigma_{bb} = 1$ [see (6)]. However, an approach to OCs that characterizes the thermalization process is valid only for limited values of detuning such as $|\delta| \ll \omega_{at}$. For any finite Ω_0 the variation of atom-field detuning δ or gas temperature T drives a coupled atom-light system out of thermal equilibrium even if such an equilibrium (or quasiequilibrium) has been initially achieved (Fig. 1). In this sense the fulfillment of condition (5) for coupled atom-light states permits us to create a population inversion in a two-level atomic ensemble and to realize nonequilibrium (or quasiequilibrium) transition to lasing as a result (cf. [15,16]).

III. PHASE TRANSITION IN A COUPLED ATOM-LIGHT SYSTEM UNDER EQUILIBRIUM

While OCs result in thermalization of coupled (dressed) atom-light states, Eq. (10) describes the phase transition under discussion only *qualitatively*. Strictly speaking, a rigorous thermodynamical approach to critical properties of a coupled atom-light system at equilibrium is needed.

We describe the interaction of a single-mode light field f with an atomic ensemble by relevant (Dicke) Hamiltonian (cf. [13,17])

$$H = \hbar\omega_L f^\dagger f + \frac{\hbar\omega_{\rm at}}{2}\sum_{j=1}^{N} S_{z,j} + \frac{\hbar\kappa}{\sqrt{N}}\sum_{j=1}^{N}(S_{-,j}^\dagger f + f^\dagger S_{-,j}), \tag{11}$$

where $\kappa = g\sqrt{N}$ is a collective parameter of atom-field interaction, $S_{-,j}$ is a transition operator for jth atom, and $S_{z,j}$ is an operator of atomic population imbalance.

From the mathematical point of view for the definition of $S_{-,j}$ and $S_{z,j}$ operators it is possible to use Pauli spin matrices representation (cf. [18]) or explore annihilation (a_j, b_j) and creation ($a_j^\dagger$, $b_j^\dagger$) operators for bosonic atoms at the ground $|a\rangle$ and excited $|b\rangle$ states in the second quantization representation, respectively. The latter one enables definitions $S_{-,j} = a_j^\dagger b_j$ and $S_{z,j} = b_j^\dagger b_j - a_j^\dagger a_j$ (see [28]).

Since the atomic medium is very dense ($n_{\rm at} \simeq 10^{16}$ cm^{-3}) one can neglect inhomogeneous (Doppler) broadening because Doppler broadening does not restrict the thermalization process and is by two orders smaller than parameter κ, that is, $\kappa/2\pi \approx 0.624$ THz for resonant Rabi frequency $\Omega_0/2\pi = 0.1$ THz (cf. [12,13]). Hence the so-called strong atom-field coupling condition that plays an important role in the problem of phase transition with polaritons is fulfilled in our case (cf. [3–5]). In this limit one can introduce collective atomic ladder operator S_- and operator of atomic population imbalance S_z as follows:

$$S_- = \sum_{j=1}^{N} S_{-,j}, \tag{12a}$$

$$S_z = \sum_{j=1}^{N} S_{z,j}. \tag{12b}$$

Operators introduced in (12) obey su(2) algebra commutation relations

$$[S_-,S_-^\dagger] = -S_z, \tag{13a}$$

$$[S_z,S_-] = -2S_-. \tag{13b}$$

For describing excitations of a two-level atomic system we define annihilation (ϕ) and creation ($\phi^\dagger$) excitation operators by using the so-called Holstein-Primakoff transformation (cf. [29])

$$S_- = \sqrt{N-\phi^\dagger\phi}\,\phi, \tag{14a}$$

$$S_-^\dagger = \phi^\dagger\sqrt{N-\phi^\dagger\phi}, \tag{14b}$$

$$S_z = 2\phi^\dagger\phi - N. \tag{14c}$$

The relations (13) are preserved when operators ϕ and $\phi^\dagger$ obey usual commutation relation for the Bose system:

$$[\phi,\phi^\dagger] = 1. \tag{15}$$

For a large number of atoms N it is possible to treat operators S_- and $S_-^\dagger$ as

$$S_- = \sqrt{N}\phi - \frac{\phi^\dagger\phi^2}{2\sqrt{N}}, \tag{16a}$$

$$S_-^\dagger = \sqrt{N}\phi^\dagger - \frac{{\phi^\dagger}^2\phi}{2\sqrt{N}}. \tag{16b}$$

The last term in (16) characterizes nonlinear effects for atomic excitations under the atom-field interaction. However, in the low density limit defined as

$$\phi^\dagger\phi \ll N \tag{17}$$

we can obtain from (16) simple expressions for operators ϕ and $\phi^\dagger$, that is, we get

$$P \equiv \phi = \frac{1}{\sqrt{N}}S_-, \tag{18a}$$

$$P^\dagger \equiv \phi^\dagger = \frac{1}{\sqrt{N}}S_-^\dagger. \tag{18b}$$

Taking into account Eq. (14c), it is easy to see that condition (17) exactly corresponds to the relation (8b) obtained for large and negative atom-field detuning $\delta < 0$. In this case collective atomic excitations described by operator ϕ characterize a macroscopic polarization P of atomic system.

Taking into account Eqs. (14c), (17), and (18), Hamiltonian (11) can be easily reduced to the form

$$H = \hbar\omega_L f^\dagger f + \hbar\omega_{\rm at}\phi^\dagger\phi + \hbar\kappa(\phi^\dagger f + f^\dagger\phi). \tag{19}$$

At the steady state for a coupled atom-light system Hamiltonian (19) can be diagonalized with the help of unitary transformations

$$\Phi_1 = \vartheta_1 f + \vartheta_2\phi, \tag{20a}$$

$$\Phi_2 = \vartheta_1\phi - \vartheta_2 f, \tag{20b}$$

where $\vartheta_{1,2}^2 = \frac{1}{2}(1 \pm \frac{\delta}{\sqrt{\delta^2+4\kappa^2}})$ are Hopfield coefficients that obey normalization condition $\vartheta_1^2 + \vartheta_2^2 = 1$.

The annihilation operators $\Phi_{1,2}$ in Eqs. (20a) and (20b) characterize two types of quasiparticles due to the atom-field interaction, that is, upper and lower branch polaritons. By using relation (15) it is easy to show that operators defined in (20a) and (20b) obey commutation relations

$$[\Phi_1,\Phi_1^\dagger] = [\Phi_2,\Phi_2^\dagger] = 1. \tag{21}$$

Below we focus on photonlike polaritons necessary for explaning photonic phase transition for the light field in the system under discussion.

The equation for order parameter λ at equilibrium can be derived from a variational (thermodynamic) approach (see, e.g., [18]). In their calculations the authors use a canonical ensemble with chemical potential $\mu = 0$ applied to Dicke Hamiltonian (11). However, this approach cannot be justified even for a pure photonic system posing phase transition at equilibrium. For example, as it is shown in [30] the chemical potential for photonic gas being under BEC condition is nonzero.

In the paper we use a grand canonical ensemble with finite chemical potential and coherent basis for the photonic field for calculating partition function $Z(N,T) = \mathrm{Tr}(e^{-H'/k_BT})$, $H' = H - \mu N_{\mathrm{ex}}$ is a modified Hamiltonian, and $N_{\mathrm{ex}} = f^\dagger f + \frac{1}{2}\sum_{j=1}^{N} S_{z,j}$ is the number of excitations.

Evaluating partition function $Z(N,T)$ under the mean-field approximation one can obtain (cf. [18])

$$\tilde{\omega}_L \lambda = \frac{\kappa^2 \lambda \tanh\left[\frac{\hbar}{2k_BT}\left(\tilde{\omega}_{\mathrm{at}}^2 + 4\kappa^2\lambda^2\right)^{1/2}\right]}{\left(\tilde{\omega}_{\mathrm{at}}^2 + 4\kappa^2\lambda^2\right)^{1/2}}, \tag{22}$$

where we introduced denotations $\tilde{\omega}_L \equiv \omega_L - \mu$, $\tilde{\omega}_{\mathrm{at}} \equiv \omega_{\mathrm{at}} - \mu$.

Equation (22) is similar to the gap equation that characterizes a second-order phase transition for various systems in solid-state physics (see, e.g., [27]). For finding chemical potential μ in our case one can use polariton number operator $N_{\mathrm{pol}} = \Phi_1^\dagger\Phi_1 + \Phi_2^\dagger\Phi_2$. With the help of Eqs. (12b), (14c), and (20) N_{pol} can be represented as

$$N_{\mathrm{pol}} = f^\dagger f + \sum_{j=1}^{N} b_j^\dagger b_j = \frac{N}{2} + N_{\mathrm{ex}}. \tag{23}$$

Density ρ of total excitations occurring in a closed atom-light field system defined in (7) can be interpreted as a polariton number density, that is, $\rho \equiv N_{\mathrm{pol}}/N$. In the thermodynamic limit it approaches

$$\rho = \lambda^2 + \frac{1}{2}\left\{1 - \frac{\tilde{\omega}_{\mathrm{at}} \tanh\left[\frac{\hbar}{2k_BT}\left(\tilde{\omega}_{\mathrm{at}}^2 + 4\kappa^2\lambda^2\right)^{1/2}\right]}{\left(\tilde{\omega}_{\mathrm{at}}^2 + 4\kappa^2\lambda^2\right)^{1/2}}\right\}. \tag{24}$$

The last term in the brackets of Eq. (24) characterizes normalized population imbalance $\tilde{S}_z = S_z/N$ at thermal equilibrium. In particular, it is (cf. [25])

$$\tilde{S}_z^{(\mathrm{eq})} = -\frac{\tilde{\omega}_{\mathrm{at}} \tanh\left[\frac{\hbar}{2k_BT}\left(\tilde{\omega}_{\mathrm{at}}^2 + 4\kappa^2\lambda^2\right)^{1/2}\right]}{\left(\tilde{\omega}_{\mathrm{at}}^2 + 4\kappa^2\lambda^2\right)^{1/2}}. \tag{25}$$

Combining (24) and (22) for chemical potential μ we get

$$\mu_{1,2} = \tfrac{1}{2}(\omega_{\mathrm{at}} + \omega_L \pm \Omega_{R,\mathrm{eff}}), \tag{26}$$

where $\Omega_{R,\mathrm{eff}} = \sqrt{\delta^2 - 8\kappa^2\left(\rho - \lambda^2 - \frac{1}{2}\right)}$ is a new effective Rabi splitting frequency.

At low polariton densities [see (8)] Eq. (26) defines normal state ($\lambda = 0$) for upper (μ_1) and lower (μ_2) polariton branch frequencies, respectively. Inserting (26) into (22) we arrive at

$$\tilde{\omega}_{L1,2} = \frac{\kappa^2 \tanh\left[\frac{\hbar}{2k_BT}\left(\tilde{\omega}_{\mathrm{at}1,2}^2 + 4\kappa^2\lambda^2\right)^{1/2}\right]}{\left(\tilde{\omega}_{\mathrm{at}1,2}^2 + 4\kappa^2\lambda^2\right)^{1/2}}, \tag{27}$$

that are BCS-like equations for upper (index "1") and lower (index "2") branch polaritons, respectively, $\tilde{\omega}_{L1,2} = \frac{1}{2}[\delta \mp \Omega_{R,\mathrm{eff}}]$, $\tilde{\omega}_{\mathrm{at}1,2} = \frac{1}{2}[-\delta \mp \Omega_{R,\mathrm{eff}}]$.

The critical temperature T_C of phase transition (for given atom-field detuning δ) can be obtained from (27) for $\lambda = 0$ and looks like

$$T_{C1,2} = \frac{\hbar|\tilde{\omega}_{\mathrm{at}1,2}|}{2k_B \tanh^{-1}[\pm(2\rho - 1)]}. \tag{28}$$

The results obtained from Eq. (28) are consistent with existing theories of BCS-like phase transition in polariton system (cf. [7,25]).

For very large atom-light detuning, that is, for $|\delta| \gg \kappa$, a normalized population imbalance (25) looks like

$$\tilde{S}_z^{(\mathrm{eq})} \approx \frac{\delta}{|\delta|}\tanh\left(\hbar\,|\delta|\,/2k_BT\right). \tag{29}$$

Equation (29) corresponds to equilibrium population of the upper level achieved at full thermalization of coupled atom-light states in the presence of OCs and described by Eq. (6). In the same limit the critical temperature T_c approaches

$$T_c = \frac{\hbar\delta}{2k_B \tanh^{-1}(2\rho - 1)}. \tag{30}$$

Equation (30) immediately comes from Eq. (7) taken for order parameter $\lambda = 0$.

At the same time Eq. (28) defines critical value α_C of α parameter (normalized atom-field detuning δ) for fixed temperature T of atomic gas for which we get

$$\alpha_C = -\ln\left(\frac{1-\rho}{\rho}\right) - \frac{2\hbar^2\kappa^2(\rho - 1/2)}{k_B^2 T^2 \ln[(1-\rho)/\rho]}. \tag{31}$$

Equation (31) characterizes phase boundary between photonic superradiant (coherent) and normal states taken at $\lambda = 0$ for the given temperature T of an atomic ensemble. In accordance with experimental conditions of [12–14], $k_BT \gg \hbar\kappa$ and the last term in (31) vanishes. In this case we recognize Eq. (9) for critical parameter α_C that characterizes the OC process at full equilibrium.

For negative atom-light detuning, that is, for $\delta < 0$, we can interpret the phase transition under consideration as a transition to superfluid state of LB photonlike polaritons ($\Phi_2 \simeq -f$) occurring under the OC process in the low-density limit (8). In this case order parameter λ can be recognized as a polariton wave function. In the Appendix we represent the main features of cylindrical waveguides for observing phase transition with polaritons. Superfluid properties of polaritons can be elucidated taking into account a weak interaction between them and can be described in the momentum representation (cf. [4]). Such an interaction arises due to nonlinear effects in atom-field interaction, described by second terms in expressions (16) for S_- and $S_-^\dagger$ (cf. [31]). We expect that in the future it will be possible to examine superfluid properties of such polaritons by using very thin-walled (up to 60 nm) waveguides for the experiment with Josephson junctions at high enough temperatures.

IV. CONCLUSIONS AND OUTLOOK

We consider a phase transition problem in a two-level atomic ensemble strongly interacting with an optical field. The main features of such a phase transition are connected with a coupled atom-light state thermalization occurring due to the

OCs process with buffer gas particles. Such a thermalization can be achieved experimentally for a large value of negative atom-field detuning δ. Using a thermodynamic approach (partition function) we established a gap equation under a mean-field approximation for order parameter λ, normalized average optical field amplitude. In the paper we present simple arguments confirming that the obtained thermalization of coupled atom-light states leads to the photonic phase transition to the superradiant (coherent) state and is characterized by some ordering (equilibrium state) for two-level atomic system. Nontrivial solution of Eq. (22) with $\lambda \neq 0$ leads to the appearance of a macroscopic stationary polarization of the atomic medium that is proportional to the order parameter λ.

The equilibrium properties of a coupled atom-light system offer a promising approach to studying the critical behavior of photonlike LB polaritons under the low-density limit (8). In this respect we discuss special metallic waveguides for providing necessary atom-field interaction and achieving suitable polariton lifetime, as a result. In particular, the phase transition under discussion can be connected with transition to the superfluid (coherent) state for such polaritons characterizing superposition of a quantized optical field and a macroscopic atomic polarization. In the presence of the phase transition the atomic polarization evolves in time with the frequency that is equal to the chemical potential $\mu_2 \approx \omega_{\rm at} - |\delta|$ for LB polaritons. This property enables us to observe the phase transition under discussion experimentally.

We note that the results obtained above can be useful for observing true BEC occurring with atomic polaritons. In this case we need to realize certain trapping potential for LB polaritons. One simple way for that is to use a biconical waveguide cavity for which waveguide radius R varies smoothly along the z coordinate. A similar dielectric (bottle-like) cavity based on tapered optical fiber has been recently proposed for improving atom-light coupling strength (see, e.g., [32]). In our case such a cavity enables us to create an appropriate trapping potential for photons as well as for polaritons in the z dimension. It is important that in this case the lifetime of photonlike polaritons can be long enough and is determined by the cavity Q factor. These problems will be a subject of intensive study both in theory and experiment in forthcoming papers.

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APPENDIX A: PHOTON CONFINEMENT IN CYLINDRICAL WAVEGUIDE

Let us briefly discuss the possibility of observing the phase transition under discussion with LB polaritons in a metallic cylindrical waveguide. The implementation of metallic microtubes for investigating coupled atom-light state thermalization under OC has been demonstrated in [14]. The properties of an optical field in the ideal empty waveguide without any losses are well known (see, e.g., [33]). In particular, wave vector component $k_{mp} = g_{mp}/R$ of the field in the cross section of the waveguide is orthogonal to the z axis and quantized; g_{mp} represents the root of the equation $J_m(k_\perp R) = 0$, R is a waveguide radius, and m and p are integer numbers characterizing azimuthal and transversal field distribution respectively. At the same time there exist continuum modes with wave vectors k_z in the z direction. Physically it means that it is possible to establish a dispersion relation for photonic field in the waveguide as

$$\omega_L \simeq ck_{\perp,mp} + \hbar k_z^2/2m_{\rm ph}, \tag{A1}$$

where we have introduced photon mass $m_{\rm ph} = \hbar k_{\perp,mp}/c$. Thus, for a cylindrical waveguide spatial degrees of freedom in x and y directions are suppressed and photons remain confined in the plane which is perpendicular to the z axis. At the same time Eq. (A1) implies the existence of a finite polariton mass in the waveguide. For example, mass $m_{\rm pol}$ of photonlike polaritons is about 2.6×10^{-36} kg which implies a high-temperature phase transition in our case (cf. [3–5]).

It is possible to define from Eq. (A1) the so-called *cutoff wave number* $k_{mp}^{(c)} = k_{\perp,mp} = g_{mp}/R$ for which a waveguide mode propagation constant vanishes. In the experiment it is preferable to use a fundamental TM_{01} mode with quantum numbers $m = 0$, $p = 1$ ($g_{01} = 2.4048$) as a cavity mode for the atom-field interaction. In particular, we require the fulfillment of conditions $\mu_{1,2},\omega_L > ck_{01}^{(c)}$ for characteristic frequencies that describe a coupled atom-light system. The condition under discussion can be represented in some other way by using radius R of the waveguide as $c\frac{g_{01}}{\omega_{\rm at}-|\delta|} < R < c\frac{g_{11}}{\omega_{\rm at}+|\delta|}$, where $g_{11} \simeq 3.8317$. The fulfillment of the last inequality guarantees that only a single waveguide mode effectively interacts with the atomic ensemble at a large atom-light detuning δ. Notably, that according to the condition presented above the diameter of the waveguide should be approximately of the order of wavelength $\lambda_{\rm Rb} \simeq 785$ nm for rubidium atom D-line transition (cf. [14]).

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