Letter

Quantum emission of light with densely packed driven dipoles

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Collective excitations in regular structures of strongly interacting driven dipoles have been found to lead to strongly antibunched statistics of the emitted photons [$g^{(2)}(0) \ll 0.5$], but precise control of the number of emitters and their spatial distribution is in general a complex experimental task. We overcome this limitation by showing quantum light emission from densely packed driven dipoles does not require their arrangement in any specific structure, pointing strongly towards possible single-photon sources based on hot thermal vapors confined within nanocells, or on irregular quantum dot ensembles.

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Collective coupling between dipoles in (for example) atomic arrays can dramatically modify the optical response of a medium [1,2]. This can respond cooperatively to resonant incident light, leading to modified decay rates, superradiance [3–6], and subradiance [7–13]. Such cooperative optical effects can be understood as the result of dipole-dipole interactions and coherent scattering of photons between quantum emitters [14,15]. Moreover, these collective effects can affect the photon statistics of the emitted light [16–21]. We refer to the case where the photon statistics correspond to the second-order correlation function $g^{(2)}(t) < 1$ as photon antibunching (or the emission of individual photons), and where $1 < g^{(2)}(t) < 2$ as photon bunching (simultaneous detection of multiple photons). The two contrasting regimes of photon superbunching and antibunching have attracted significant attention in recent years for numerous applications [22–24], in addition to their fundamental interest. Another exciting feature is that although a single emitter can exhibit only antibunching, independent of the angle of observation [25–27], for two emitters, the photon statistics depend on several parameters, the direction of observation included [28–30]. Antibunching and single-photon sources are crucial for quantum technologies, with applications in quantum computation, simulation, and sensing [31–33]. They have been demonstrated using cold atoms, ions, and molecules [34–36], as well as with thermal vapors [37,38]. Casimir dispersion forces have also been explored in order to control bunching and antibunching of photons from quantum emitters near surfaces [39,40].

Recent theoretical work shows that strongly antibunched photon statistics [$g^{(2)}(0) \approx 10^{-2}$] should arise from regular arrays of driven dipoles with sufficiently short interparticle distances [41], within a variety of proposed experimental configurations. Achieving a regular array when the emitters must be separated by distances significantly less than the transition wavelength is in general challenging. Atomic and optical physics has witnessed impressive advances in trapping and controlling quantum emitters under a variety of conditions, in particular trapped cold atoms in optical lattices [42–45]; the precise control and well-understood interactions offer unprecedented and exciting opportunities to study many-particle physics [46,47], and a potentially valuable resource for quantum information applications [48,49]. Unit filling and the controlled loading of individual atoms into lattices remain experimentally challenging to achieve, although defect-free arrays in one [50] and two dimensions [51] have been successfully assembled. Assemblies of carefully structured molecular aggregates have also attracted significant interest, as their cooperative behavior can be used to alter the properties of optoelectronic devices [52]. It is also possible to achieve interparticle separations of a few Å in quantum dot (QD) superlattices [53], upon which dipole-dipole interactions lead to energy transfer between neighboring QDs [54,55]. These can be grown in 1D, 2D, and 3D structures [56]; the significant complexity of assembly, however, can easily lead to lattice irregularities and defects.

In this Letter we will show that such structural regularity is unnecessary, eliminating the need for many experimental complexities. Recent experimental and theoretical progress in understanding the properties of light propagating through thin vapor nanocells, for example, has revealed their potential in a number of quantum technology applications, including as single-photon sources [37,38,57]. This ongoing interest has led to the development of a variety of cell designs, and it is possible to create etched two-dimensional and one-dimensional nanochannels [58], where thermal vapor placed in such etched arrays will typically have more than one atom per site ($N \gg 1$). We use a model developed to treat the collective scattering of coherent light from a thermal vapor [59] to theoretically study the photon statistics of the emitted light for ensembles of dipoles in different spatial configurations, including regular or random two-dimensional arrangements (see Fig. 1). Measuring the collective light field characterizes the quantum state of the emitters in a way that is nondestructive with respect to their spatial configuration. We assume

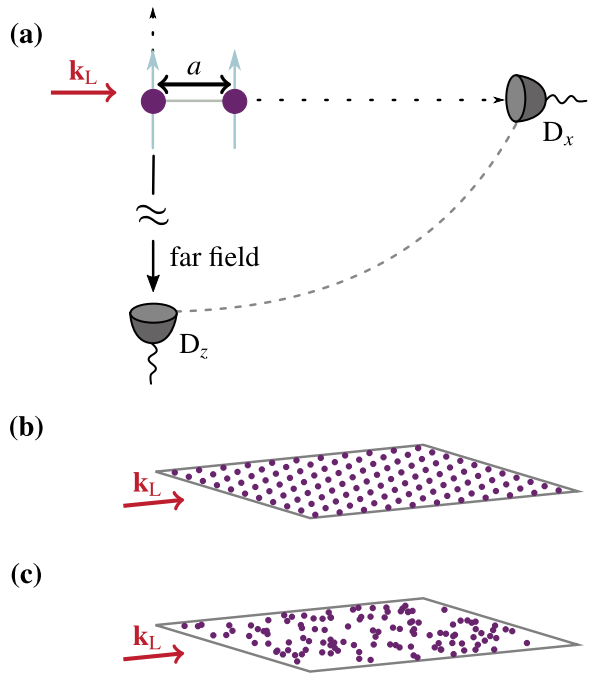

FIG. 1. Scheme depicting the three cases we consider (not to scale). (a) Two identical emitters aligned on the x axis and separated by a distance a. These interact via the dipole-dipole interaction. We consider photon statistics, measured in the far-field zone of the emitted radiation, at two distinct positions, D_x and D_z. (b) Many emitters, confined to a tightly packed regular two-dimensional (2D) lattice in the xy plane, and in (c) we consider a system with randomly located dipoles having the same overall density. The bold (red) arrows represent running laser fields with wave vector $\mathbf{k}_L = (k_L,\ 0,\ 0)$ that drive the dipoles, which we consider to be oriented in the z direction.

throughout that a coherent running laser field drives all the dipoles in the system, and the emitters interact with each other via dipole-dipole interactions. Hence, we show that, for larger numbers of emitters ($N \gg 2$), to achieve low values of $g^{(2)}(0)$ we require only that the emitters are densely packed, and discuss possible realizations within thin vapor nanocells and quantum dot assemblages.

We first consider two identical two-level systems (TLS) at fixed positions $\mathbf{r}_1$ and $\mathbf{r}_2$, with dipole moment $\mathbf{d}_{eg}$, for ground $|g\rangle_j$ and excited states $|e\rangle_j$, transition frequency ω_0, and where $j \in \{1, 2\}$. We drive both dipoles by an external laser field with wave vector $\mathbf{k}_L$. We assume the only dissipative terms are due to spontaneous decays from $|e\rangle_j$. Following Refs. [60,61], the system reduced dipole density operator ρ evolves, in the laboratory frame, according to the master equation

$$\begin{aligned}\frac{\partial \rho}{\partial t} &= -i\omega_0 \sum_{j=1}^{2} \left[\hat{\sigma}_j^z, \rho\right] - \frac{i}{2} \sum_{j\neq l} g_{jl}[\hat{\sigma}_j^+ \hat{\sigma}_l^- + \text{ H.c.}, \rho] \\ &+ \frac{i}{2} \sum_{j=1}^{2} [\Omega_j \hat{\sigma}_j^+ \exp(i\omega_L t) + \text{H.c.}, \rho] \\ &- \sum_{j,l=1}^{2} \gamma_{jl}(\hat{\sigma}_j^+ \hat{\sigma}_l^- \rho + \rho \hat{\sigma}_j^+ \hat{\sigma}_l^- - 2\hat{\sigma}_l^- \rho \hat{\sigma}_j^+), \end{aligned} \tag{1}$$

where $\hat{\sigma}_j^+ = |e\rangle_{jj}\langle g|$ and $\hat{\sigma}_j^- = |g\rangle_{jj}\langle e|$ are the usual raising and lowering operators for the jth emitter, and $2\gamma_{jj} = 2\Gamma$ is the Einstein A coefficient for spontaneous emission from a single dipole. The collective parameters γ_{jl} ($j \neq l$) and g_{jl} describe the damping rate and the dipole-dipole coupling arising from the mutual influence of the emitters via the electromagnetic field [59]. In a running-wave laser field we write $\Omega_j = \Omega_R \exp(-i\,\mathbf{k}_L \cdot \mathbf{r}_j)$, where Ω_R is the maximum Rabi frequency. We use Eq. (1) to study the system dynamics of the system by determining correlation functions for the operators $\hat{\sigma}_j^{\pm}$ ($j = 1, 2$), via the identity $\langle \hat{Q} \rangle = \text{Tr}_S\{\rho\, \hat{Q}\}$ for any operator $\hat{Q}$. For the two-dipole configuration, we obtain a closed system of 15 first-order differential equations of motion for the expectation values of the operators and correlations [59,60], and solve these for the steady state by assuming $(\hat{\sigma}_j^{\pm})^2 = 0$ and noting that operators for different emitters commute at the same time. In general we wish to study the second-order correlation function of fluorescence photons emitted by an ensemble of $N \gg 2$ dipoles and detected by a single photodetector at a point $\mathbf{R}$ in the far-field zone of the radiation emitted by the emitter system [61],

$$g^{(2)}(\mathbf{R}; t, t+\tau) = \frac{G^{(2)}(\mathbf{R}; t, t+\tau)}{G^{(1)}(\mathbf{R}, t) G^{(1)}(\mathbf{R}, t+\tau)}, \tag{2}$$

where for N emitters,

$$\frac{G^{(1)}(\mathbf{R}, t)}{f(\mathbf{R})} = \sum_{j,l}^{N} \langle \hat{\sigma}_j^+(t) \hat{\sigma}_l^-(t) \rangle e^{ik\hat{\mathbf{R}} \cdot \mathbf{r}_{jl}}, \tag{3}$$

$$\frac{G^{(2)}(\mathbf{R}, t)}{f^2(\mathbf{R})} = \sum_{j,l,m,n}^{N} \langle \hat{\sigma}_j^+(0) \hat{\sigma}_m^+(t) \hat{\sigma}_n^-(t) \hat{\sigma}_l^-(0) \rangle e^{ik[\hat{\mathbf{R}} \cdot (\mathbf{r}_{jl} + \mathbf{r}_{mn})]}. \tag{4}$$

Solving the exact dynamics of the system rapidly becomes very complex, as the dimensions of the density matrix ρ grow as $2^N \times 2^N$. Following the procedure in Ref. [59], we use Eq. (1) to solve the dynamics for random pairs of emitters and average the photon statistics of multiple different random pairs. In the limit of many emitters, the single-dipole contributions to the photon statistics become insignificant. To prevent overcounting of these contributions, we neglect single-dipole contributions from Eqs. (3) and (4). Furthermore, contributions of the type $\langle \sigma_j^+(0)\, \sigma_m^+(t)\, \sigma_n^-(t)\, \sigma_j^-(0) \rangle$ (and various permutations) drop out upon time averaging [62], leaving only terms with phase equal either to zero or to $\pm 2ik(\hat{\mathbf{R}} \cdot \mathbf{r}_{jl})$, where $j \neq l$.

We always have antibunching from a single TLS; upon emission of a photon the TLS returns to the ground state, and must again be excited before the next emission. With two noninteracting TLS, one is in its ground state after an emission while the other is unaffected, giving a nonzero probability for two photons to be observed simultaneously, i.e., $g^{(2)}(0) = 1/2$ [63]. Introducing dipole-dipole interactions between two TLS means the photon-emission statistics depend on separation, driving frequency, detuning, and direction of observation. For separations below $\lambda/4$ ($\lambda = 2\pi/|\mathbf{k}_L|$ is the laser wavelength) it becomes impossible to ignore this interaction, and the dipole-dipole parameter $g_{jl} \propto (kr_{jl})^{-3}$ tends to infinity as the emitters' separation tends to zero (note that retardation effects,

which may in principle become important for $r_{jl} > \lambda/2$, are not accounted for in our treatment).

We align the dipoles, assuming the orientation of the dipole oscillations to be along the z axis, and that the wave vector of the driving field propagates along the x axis as $\mathbf{k}_L = (k_L, 0, 0)$ (see Fig. 1), and set the laser frequency equal to ω_0. There is nevertheless in general a nonzero detuning, as in a moving dipole's frame of reference the laser light will be Doppler shifted. We present our results for two TLS in Fig. 2, calculated using Eqs. (2)–(4). We calculate the photon statistics for two different detunings ($\Delta_{1,2}/\Gamma = 0$, and 10) and driving frequency $\Omega_R/\Gamma = 15$. In Figs. 2(a), 2(b) we show $g^{(2)}(t)$ for the case of two TLS placed along the x axis with $a/\lambda = 0.3$, for different directions of observation D_x and D_z. We observe periodic oscillations in the photon statistics; these revivals with time are related to the coherent Rabi oscillations that the dipoles undergo, and is in agreement with experimental observation [64]. In Figs. 2(c)–2(h), we have plotted $g^{(2)}(0)$ as a function of emitter separation a/λ for dipoles placed along the x, y, and z axes. When $\Delta_{1,2}/\Gamma = 0$, decreasing the emitter separation (keeping other parameters fixed) leads to a transition from antibunching to bunching, for both weak and strong driving [65]. When the emitters are not in resonance, we observe different behavior in the photon statistics; depending on the system geometry, superbunching and antibunching photon statistics appear for specific values of detuning and direction of observation. This directional effect in the photon statistics can be explained within the framework of the two-dipole system's collective states and is connected with the symmetric and antisymmetric Dicke states [64]. As we wish to study a system of many TLS with random positions, we also show the averaged photon statistics for randomly oriented pairs of TLS in Figs. 2(i), 2(j) placing one dipole at the origin and the other at a distance a/λ in any direction within a half-sphere.

Possible $N \gg 2$ configurations include atomic optical lattices [66], QD superlattices [67], or unstructured mesoscopic systems such as confined hot vapors in nanocells [58], or ensembles of QDs [68], where many emitters are confined close together with no particular arrangement; we will consider TLS ensembles with $N \gg 2$ both in periodic structures and randomly dispersed. Taking the same approach as in Ref. [59], we consider the photon statistics of the ensembles as an average over different dipole pairs. Specifically, we consider an ensemble of $N = 11\times11$ TLS confined to a 2D structure in the xy plane driven by a running laser field (see Fig. 1). The TLS interact via the dipole-dipole interaction, and the dipoles are aligned along the z direction. Such a pairwise approach is clearly a simplification, however, has been demonstrated to be in good agreement with experimental data available for thermal vapors [59,69]. For a perfect lattice the treatment is potentially incomplete, as an assumption that certain phases time average away is not necessarily justified. With increasing size and temperature of a mesoscopic system, realistic accounting of thermal jitter also increasingly justifies this assumption, as we approach a more random distribution of the emitter positions.

At $\Delta_{1,2}/\Gamma = 0$, our simulations show no significant differences between the photon statistics measured from different directions D_x and D_z. Moreover, we have previously shown

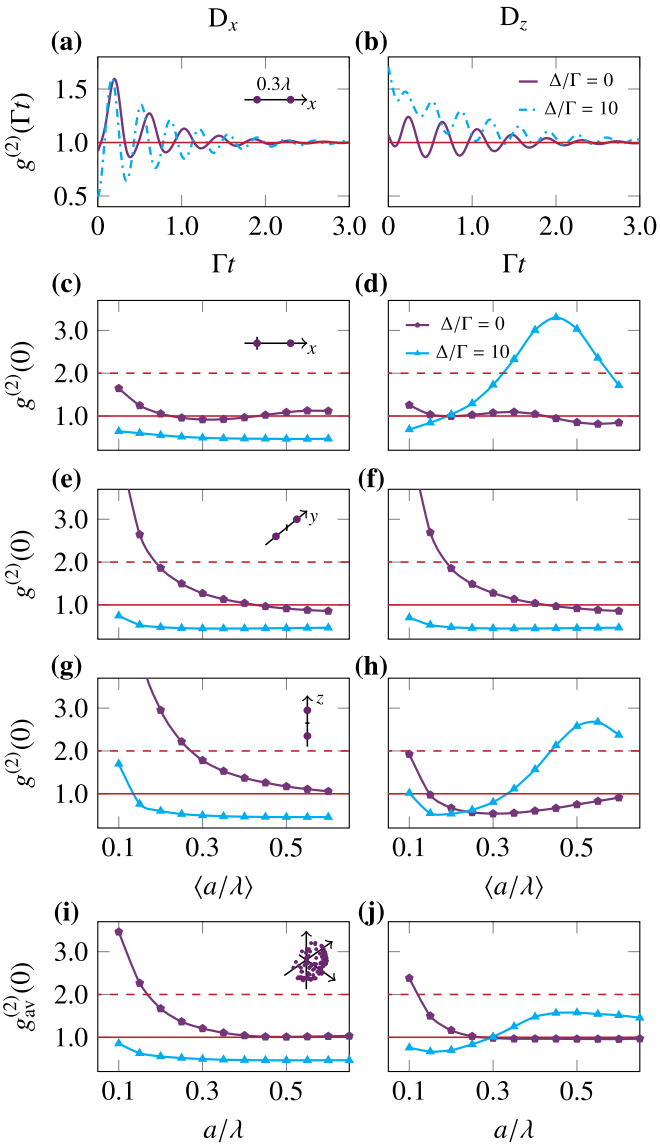

FIG. 2. Photon statistics for two identical dipoles separated by a distance a/λ, where we set the $\Omega_R/\Gamma = 15$. We observe the emitted fluorescence in the D_x (left-hand column) and D_z (right-hand column) directions, as depicted in Fig. 1. (a), (b) Time dependence of $g^{(2)}(t)$ for two dipoles fixed along the x axis for $a/\lambda = 0.3$ and $\Delta_{1,2}/\Gamma = 0$ (solid purple lines) or $\Delta_{1,2}/\Gamma = 10$ (blue dash-dotted lines). (c), (d) $g^{(2)}(0)$ of the same system for varying a/λ. Purple pentagons correspond to $\Delta_{1,2}/\Gamma = 0$ and blue triangles to $\Delta_{1,2}/\Gamma = 10$. We repeat this for dipoles placed (e), (f) along the y axis, and (g), (h) along the z axis. For (a)–(h), we also qualitatively model the effects of such things as thermal jitter by adding a random offset along the relevant axis to the locations of the two dipoles of between 0 and 0.02 λ, and averaging over 100 repetitions. Finally, in (i), (j), we average the photon statistics for different random directions, by locating one dipole at the origin, and the other at a random location on a radius a sphere, averaging over 500 repetitions. We highlight $g^{(2)}(t) = 1$ and $= 2$ by the (red) solid and dashed horizontal lines, respectively. Our units are chosen such that the quantities are scaled by λ or Γ.

[59] that (for $\Delta_{1,2}/\Gamma \neq 0$) larger detunings lead to smaller values of $g^{(2)}(0)$ at larger dipole-dipole separations, but to no significant changes at smaller separations. Furthermore,

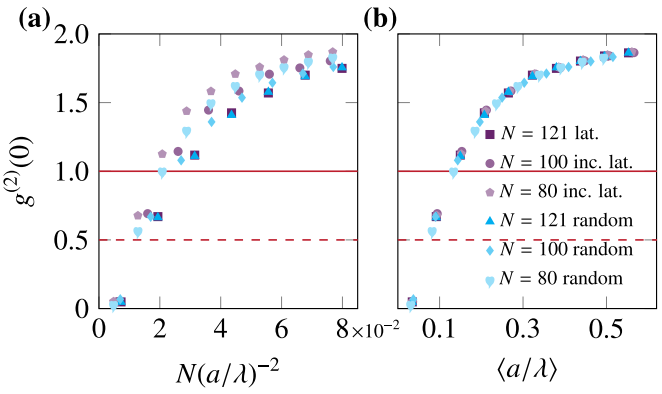

FIG. 3. Dependence of the photon statistics $g^{(2)}(0)$ for N identical emitters confined to the xy plane, (a) as a function of density, and (b) of average separation $\langle a/\lambda \rangle$, for both structured and random configurations. We have set $\Omega_R/\Gamma = 15$ and $\Delta_{1,2}/\Gamma = 0$, and observe the emitted fluorescence in the D_z direction. We arrange $N = 121$ emitters in an 11×11 sites square lattice (squares). We discard emitters at random until $N = 100$ (circles) or 80 (pentagons). We also consider entirely random distributions for $N = 121$ (triangles), 100 (diamonds), and 80 (hearts) emitters. We highlight $g^{(2)}(t) = 1$ and 0.5 by the (red) horizontal solid and dashed lines, respectively. Our units are chosen such that the quantities are scaled by λ or Γ.

we consider that differences in the detuning due to differences in the emitters, such as different sizes in QD ensembles or different particle velocities in a thermal vapor, will only lead to small variations around an average. Many techniques have been developed to make consistently sized quantum dots [55,70,71], as a broader size distribution within optoelectronic devices also leads to undesirable broadening of the energetic emission. Within a thermal vapor one would by default expect a broad distribution of atomic velocities at the envisaged temperatures. At the corresponding envisaged atomic densities, it is necessary to consider the vapor to be confined within a narrow nanocell, as otherwise the sample would be optically thick. Within such narrow nanocells, high-velocity atoms would hit the nanocell walls and not contribute to the measured photon statistics. Hence, the confinement of the thermal vapor effectively selects only a narrow velocity window, i.e., slower atoms [58]. Other possible experimental alternatives to these two platforms, such as single colloidal nanomaterials with multiple emitters (which can also be made to be nearly identical [72]), or molecular ensembles [73], could also have inherent disorder, but if small, these fluctuations in the detuning do not affect our results, as previously shown in Ref. [59]. Our investigations therefore focus on when the laser is in resonance with the dipoles if they are at rest. In Fig. 3, we show our numerical results for $g^{(2)}(0)$, for structured and random spatial distributions, as a function of density and average separation. This we do by starting with $N = 121$ TLS in a square lattice and removing emitters at random until $N = 100$ or 80. We also show results for entirely random distributions of the same particle number and density. We observe that as the average separation increases, the $g^{(2)}(0)$ reaches a plateau (which in general depends on the detuning [59]). For sufficiently small particle separations, we achieve $g^{(2)}(0) < 0.5$, a regime of highly nonclassical photon statistics; i.e., higher densities lead to smaller values of $g^{(2)}(0)$. Note that this differs from what is shown in Figs. 2(i), 2(j) as naive averaging over the directions leads to incorrect results, due to double counting of cases when both photons originate from a single emitter [59]. Moreover, we observe a universal trend line for $g^{(2)}(0)$ as a function of the average TLS separation, which is independent of the precise spatial distribution of the emitters. Note we have not accounted for effects, which may become significant in thermal vapors for very small average separations, such as collisions or atomic motional dephasing; these effects are not an issue for fixed emitters within a matrix, such as QDs.

In conclusion, we find that achieving $g^{(2)}(0) < 0.5$ from an ensemble of emitters appears to be solely due to their being densely packed; there is no requirement for a regular structure of dipoles. We have explored different dimensionalities, achieving the same results with both 2D and 3D ensembles, adding weight to this conclusion. The necessary mean interparticle spacings could be achieved relatively straightforwardly; for example, in a nanocell-confined thermal vapor, such as that described in Ref. [58], at a temperature of 500 K (equivalent to an average distance of $0.1\,\lambda$), compared to current experiments at 450 K. These results apply to ensembles of any TLS, from solid-state systems such as quantum dots to molecular ensembles [74]. As such this is a significant step to the future routine generation of quantum light, and can be straightforwardly confirmed experimentally.

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