Dense dipole-dipole-coupled two-level systems in a thermal bath

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The quantum dynamics of a dense and dipole-dipole-coupled ensemble of two-level emitters interacting via their environmental thermostat is investigated. The static dipole-dipole interaction strengths are being considered strong enough but smaller than the transition frequency. Therefore, the established thermal equilibrium of ensemble's quantum dynamics is described with respect to the dipole-dipole-coupling strengths. We have demonstrated the quantum nature of the spontaneously scattered light field in this process for weaker thermal baths as well as non-negligible dipole-dipole couplings compared to the emitter's transition frequency. Furthermore, the collectively emitted photon intensity suppresses or enhances depending on the environmental thermal baths' intensities.

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I. INTRODUCTION

The mutual interactions among multiple excited two-level emitters are being mediated by their environmental vacuum electromagnetic field reservoir. Depending on the interparticle separations, these interactions diminish or can be enhanced, respectively [1–9]. Certain of these emission-absorption collective features were demonstrated experimentally which resulted, for instance, in superradiance and subradiance [10–13] and superabsorption [14–16], as well as in various phase transitions phenomena [17,18]. Collective interactions are found useful in explaining the sonoluminescence phenomenon [19] or the tandem superradiant emission of light and atoms in a Bose-Einstein condensate [20], respectively. Furthermore, applications of collective phenomena towards lasing effects in various frequency ranges are being widely recognized [21,22].

In a thermal electromagnetic field environmental reservoir, the quantum dynamics of a small two-level ensemble changes accordingly. Particularly, it was demonstrated that the entanglement between two initially independent qubits can be generated if the two qubits interact with a common heat bath in thermal equilibrium [23–26]. Larger two-level ensembles, under the Dicke approximation [1–5] and in a common thermostat, obey the Bose-Einstein statistics and there are no critical steady-state behaviors in the thermodynamic limit which are typical to laser-driven atomic ensembles [5,17,18]. Moreover, quantum light features are characteristic only for few atoms in these setups [27,28]. Contrarily, the electromagnetic fields emitted on the two transitions of a three-level Dicke-like ensemble, surrounded by the thermostat, are strongly correlated or anticorrelated and exhibit quantum light properties also for larger numbers of atoms. This is demonstrated via violation of the Cauchy-Schwarz inequality [29].

The multiqubit collective phenomena, mediated by environmental thermal baths, have attracted an additional interest recently due to feasible applications related to quantum thermodynamics [30–33]. In this respect, the performance of quantum heat engines [34–39], quantum refrigerators [40,41], or quantum batteries [42–44] has been shown to improve considerably due to cooperativity among their constituents.

Motivated by these advances regarding multiqubit ensembles and their applications, here we investigate the quantum dynamics of a small and strongly dipole-dipole interacting two-level ensemble in thermal equilibrium with its surrounding thermostat. The spontaneously scattered quantum light features, in this process, were addressed for the case when the dipole-dipole interaction strength among the two-level radiators is comparable to, but still smaller than, the transition frequency of a single emitter, respectively. We have obtained the corresponding master equation describing this multiqubit ensemble and solved it analytically in the steady state which characterizes the established thermal equilibrium. Furthermore, we have demonstrated that the thermal driven small two-level ensemble emits light which has sub-Poissonian statistics for weaker thermal baths, i.e., for lower temperatures. For negligible dipole-dipole couplings, it is known, see, e.g., Ref. [27], that this photon statistics occurs for a few emitters only and stronger thermal baths that may prevent its detection because of the thermal photon background. The collectively spontaneously scattered photon intensity enhances for stronger as well as weaker thermal baths due to dipole-dipole interactions among the involved two-level emitters, while it suppresses for moderately intense to moderately weak intensities of the environmental heat reservoirs, respectively. Actually, strong dipole-dipole interactions are characteristics of Rydberg atoms and the dipole-dipole coupling among two Rydberg atoms separated by 15 μm was experimentally

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demonstrated in [45]. Therefore, ensembles of two-level Rydberg atoms [46–50] may form a physical platform where the effects described here might be realized experimentally.

This paper is organized as follows. In Sec. II we describe the analytical approach and the system of interest, while in Sec. III we discuss the first- and the second-order photon correlation functions, respectively. Section IV presents and analyzes the obtained results. The article concludes with a summary given in Sec. V.

II. THEORETICAL FRAMEWORK

We consider an ensemble of N dipole-dipole-coupled two-level emitters, each having the transition frequency ω_0, and interacting with the environmental thermal reservoir at temperature T. The atomic subsystem is being densely packed so that its linear dimensions are smaller than the photon emission wavelength λ, i.e., within the Dicke approximation [1–4]. The static dipole-dipole-coupling strength $\delta \sim d^2/r^3$ [4,14], where r is the mean distance among any of the atomic pairs characterized by the dipole d, is strong such that the ratio $\delta/\omega_0 < 1$ is not negligible and may play a relevant role over the established thermal equilibrium features. Also, the dipole-dipole-coupling strength is taken equal for any atomic pair—a reasonable approximation in the Dicke limit [4]. Alternatively, one can assume a Gaussian-distributed atomic cloud to obtain an averaged dipole-dipole-coupling strength δ; see, e.g., Ref. [51]. Hence the Hamiltonian describing this system in the dipole and rotating-wave approximations [1–6,17] can be represented as follows: $H = H_0 + H_i$, where

$$H_0 = \sum_k \hbar\omega_k a_k^\dagger a_k + \hbar\omega_0 S_z - \hbar\delta \sum_{j\neq l} S_j^+ S_l^- \tag{1}$$

and

$$H_i = i\sum_k \{(\vec{g}_k \cdot \vec{d})a_k^\dagger S^- - \text{H.c.}\}. \tag{2}$$

Here, the collective atomic operators $S^+ = \sum_{j=1}^N S_j^+ = \sum_{j=1}^N |e\rangle_{jj}\langle g|$ and $S^- = [S^+]^\dagger$ obey the usual commutation relations for su(2) algebra, namely, $[S^+, S^-] = 2S_z$ and $[S_z, S^\pm] = \pm S^\pm$, where $S_z = \sum_{j=1}^N S_{zj} = \sum_{j=1}^N (|e\rangle_{jj}\langle e| - |g\rangle_{jj}\langle g|)/2$ is the bare-state inversion operator. Further, $|e\rangle_j$ and $|g\rangle_j$ are the excited and ground state of the emitter j, respectively, while $a_k^\dagger$ and a_k are the creation and the annihilation operators of the environmental electromagnetic field (EMF) thermal reservoir, which satisfy the standard bosonic commutation relations, that is, $[a_k, a_{k'}^\dagger] = \delta_{kk'}$ and $[a_k, a_{k'}] = [a_k^\dagger, a_{k'}^\dagger] = 0$ [52]. In the Hamiltonian (1), the free energies of the environmental EMF thermal modes and atomic subsystems are represented by the first two terms, respectively. The dipole-dipole interaction Hamiltonian is given by the third term of the Hamiltonian (1). Since $S^+S^- = \sum_{j\neq l} S_j^+ S_l^- + \sum_j S_{zj} + N/2$, the Hamiltonian H_0, can be represented as

$$H_0 = \sum_k \hbar\omega_k a_k^\dagger a_k + \hbar\bar{\omega}_0 S_z - \hbar\tilde{\delta} S^+ S^-, \tag{3}$$

where $\bar{\omega}_0 = \omega_0 + \tilde{\delta}$, while the constant $N/2$ is being dropped. Note here that the Hamiltonian describing many two-level radiators is an additive function, i.e., it consists of a sum of individual Hamiltonians, characterizing separately each two-level emitter. From this reason, the dipole-dipole-coupling strength δ was divided on $N-1$, i.e., $\tilde{\delta} = \delta/(N-1)$, since there are $N(N-1)$ terms describing the dipole-dipole interacting atoms; see, e.g., Ref. [4]. The Hamiltonian (2), i.e., H_i, accounts for the interaction of the whole ensemble with the environmental thermostat. There, $\vec{g}_k = \sqrt{2\pi\hbar\omega_k/V}\vec{e}_p$ is the coupling strength among the few-level emitters and the thermal EMF modes. Here, $\vec{e}_p$ is the photon polarization vector with $p \in \{1, 2\}$ and V is the quantization volume, respectively.

The general form of the master equation, describing the atomic subsystem alone in the interaction picture, is given by [5]

$$\frac{d}{dt}\rho(t) = -\frac{1}{\hbar^2}\text{Tr}_\text{f}\left\{\int_0^t dt'[H_I(t), [H_I(t'), \rho(t')]]\right\}, \tag{4}$$

where

$$H_I(t) = U(t)H_i U^{-1}(t), \tag{5}$$

with $U(t) = e^{iH_0 t/\hbar}$, and the notation $\text{Tr}_\text{f}\{\cdots\}$ means the trace over the thermal EMF degrees of freedom. The explicit expression of the interaction Hamiltonian in the interaction picture is then

$$H_I = i\sum_k (\vec{g}_k \cdot \vec{d})a_k^\dagger e^{i(\omega_k - \hat{\omega})t} S^- + \text{H.c.}, \tag{6}$$

where

$$\hat{\omega} = \bar{\omega}_0 + 2\tilde{\delta} S_z. \tag{7}$$

Next, one substitutes the Hamiltonian (6) in the equation (4) and performs the trace over the thermal EMF degrees of freedom taking as well into account the operator nature of the exponential function. Subsequently, under the Born-Markov approximations, one arrives at the following master equation describing the atomic subsystem only:

$$\begin{aligned}\frac{d}{dt}\rho(t) &+ i[\bar{\omega}_0 S_z - \tilde{\delta} S^+ S^-, \rho] \\ &= -\left[S^+, \frac{\hat{\Gamma}(\hat{\omega})}{2}\big(1 + \bar{n}(\hat{\omega})\big)S^-\rho\right] \\ &\quad -\left[S^-, S^+\frac{\hat{\Gamma}(\hat{\omega})}{2}\bar{n}(\hat{\omega})\rho\right] + \text{H.c.}\end{aligned} \tag{8}$$

Here $\hat{\Gamma}(\hat{\omega}) = 2d^2\hat{\omega}^3/(3\hbar c^3)$ is the emitters' spontaneous decay rate, while $\bar{n}(\hat{\omega}) = [\exp(\hbar\hat{\omega}/k_B T) - 1]^{-1}$ is the mean thermal photon number, both at the eigenvalues of $\hat{\omega}$, respectively.

While finding the general solution of the master equation (8) is not a trivial task at all for a many-atom sample, it possesses a steady-state solution. One can observe, by a direct substitution in Eq. (8), that the expression

$$\rho_s = Z^{-1} e^{-\beta H_a} \tag{9}$$

is a steady-state solution of it with the parameter Z being the normalization constant determined by the requirement $\text{Tr}\{\rho_s\} = 1$. Here,

$$H_a = \hbar\bar{\omega}_0 S_z - \hbar\tilde{\delta} S^+ S^- \tag{10}$$

and $\beta = (k_B T)^{-1}$, whereas k_B is the Boltzmann's constant. Considering an atomic coherent state $|n\rangle$, denoting a symmetrized N-atom state in which $N-n$ particles are in the lower state $|g\rangle$ and n atoms are excited to their upper state $|e\rangle$, and that $S^-|n\rangle = \sqrt{n(N-n+1)}|n-1\rangle$, $S^+|n\rangle = \sqrt{(N-n)(n+1)}|n+1\rangle$, and $S_z|n\rangle = (n-N/2)|n\rangle$, one can calculate the steady-state expectation values of any atomic correlators of interest.

For a negligible ratio of the dipole-dipole-coupling strength over the transition frequency ω_0, i.e., $\delta/\omega_0 \to 0$, the master equation (8) would turn into the usual master equation describing a collection of two-level atoms collectively interacting via their thermal reservoir [3,5,27] in the Dicke limit, namely, $\dot\rho(t) + i[\bar\omega_0 S_z - \tilde\delta S^+S^-, \rho] = -\Gamma(\omega_0)(1+\bar n(\omega_0))[S^+, S^-\rho]/2 - \Gamma(\omega_0)\bar n(\omega_0)[S^-, S^+\rho]/2 + \text{H.c.}$, where the overdot means differentiation with respect to time; see Appendix A. Its steady-state solution is known, i.e., $\rho_s = Z^{-1}\exp[-\hbar\omega_0 S_z/k_B T]$, see, e.g., Refs. [5,27], and the corresponding steady-state atomic quantum dynamics does not depend on δ as long as the dipole-dipole interactions are too weak.

Therefore, in the next sections, we shall focus on the quantum statistics of the spontaneously scattered photons in the process of dipole-dipole interacting two-level emitters via their environmental thermal reservoir and for non-negligible dipole-dipole-coupling strengths such that $\delta/\omega_0 < 1$. In this regard, we introduce, respectively, the first- and the second-order photon correlation functions describing this feature.

III. PHOTON CORRELATION FUNCTIONS

The first- and the unnormalized second-order photon correlation functions at position $\vec R$ can be represented as follows [52–55]:

$$G_1(\vec R, t) = \langle \vec E^{(-)}(\vec R, t)\vec E^{(+)}(\vec R, t)\rangle,$$
$$G_2(\vec R, t) = \langle : \vec E^{(-)}(\vec R, t)\vec E^{(+)}(\vec R, t) \times \vec E^{(-)}(\vec R, t)\vec E^{(+)}(\vec R, t) :\rangle, \quad (11)$$

where

$$\vec E^{(-)}(\vec R, t) = \sum_k \vec g_k a_k^\dagger(t) e^{-i\vec k\cdot\vec R},$$
$$\vec E^{(+)}(\vec R, t) = \sum_k \vec g_k a_k(t) e^{i\vec k\cdot\vec R}, \quad (12)$$

while $: f(\vec R, t) :$ indicates normal ordering. In the far-zone limit of experimental interest $R = |\vec R| \gg \lambda$, one can express the first- and second-order correlation functions via the collective atomic operators. In this respect, one solves formally the Heisenberg equations for EMF operators $\{a_k^\dagger(t), a_k(t)\}$ using the Hamiltonian (6). Then, introducing those solutions in Eqs. (11) and integrating over all involved variables, see, e.g., Ref. [56], one arrives finally at

$$G_1(\vec R, t) = \Phi(R)\langle S^+(t)\hat\omega^4 S^-(t)\rangle,$$
$$G_2(\vec R, t) = \Phi(R)^2\langle S^+(t)\hat\omega^2 S^+(t)\hat\omega^4 S^-(t)\hat\omega^2 S^-(t)\rangle. \quad (13)$$

Here $\Phi(R) = d^2(1-\cos^2\zeta)/(c^4R^2)$, while ζ is the angle between the direction of vector $\vec R$ and the dipole $\vec d$.

The normalized second-order photon-photon correlation function, defined in the usual way, namely,

$$g^{(2)}(t) = \frac{G_2(\vec R, t)}{G_1(\vec R, t)^2}, \quad (14)$$

takes the next form in the steady state,

$$g^{(2)}(0) = \frac{\langle S^+\hat\omega^2 S^+\hat\omega^4 S^-\hat\omega^2 S^-\rangle}{\langle S^+\hat\omega^4 S^-\rangle^2}. \quad (15)$$

Note that $g^{(2)}(0) < 1$ characterizes sub-Poissonian, $g^{(2)}(0) > 1$ super-Poissonian, and $g^{(2)}(0) = 1$ Poissonian photon statistics.

In the following section, we shall investigate the second-order photon-photon correlation function (15) with the help of the steady-state solution (9). Actually, the focus will be on comparing $g^{(2)}(0)$ when $\delta/\omega_0 \to 0$, a case which is known in the scientific literature, with the situation considered here, namely, when $\delta/\omega_0 \neq 0$ but still $\delta/\omega_0 < 1$.

IV. RESULTS AND DISCUSSION

We start our discussions by presenting some limiting cases of the second-order photon-photon correlation function given by Eq. (15). Particularly, for $\eta \to 0$, where

$$\eta = \frac{\delta}{\omega_0},$$

we recover the known results (see, e.g., Ref. [27]), namely,

$$g^{(2)}(0) = \frac{6(N+3)(N-1)}{5N(N+2)}, \quad \text{if } x \to 0,$$
$$g^{(2)}(0) = 2 - \frac{2}{N}, \quad \text{when } x \gg 1, \quad (16)$$

where

$$x = \frac{\hbar\omega_0}{k_B T}.$$

However, for $\eta \neq 0$ and strong thermal baths, i.e., $x \to 0$, and to the second order in the small parameter η, we have obtained that

$$g^{(2)}(0) = \frac{6(N+3)(N-1)}{5N(N+2)} + \frac{48(N+3)(N^2+2N-18)\eta^2}{25N(N-1)(N+2)}. \quad (17)$$

Contrary, for weaker thermal baths when $x \gg 1$, we have obtained the following expression for the second-order photon-photon correlation function:

$$g^{(2)}(0) = 2\left(1 - \frac{1}{N}\right)\left(\frac{N-1-(N-3)\eta}{(N-1)(1-\eta)}\right)^4 \times \exp\left(-\frac{2\eta x}{N-1}\right). \quad (18)$$

Notice that Eqs. (17) and (18) turn, respectively, into those given by Eq. (16), when $\eta \to 0$. Moreover, the photon statistics, characterized by Eq. (18) for larger values of the parameter x differs considerably between the cases $\eta = 0$ and $\eta \neq 0$, respectively. However, if $T = 0$

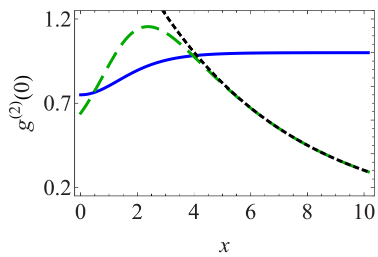

FIG. 1. Steady-state second-order photon-photon correlation function $g^{(2)}(0)$ as a function of $x = \hbar\omega_0/(k_B T)$ for a two-atom system, $N = 2$. Here the solid line is plotted for a negligible dipole-dipole-coupling strength, i.e., $\delta/\omega_0 \to 0$, whereas the dashed one stands for $\delta/\omega_0 = 0.1$. The short-dashed curve is plotted based on Eq. (18), which is valid for $x \gg 1$.

then $\{G_1(\vec{R}), G_2(\vec{R})\} = 0$ and the normalized second-order correlation function $g^{(2)}(0)$ loses its meaning.

Figures 1 and 2 depict the photon statistics of the spontaneously scattered photons by a dipole-dipole interacting few-atom system, i.e., for $N = 2$ and $N = 3$, respectively, via their environmental thermostat; see also Appendix B. Particularly, the solid lines are plotted for a negligible dipole-dipole-coupling strength, i.e., $\delta/\omega_0 \to 0$, whereas the dashed ones stand for $\delta/\omega_0 = 0.1$. The short-dashed curves represent Eq. (18), which is valid for $x \gg 1$. Interestingly, the photon statistics turns into a quantum photon statistics, i.e., $g^{(2)}(0) < 1$, when $\eta \neq 0$ and for lower temperatures, that is, $x \gg 1$. This behavior is typical for several dipole-dipole interacting atoms; see also [57,58]. From this reason, in Fig. 3 we plot the steady-state second-order photon-photon correlation function $g^{(2)}(0)$ for $N = 7$ dipole-dipole interacting atoms which supports it. Thus the thermal mediated dipole-dipole interactions among two-level emitters in a several-atom Dicke-like sample are responsible for the sub-Poissonian photon statistics of the spontaneously scattered photons. In the absence of the dipole-dipole interactions, that is, when $\eta \to 0$, sub-Poissonian photon statistics occurs in this system only for $N \in \{1, 2, 3\}$ and for higher environmental temperatures, i.e., $x \ll 1$; see Figs. 1 and 2. Also, one can observe a nice concordance with the limiting cases of the second-order photon-photon correlation function $g^{(2)}(0)$, given by Eqs. (16)–(18) and the plots in Figs. 1–3.

While this tendency, i.e., the occurrence of sub-Poissonian photon statistics for bigger values of $x = \hbar\omega_0/(k_B T)$, persists for larger atomic ensembles, it will be less useful since the photon flux might be too weak in this case. To

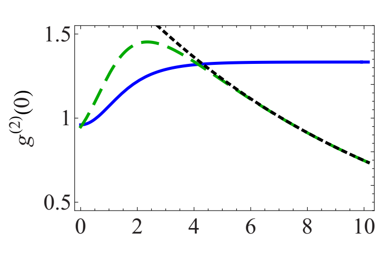

FIG. 2. Same as in Fig. 1, but for $N = 3$.

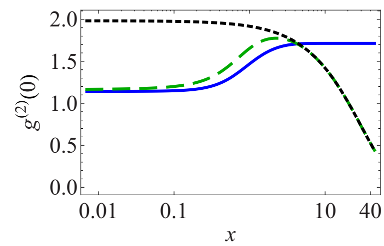

FIG. 3. Log-linear plot of the steady-state second-order photon-photon correlation function $g^{(2)}(0)$ as a function of $x = \hbar\omega_0/(k_B T)$, for $N = 7$. Other involved parameters are as in Fig. 1.

clarify this issue, one represents the cooperative intensity of spontaneously scattered EMF by the dipole-dipole interacting two-level emitters in a weaker thermal environment, i.e., for $x \gg 1$, as follows:

$$G_1(\vec{R})/\Phi(R) = N(1-\eta)^4 \exp\left(-x(1-\eta)\right). \tag{19}$$

On the other hand, from Eq. (18) when setting $g^{(2)}(0) = 1$, one can obtain the η's ranges in order for the sub-Poissonian photon statistics to occur for $N \gg 1$ and $x \gg 1$, namely,

$$\eta > (N/x)\ln(\sqrt{2}). \tag{20}$$

Note here that $\eta = (N/x)\ln(\sqrt{2})$ is the condition for Poissonian, whereas $\eta < (N/x)\ln(\sqrt{2})$ is for super-Poissonian photon statistics, respectively. Although Eq. (20) is not difficult to fulfill, the photon intensity, given by Eq. (19), will be too low for larger ensembles and weaker thermal baths, $x \gg 1$. The reason consists in the Bose-Einstein nature of the atomic statistics, meaning that in a thermal environment at equilibrium, the two-level emitters tend to reside in their ground state for $\{N, x\} \gg 1$. Under these circumstances, less atoms get excited and, therefore, the spontaneously generated EMF might be weak, as well. Finally, the photon statistics changes from sub-Poissonian to super-Poissonian if $\delta \to -\delta$ and $x \gg 1$.

Note that from Eq. (19) follows that for weaker thermal baths when $x \gg 1$, but still $T \neq 0$, $G_1(\vec{R}, \eta \neq 0) > G_1(\vec{R}, \eta = 0)$; see Fig. 4. However, from the general expression of the intensity, i.e., Eq. (13), one has that $G_1(\vec{R}, \eta \neq 0) < G_1(\vec{R}, \eta = 0)$, within moderately weak, $x > 1$, to moderately intense, $x \leqslant 1$, heat baths, respectively. This will turn again to $G_1(\vec{R}, \eta \neq 0) > G_1(\vec{R}, \eta = 0)$ for stronger thermal baths, i.e., when $x \to 0$; see Fig. 4. Particularly, for higher

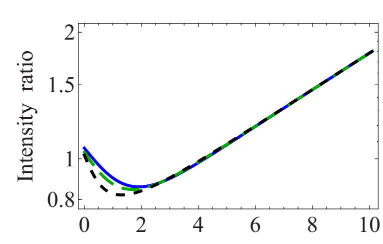

FIG. 4. Log plot of the ratio $G_1(\vec{R}, \eta \neq 0)/G_1(\vec{R}, \eta = 0)$ as a function of $x = \hbar\omega_0/(k_B T)$ for $\eta = 0.1$. The solid line is plotted for $N = 2$ and the dashed one for $N = 3$, while the short-dashed curve is for $N = 7$, respectively.

environmental temperatures when $x \to 0$, one has that

$$\frac{G_1(\vec{R}, \eta \neq 0)}{G_1(\vec{R}, \eta = 0)} = 1 + 72\eta^2 + 2064\eta^4/7. \quad (21)$$

Generalizing, the thermal mediated dipole-dipole interactions are responsible for sub-Poissonian statistics of the spontaneously scattered photons by a small two-level ensemble, in a weak thermal bath and within the Dicke limit. Furthermore, due to the presence of the dipole-dipole interactions between the two-level emitters, the collectively scattered spontaneous emission light suppresses or enhances depending on the intensities of the environmental thermal baths; see also [59]. These behaviors can be understood if one uses the symmetrical cooperative multiatom Dicke states [1,4]. The symmetric atomic Dicke states shift, because of the dipole-dipole interaction among the two-level emitters, leading to different collective populations of these states in a thermal environment; see Appendix B demonstrating this particularly for a $N = 2$ atomic sample.

In principle, the relationship among the quantum nature of the spontaneously scattered photons in this process and the ensemble's quantum thermodynamics would be an interesting issue to address in future investigations.

V. SUMMARY

We have investigated the steady-state quantum dynamics of an ensemble of dipole-dipole interacting two-level emitters, in the Dicke limit, which is mediated by the environmental thermostat. Particularly, the focus was on the quantum nature of the collective spontaneously scattered photons in this process. We have demonstrated that the dipole-dipole interaction among the two-level emitters is responsible for sub-Poissonian photon statistics scattered by a small ensemble consisting from few to several atoms, respectively. The quantum light effect occurs when the energy of a free atom is larger than the corresponding one due to the thermal reservoir. This quantum feature survives for larger ensembles; however, the photon intensity might be too weak in this case. Without the dipole-dipole interactions among the two-level radiators, the sub-Poissonian statistics occurs for few atoms and stronger thermal baths only. The collectively scattered spontaneous emission light intensity suppresses or enhances depending on the intensities of the environmental thermal baths.

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APPENDIX A: MASTER EQUATION FOR WEAK DIPOLE-DIPOLE INTERACTING TWO-LEVEL EMITTERS, i.e., $\delta/\omega_0 \to 0$

For the sake of comparison, let us discuss here the situation of weak dipole-dipole interaction strengths, compared to the transition frequency. The Hamiltonian describing N two-level emitters in a thermal bath is then $H = H_F + H_A + H_{AF}$, where

$$H_F = \sum_k \hbar\omega_k a_k^\dagger a_k \quad (A1)$$

is the free energy of the thermal reservoir, while

$$H_A = \sum_{j=1}^{N} \hbar\omega_0 S_{zj} \quad (A2)$$

is the corresponding free energy of the atomic subsystem. The interaction among the two subsystems, assumed weak, is given by the following Hamiltonian:

$$H_{AF} = i\sum_k \sum_{j=1}^{N} (\vec{g}_k \cdot \vec{d}_j)(a_k^\dagger S_j^- e^{-i\vec{k}\cdot\vec{r}_j} - \text{H.c.}), \quad (A3)$$

where all the involved parameters or operators have the usual meaning. Eliminating the thermal reservoir degrees of freedom, see, e.g., Ref. [6], in the usual way and in the Born-Markov approximations, one arrives at the following master equation describing the two-level ensemble in a thermal heat bath:

$$\dot{\rho}(t) - i\sum_{j\neq l}^{N} \delta_{ij}[S_j^+ S_l^-, \rho] = -\sum_{j,l=1}^{N} \chi_{jl}\{(1 + \bar{n}(\omega_0))[S_j^+, S_l^- \rho] + \bar{n}(\omega_0)[S_j^-, S_l^+ \rho]\} + \text{H.c.}, \quad (A4)$$

where

$$\delta_{jl} = \frac{3\Gamma(\omega_0)}{4}\bigg\{[\cos^2\xi_{jl} - 1]\frac{\cos(\omega_0 r_{jl}/c)}{(\omega_0 r_{jl}/c)} + [1 - 3\cos^2\xi_{jl}]\bigg[\frac{\sin(\omega_0 r_{jl}/c)}{(\omega_0 r_{jl}/c)^2} + \frac{\cos(\omega_0 r_{jl}/c)}{(\omega_0 r_{jl}/c)^3}\bigg]\bigg\}$$

are the vacuum-induced dipole-dipole interactions among any two atoms, separated by $r_{jl} = |\vec{r}_j - \vec{r}_l|$, whereas

$$\chi_{jl} = \frac{3\Gamma(\omega_0)}{2}\bigg\{[1 - \cos^2\xi_{jl}]\frac{\sin(\omega_0 r_{jl}/c)}{(\omega_0 r_{jl}/c)} + [1 - 3\cos^2\xi_{jl}]\bigg[\frac{\cos(\omega_0 r_{jl}/c)}{(\omega_0 r_{jl}/c)^2} - \frac{\sin(\omega_0 r_{jl}/c)}{(\omega_0 r_{jl}/c)^3}\bigg]\bigg\}$$

are the corresponding incoherent couplings contributing to the spontaneous decay [6]. Here, ξ_{jl} are the angles between the dipole moments $\vec{d}$, assumed identical for all atoms and parallel, and $\vec{r}_{jl}$.

For small interparticle separations compared to the emission wavelength, i.e., $\omega_0 r_{jl}/c \to 0$ in the Dicke limit, one has that $\chi_{jl} \to \Gamma(\omega_0)/2$, while δ_{jl} reduces to the static dipole-dipole interaction potential, i.e., $\delta_{jl} \to 3\Gamma(\omega_0)/[4(\omega_0 r_{jl}/c)^3]$, and $j \neq l$. Now introducing the collective atomic operators and considering that the dipole-dipole interactions are equal for any atomic pair, we obtain the corresponding master equation given at the end of Sec. II. Notice here that although $\delta/\omega_0 \to 0$, the dipole-dipole-coupling strength δ can be smaller and of the same order or larger than the single-emitter spontaneous decay rate Γ; however, also $\Gamma/\omega_0 \to 0$.

Actually, the above master equation (A4) given in the Dicke limit is valid only if the static dipole-dipole-coupling strength δ, or generally the dipole-dipole interaction, is negligible compared to the transition frequency ω_0, that is, $\delta/\omega_0 \to 0$. This is because we have obtained the corresponding expression for the dipole-dipole interactions δ_{jl} assuming *weak* interaction among the atomic and thermal reservoir subsystems, respectively, that is, one cannot get something stronger than the free energy of the atomic subsystem given by H_A. Therefore, for stronger dipole-dipole interactions δ, but still $\delta/\omega_0 < 1$ which is the case considered here, the static dipole-dipole interaction potential should be included in the Hamiltonian H_A, namely,

$$H_A = \sum_{j=1}^{N} \hbar\omega_0 S_{zj} - \hbar\delta \sum_{j\neq l} S_j^+ S_l^-, \tag{A5}$$

and eliminate the thermal bath degrees of freedom as it is described in Sec. II. The positions r_j of the atoms are excluded in the Dicke limit since the stronger contribution to the dipole-dipole interaction comes from the static dipole-dipole part which is already included in H_A; see also the Hamiltonian (1), that is, $H_0 = H_F + H_A$.

APPENDIX B: TWO DIPOLE-DIPOLE INTERACTING ATOMS IN A THERMAL BATH

Here we shall discuss the two-atom cooperative dynamics in a thermal bath using the Dicke collective states for $N = 2$. Introducing the collective two-atom Dicke states [2]

$$|E\rangle = |e_1 e_2\rangle,$$
$$|S\rangle = (|e_1 g_2\rangle + |e_2 g_1\rangle)/\sqrt{2},$$
$$|A\rangle = (|e_1 g_2\rangle - |e_2 g_1\rangle)/\sqrt{2},$$
$$|G\rangle = |g_1 g_2\rangle,$$

and taking into account that

$$S_1^+ = \left(R_{ES} - R_{EA} + R_{SG} + R_{AG}\right)/\sqrt{2}$$

and

$$S_2^+ = (R_{ES} + R_{EA} + R_{SG} - R_{AG})/\sqrt{2},$$

one obtains, from (1) and (2), the following Hamiltonians in the cooperative two-atom bases:

$$H_0 = \sum_k \hbar\omega_k a_k^\dagger a_k + \hbar\omega_0(R_{EE} - R_{GG}) - \hbar\delta R_{SS},$$
$$H_i = i\sqrt{2}\sum_k (\vec{g}_k \cdot \vec{d})((R_{ES} + R_{SG})a_k - \text{H.c.}), \tag{B1}$$

where the atomic operators acting in the two-atom collective bases are defined as follows: $R_{\alpha\beta} = |\alpha\rangle\langle\beta|$ and satisfying the commutation relations $[R_{\alpha\beta}, R_{\beta'\alpha'}] = R_{\alpha\alpha'}\delta_{\beta\beta'} - R_{\beta'\beta}\delta_{\alpha'\alpha}$ and $\{\alpha, \beta \in E, S, G\}$. The antisymmetrical state $|A\rangle$ was dropped since it does not couple to the environmental electromagnetic field reservoir.

Introducing the Hamiltonian H_i, but in the interaction picture, in Eq. (4) and performing the trace over the thermal EMF degrees of freedom, one arrives at the following master equation describing the two dipole-dipole interacting atoms in a thermal environment, namely,

$$\begin{aligned}
&\frac{d}{dt}\rho(t) + i[\omega_0(R_{EE} - R_{GG}) - \delta R_{SS}, \rho] \\
&\quad = -\gamma^{(+)}(1 + \bar{n}^{(+)})([R_{ES}, R_{SE}\rho] + [R_{SG}, R_{SE}\rho]) \\
&\qquad - \gamma^{(-)}(1 + \bar{n}^{(-)})([R_{SG}, R_{GS}\rho] + [R_{ES}, R_{GS}\rho]) \\
&\qquad - \gamma^{(+)}\bar{n}^{(+)}([R_{SE}, R_{ES}\rho] + [R_{GS}, R_{ES}\rho]) \\
&\qquad - \gamma^{(-)}\bar{n}^{(-)}([R_{GS}, R_{SG}\rho] + [R_{SE}, R_{SG}\rho]) + \text{H.c.},
\end{aligned} \tag{B2}$$

where

$$\bar{n}^{(\pm)} = \bar{n}(\omega_0 \pm \delta), \quad \gamma^{(\pm)} = \Gamma(\omega_0 \pm \delta)$$

are given at the corresponding transition in the two-atom basis; see also [49]. Notice here that Eq. (B2) implies that $\omega_0 \gg \delta$, meaning that the corrections to the obtained results are of the order of $(\delta/\omega_0)^2$. Now, one can obtain the equations of motion for the variables of interest. For instance, the populations in the two-atom collective states are given by the following equations:

$$\begin{aligned}
\frac{d}{dt}\langle R_{EE}\rangle &= -2\gamma^{(+)}(1 + \bar{n}^{(+)})\langle R_{EE}\rangle + 2\gamma^{(+)}\bar{n}^{(+)}\langle R_{SS}\rangle, \\
\frac{d}{dt}\langle R_{GG}\rangle &= 2\gamma^{(-)}(1 + \bar{n}^{(-)})\langle R_{SS}\rangle - 2\gamma^{(-)}\bar{n}^{(-)}\langle R_{GG}\rangle,
\end{aligned} \tag{B3}$$

with

$$\langle R_{EE}(t)\rangle + \langle R_{SS}(t)\rangle + \langle R_{GG}(t)\rangle = 1.$$

Their steady-state solutions are, respectively,

$$\begin{aligned}
\langle R_{EE}\rangle_s &= \frac{\mu^{(-)}\mu^{(+)}}{1 + \mu^{(-)}(1 + \mu^{(+)})}, \\
\langle R_{SS}\rangle_s &= \frac{\mu^{(-)}}{1 + \mu^{(-)}(1 + \mu^{(+)})}, \\
\langle R_{GG}\rangle_s &= \frac{1}{1 + \mu^{(-)}(1 + \mu^{(+)})},
\end{aligned} \tag{B4}$$

where $\mu^{(\pm)} = \exp[-x(1 \pm \eta)]$, and recalling that $x = \hbar\omega_0/k_B T$ and $\eta = \delta/\omega_0$ with $\eta < 1$.

For a dipole-dipole interacting two-atom sample, the steady-state second-order correlation function, $g^{(2)}(0)$, is represented as follows:

$$g^{(2)}(0) = \frac{(1 - \eta^2)^4\langle R_{EE}\rangle_s}{((1 + \eta)^4\langle R_{EE}\rangle_s + (1 - \eta)^4\langle R_{SS}\rangle_s)^2}. \tag{B5}$$

This expression is equivalent with the one which would be obtained from Eq. (15), with the help of the solution (9) when taking $N = 2$. Because of strong dipole-dipole interactions among the two-level emitters one has that $\langle R_{SS}(\delta \neq 0)\rangle_s \neq \langle R_{SS}(\delta = 0)\rangle_s$ for $\eta < 1$ and x varying from zero up to several units, whereas $\langle R_{EE}(\delta \neq 0)\rangle_s \approx \langle R_{EE}(\delta = 0)\rangle_s$. Actually, the transition frequencies among the two-atom collective states would modify in the presence of dipole-dipole interaction among the two-level emitters, namely, $(\omega_0 - \delta)$ is the frequency of the $|G\rangle \leftrightarrow |S\rangle$ transition, whereas $(\omega_0 + \delta)$ is the frequency of the $|S\rangle \leftrightarrow |E\rangle$ two-atom transition, respectively. The mean thermal photon numbers corresponding to these frequencies, i.e., $\bar{n}^{(\pm)}$, are different as well in the presence of dipole-dipole interaction. That is why the population on the symmetrical state $|S\rangle$ differs, while on the most excited state $|E\rangle$ is almost the same, regardless of the dipole-dipole-coupling strength δ. This will lead to behaviors shown in Fig. 1 for $g^{(2)}(0)_{\delta\neq 0}$ and $g^{(2)}(0)_{\delta=0}$, respectively. If the dipole-dipole-coupling strength δ changes the sign, i.e., $\delta \to -\delta$, the photon statistics modifies too, that is, from sub-Poissonian to super-Poissonian photon statistics. Again, this happens due to the change of the transition frequencies among the two-atom cooperative states.

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