Photon liquefaction in time

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We provide a mechanism to imprint local temporal correlations in photon streams which have the same character as spatial correlations in liquids. Usual single-photon emitters correspond, in this picture, to a (temporal) gas while uncorrelated light is the ideal gas. We argue that good single-photon sources are those that exhibit such temporal liquid features, i.e., with a plateau for their short-time correlations (as opposed to a linear dependence) and oscillations at later times, which is a direct manifestation of photon time-ordering. We obtain general, closed-form analytical expressions for the second-order coherence function of a broad family of "liquid light" which can be arbitrarily correlated, though never completely crystallized.

A liquid is a condensed phase of matter whose definition at the microscopic level has been the subject of much debate [1]. It consists of a dense, disordered assembly of molecules exhibiting short-range order in space. Simplest liquids like monatomic argon are well modeled as jumbled closely packed spheres, with an ordering governed by integer multiples of the molecular diameter [2]. The molecular arrangement can be revealed experimentally by diffracting X-rays or neutrons on the fluid [3]. The absence of a Bragg peak indicates that there is no long-range order, but oscillations in the radial diffracted intensities reveal short-range correlations, whereby each molecule is locally attached to a shell of its surrounding neighbors, that remain free to move around and distort, but with more or less probability to be at a given distance. A gas, in contrast, presents no such correlations and has no short-range order. Two molecules still cannot sit at the same position so there remains a depletion of probabilities for close distances, but there are no oscillations. In condensed-matter physics, this is described by the structure factor, whose Fourier transform provides the so-called pair-correlation function $g(r)$ that yields the probability of finding a molecule at a distance r from another molecule, relative to an uncorrelated—i.e., ideal—gas [4]. All these correlations are in space.

Independently from these statistical considerations for correlations of distances between molecules, quantum optics arrived at the modern definition of quantum coherence of light through correlations of photons in time [5]. This relies on the so-called second-order coherence function $g^{(2)}(\tau)$, with a notation eerily reminiscent of the condensed-matter case, although, again, there appears to have no trace of any connection from one field to the other. The $g^{(2)}(\tau)$ function similarly quantifies the den-

sity of two-photons separated by a time delay τ, as compared to an uncorrelated (Poissonian or, as the optical terminology goes, "coherent") photon stream [6].

There are obvious differences between the two cases: fluids are typically three-dimensional and their correlations are in space, while quantum optics treats with one-dimensional photon correlations in time. There are, however, more similarities than seems to have been previously appreciated. In quantum optics, the most studied type of quantum correlations is for single-photon sources [7], with a suppression of two-photon coincidences, i.e., two photons are never detected at exactly the same time. This is not trivial since photons, being bosons, have the natural tendency of exhibiting the opposite behavior of bunching. Some order must be imbued to the photon stream to fight their urge of coming together. The simplest way to achieve this is to recourse to a two-level system σ, put in its excited state at a rate P_σ. If the emitter has a radiative decay rate γ_σ, one finds for its second-order coherence [8]:

$$g^{(2)}(\tau) = 1 - \exp\left(-(P_\sigma + \gamma_\sigma)\tau\right). \quad (1)$$

This has coherence time $P_\sigma + \gamma_\sigma$ with a linear τ short-time loss of coherence from perfect two-photon suppression $g^{(2)}(0) = 0$ to uncorrelated emission $\lim_{\tau\to\infty} g^{(2)}(\tau) = 1$. Another paradigmatic type of excitation is coherent excitation where a classical field (typically a laser) drives resonantly the two-level system, bringing it in another, much richer regime including quantum dressing of the transitions and coherent scattering. In the high-driving regime [9]:

$$g^{(2)}(\tau) = 1 - e^{-\frac{3}{4}\gamma_\sigma\tau}\left[\cosh\left(\frac{\gamma_M\tau}{4}\right) + \frac{3\gamma_\sigma}{\gamma_M}\sinh\left(\frac{\gamma_M\tau}{4}\right)\right] \quad (2)$$

where $\gamma_M \equiv \sqrt{\gamma_\sigma^2 - (8\Omega_\sigma)^2}$ is the Mollow (also known as Rabi) splitting. In the low-driving, so-called Heitler regime, when $\Omega_\sigma \ll \gamma_\sigma$, the two-photon correlations

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takes the simpler form:

$$g^{(2)}(\tau) = \left(1 - \exp(-\gamma_\sigma \tau)\right)^2. \qquad (3)$$

In both cases (Eqs. (2) and (3)), there is a qualitative change of the short-time correlations—where photon suppression occurs—from a linear τ dependence in the incoherent case of Eq. (1) to a quadratic τ^2 one for the coherent cases. Such change from linear to power dependence on time typically reflects a qualitative transformation of the response of a system. For fluids, such dependencies in an energy spectrum are for instance responsible for its diffusive or superfluid character [10].

Although not still compelling at this point of our discussion, we highlight that the two-photon correlation function (1) exhibits oscillations when γ_M becomes imaginary, i.e., when $\Omega_\sigma > \gamma_\sigma/8$, marking the onset of Mollow Physics. In this case, they are understood as Rabi oscillations of the two-level system which gets dressed by the laser [11]. There is then a transition from a monotonous $g^{(2)}(\tau) = 1 - e^{-3\gamma_\sigma \tau/4}(1 + \gamma_\sigma \tau/4)$ at threshold to one featuring all-times oscillations: $g^{(2)}(\tau) = 1 - e^{-3\gamma_\sigma \tau/4}\left(\cos(\Omega_M \tau/4) + \frac{\gamma_M}{\omega_M} \sin(\Omega_M \tau/4)\right)$ with $\Omega_M \equiv \sqrt{(8\Omega_\sigma)^2 - \gamma_\sigma^2}$ a real parameter. The maximum $g^{(2)}(\tau_M) = 1 + \exp(-3\pi\gamma_\sigma/\Omega_M)$ is obtained at $\tau_M = 4\pi/\Omega_M$ and thus is at most 2, in the limit of $\Omega_\sigma \to \infty$. Such oscillations are understood as Rabi oscillations of the populations since in this case $g^{(2)}(\tau) = n_0(\tau)/n_{ss}$ [12] where $n_0(\tau)$ is the dynamics of the system when starting from its ground state and n_{ss} is the steady state population, which is at most $\frac{1}{2}$ for the coherently driven system since stimulated emission prevents population inversion.

While such a Rabi interpretation is completely valid [13–15], in the following, we shall argue that such familiar oscillations are a particular case of a more general trend, namely, photon liquefaction in time, by what we mean temporal ordering of the photons similar to that in space when a gas becomes liquid. The terminology of "condensation" is more common to describe gas-to-liquid transition, but given the predominance of Bose condensation for bosons, we prefer here to refer to that phenomenon with the alternative denomination of "liquefaction" which, we highlight again, further occurs in time. This approach is motivated by the notion of a "perfect single photon source" [16] understood as a source which suppresses photon coincidences not only at exactly $\tau = 0$, but over a temporal window large enough or robust enough so that a physical detector will be resilient to the unavoidable time uncertainty associated to the photodetection process. Such temporal limitations of physical detectors were first highlighted for single-photon observables by Eberly and Wódkiewicz [17] and later upgraded to multiphoton detection by del Valle *et al.* [18]. For two-photon suppression, this results in photons correlations of the type of Eqs. (2) and (3) to be much more resilient to time-frequency uncertainties and, correspondingly, to provide much better antibunching and less "accidental" coincidences, due to the flatter short-time correlation τ^2 [19]. In Ref. [16], it was shown that in the

mathematical idealization where the correlation is flattened so much as to actually open a non-analytic time-gap, i.e., forbidding completely two photons to be closer than a given time t_G, then oscillations ensue in $g^{(2)}(\tau)$ as a result of time-ordering, thus being a direct counterpart, but in time, of the transition from a gas to a liquid. In fact, for the case of a perfect, rigid time-gap, correlations are precisely those, in space, for a system of hard rods, as was first described by Prins [20] who also was the first to derive the expression to compute diffracted intensities from molecular arrangements.

This mathematically-perfect single photon source [16] was analyzed with no underlying physical mechanism to realize it. Here, we provide a broad class of photon temporal liquids, based on a simple mechanism whereby the excitation undergoes a cascade of transitions between various states before ultimately emitting a photon. This is particularly relevant for solid-state systems [21] where the two-level system is implemented by an artificial atom embedded in an environment which comes with various intermediate states, shell structures, metastable states, etc., which could even be controlled or ultimately engineered [35]. We find the interesting result that even *incoherent* driving, insofar as it involves intermediate steps in the cascade, can feature a two-photon correlation function that corresponds to the liquid phase, with oscillations and a power-law dependence for the short-times correlations. The power is furthermore directly related to the number of cascades, and produces the flat plateau typical of hard-sphere repulsions in condensed matter, as well as the characteristic bunching elbows that mark the onset of local ordering [16].

As opposed to solving a quantum optical master equation—which yields the same result, as we shall show for a particular case—we take a more insightful statistical and condensed-matter-inspired approach based on two-photon correlation functions and their underlying waiting time distributions. The incoherent two-photon emission (1) can be understood as a two-steps process with, first, an underlying, backbone Poisson (uncorrelated) stream of events, corresponding to the incoherent excitation at rate P_σ that brings the system in its excited state. Each such event then draws another Poisson-distributed random number with parameter γ_σ, describing the spontaneous emission (second step) [16]. This is sketched in Fig. 1(a) where the exponential (Poisson) distributions alternate sampling of uncorrelated excitation T_1 and spontaneous emission T_2 times. Since we are only interested in the emitted photons, we can equivalently consider directly the distribution for $T_1 + T_2$, which is given by $\tau \exp(-(P_\sigma + \gamma_\sigma)\tau)$. If the emission involves N steps, first with parameter P_σ and each subsequent one with parameter γ_i ($2 \leq i \leq N$), then one needs to similarly replace the exponential decay by the distribution for the sum of N independent exponential random variables, which is one of the phase-type class of distributions, known as the Hypoexponential distribution with N parameters. When those are all equal, the distribution
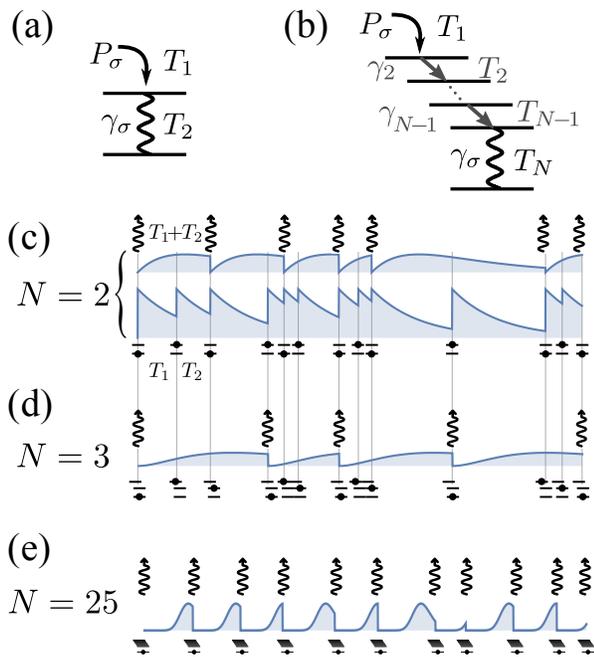
FIG. 1. Single-photon emission from (a) a two-level system under incoherent pumping P_σ and decay γ_σ and (b) with a prior cascade of intermediate transitions. (c) Photon emissions from successive random processes of excitation and emission, described by exponential distributions or directly with the corresponding Erlang distributions for two events. (d) Case with $N = 3$ featuring one intermediate cascade and the corresponding Erlang distributions. (e) Case with $N = 25$ (with a rescaled time axis) with the corresponding Erlang distributions and only the final states (emission) of the cascade depicted. The photon stream is neatly ordered in time with short-range correlations of a liquid, although there are no interactions and all processes are incoherent.

is more popularly known as the Erlang distribution and we shall focus on this case for conceptual simplicity. The waiting time distribution for N steps (one excitation plus $N - 1$ cascades) can then be simply obtained as:

$$w(\tau) = \frac{\gamma^N \tau^{N-1} e^{-\gamma\tau}}{(N-1)!} \,. \tag{4}$$

Since two-photon correlation functions can be computed from the waiting time distribution $w(\tau)$ by transiting to the Laplace space [23]

$$\tilde{g}^{(2)}(s) \equiv \int_0^\infty g^{(2)}(\tau) e^{-s\tau}\, d\tau = \gamma \frac{\tilde{w}(s)}{1 - \tilde{w}(s)} \,, \tag{5}$$

with $\tilde{w}(s) = \gamma^N / [(s+\gamma)^N - \gamma^N]$ the Laplace transform of Eq. (4), we then find, by inverse Laplace transform:

$$g_N^{(2)}(\tau) = 1 + \sum_{p=1}^{N-1} z_N^p \exp\left(-\gamma(1 - z_N^p)\tau\right) \tag{6}$$

where $z_N \equiv \exp(i2\pi/N)$ are the Nth roots of unity. This succinct general expression can be easily made explicit

for particular cases, e.g., with no cascade ($N = 1$ for the excitation alone), we have an uncorrelated (or coherent) photon stream, Fig. 2(a), while $N = 2$ describes the excitation plus spontaneous emission of Fig. 1(c) and thus recovers Eq. (1). We get new results with two cascades ($N = 3$, Fig. 1(d)), for which Eq. (6) simplifies to

$$g^{(2)}(\tau) = 1 - 2\sin\left(\frac{\sqrt{3}}{2}\gamma\tau + \frac{\pi}{6}\right) e^{-\frac{3}{2}\gamma\tau} \tag{7}$$

and for three cascades ($N = 4$):

$$g^{(2)}(\tau) = 1 - e^{-2\gamma\tau} - 2e^{-\gamma\tau}\sin(\gamma\tau) \tag{8}$$

with the possibility to derive similar closed-form expressions for other N.

These cascaded chains of incoherent relaxation produce, interestingly, two-photon correlation functions that are more like in character those of the coherently driven case (2) than the incoherent case (1). Even with one cascade only, there is some onset of liquefaction with oscillations which, although not compelling numerically, are clear from the analytical expressions in Eq. (7). The maximum $g^{(2)}(\tau_M) = 1 + e^{-\sqrt{3}\pi} \approx 1.0043$ at $\gamma_\sigma\tau_M = 2\pi/\sqrt{3}$, however, is only marginally different from unity. Higher $g^{(2)}$ are obtained for a higher number of cascades but at time delays which are zeros of transcendental equations (for instance, as a solution of $\exp(\tau)\sin(\tau + \pi/4) = c$ for $N = 2$) but these can be easily obtained numerically. Another manifestation of temporal liquefaction is the hardening of the photons that increasingly repulse each other, with short-time expansion from N cascades providing the plateaus:

$$g^{(2)}(\tau) \approx \frac{N^2}{N!}(\gamma\tau)^N \,. \tag{9}$$

This dependency of the short-time correlations mean that a physical detector will be increasingly less affected, at a qualitative level, by the fundamental time uncertainty attached to photodetection. These qualities are directly inherited from the waiting time distribution (4) and the shape of the probability distribution for each photon emission. The flattening of these quantities at short times makes it possible to open an effective temporal gap. This paves the way towards a genuine perfect single-photon source, with no multiphoton emission for detectors with a better temporal resolution than the time gap, just as a superconductor is a genuine perfect conductor, with no loss whatsoever. The peak following the plateau in the emission probability also imprints a clear time-ordering of the photons, as a result of the compound-time averaging out its more extreme fluctuations. Together, these two features produce a regular stream of photons. This is obvious in Fig. 1(e) where photons appear to be equidistantly spaced in time with small only fluctuations, as would be expected from an externally-controlled (pulsed) single-photon source [16]. Such an order is local only,

however, as fluctuations, however small, pile up and eventually wash out correlations for photons distant enough. This thus corresponds to a photon liquid, and never a crystal, as the stream is intrinsically stationary for long-enough times. This is, however, of little concern for the single-photon character.

While these properties are well-understood from such a point-process statistical approach, which is furthermore rooted in statistical arguments that have been thoroughly studied in condensed-matter physics, these results would also be obtained from a quantum treatment, i.e., with a master equation for a multilevel system. The case $N = 3$, for instance, with level structure $|i\rangle$ for $0 \leq i \leq 2$ and Hamiltonian $H \equiv \sum_{i=1,2} \omega_i |i\rangle\langle i|$ can be described by the Lindbladian $\mathscr{L}\rho \equiv = -i[H, \rho] + \left\{ \frac{P_\sigma}{2}\mathscr{L}_{|2\rangle\langle 0|} + \frac{\gamma_1}{2}\mathscr{L}_{|1\rangle\langle 2|} + \frac{\gamma_2}{2}\mathscr{L}_{|0\rangle\langle 1|} \right\}\rho$ where for any operator Ω, $\mathscr{L}_\Omega\rho \equiv 2\Omega\rho\Omega^\dagger - \Omega^\dagger\Omega\rho - \rho\Omega^\dagger\Omega$. One can then compute steady-state two-time correlators like $G_{10}^{(2)}(\tau) \equiv \langle \sigma^\dagger_{10}(0)\sigma^\dagger_{10}(\tau)\sigma_{10}(\tau)\sigma_{10}(0)\rangle$ for $\sigma_{10} \equiv |0\rangle\langle 1|$ from the quantum regression theorem, i.e., $G_{10}^{(2)}(\tau) = \mathrm{Tr}\left(\sigma^\dagger_{10}\sigma_{10}e^{\mathscr{L}\tau}[\sigma_{10}\rho_{\mathrm{ss}}\sigma^\dagger_{10}]\right)$ on the steady-state density matrix which is diagonal:

$$\rho_{\mathrm{ss}} = \frac{1}{\gamma_1\gamma_2 + P_\sigma(\gamma_1 + \gamma_2)} \begin{pmatrix} \gamma_1\gamma_2 & & \\ & P_\sigma\gamma_1 & \\ & & P_\sigma\gamma_2 \end{pmatrix}. \quad (10)$$

The normalization of $G_{10}^{(2)}$ yields $g^2(\tau)$ for this transition, which recovers Eq. (7) for the case $P_\sigma = \gamma_1 = \gamma_2$ that we considered previously.

There have been many studies of single-photon emission both theoretically and experimentally, for which our approach is relevant and that indeed prefigure our small N phenomenology. This includes other full quantum optical treatments (with a master equation) which typically consider three-level systems [24, 25], which however do not report oscillations but merely a multi-exponential return to uncorrelated emission. There is indeed a range of parameters that accommodate oscillations in $g^{(2)}(\tau)$ and our Erlang particular case fulfills them. Such oscillations are conceptually noteworthy as they occur in a completely incoherent system, exposing the time-ordering as the system liquefies. While the parameter range is not particularly strict, it is easy to overlook if one is not aware of such a possibility. Elbows to antibunching have also been observed in various systems, where they are often attributed to intermediate states in a way that is compatible with our mechanism [26–32]. Descriptions have been, however, based on rate equations and the consequences in terms of better single-photon emission have been either ignored or even considered problematic, the case of Eq. (1) [Fig. 2ii] being considered, misguidedly, as an ideal. Multilevel rate equations can provide powerful results, including impressive accounts of $g^{(2)}(\tau)$ over 11 orders of magnitude in time, for systems bathed in highly-complex semiconductor environments featuring a plethora of dark states and fluorescence intermittency [33]. Whether such models
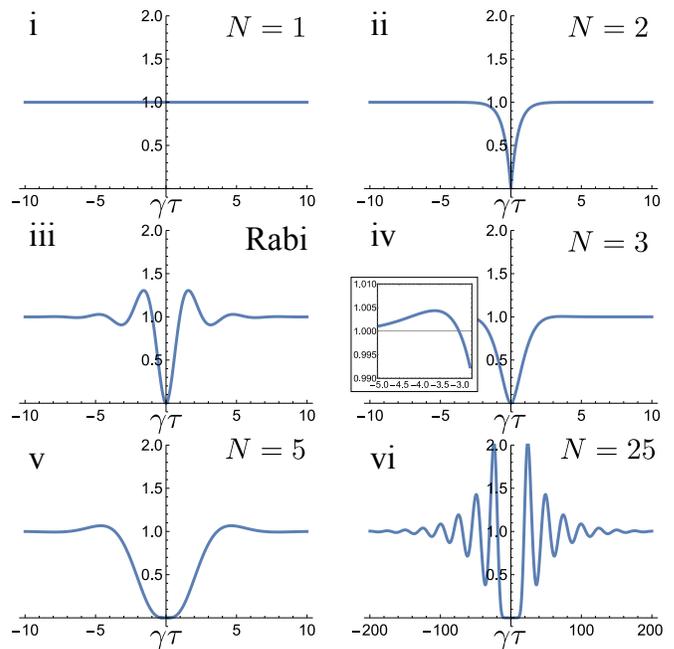
FIG. 2. Two-photon correlation functions for i) coherent light (ideal gas) ii) an incoherently pumped 2LS (gas), iii) a strongly and coherently-driven 2LS (Rabi liquid), iv) an incoherently-driven one-cascade 2LS (onset of liquid with a zoom in inset showing the $g^{(2)} = 1.0043$ bunching), v) liquefaction with 5 cascades and vi) liquid with 25 cascades. Note that the τ axis is rescaled in the latter case as over the range $\pm10/\gamma$ of the other plots, $g^{(2)}$ remains below 2×10^{-3}. We assumes all rates to be equal.

compete or complement our mechanism, which endows photons with qualitatively superior features of single-photon emission, remains to be ascertained. Our approach furthermore focused on the most basic configuration to highlight the conceptual novelty, and should be extended in several ways to tackle realistic experiments. Let alone that the Erlang (degenerate) distribution is a very particular case (with one parameter only whereas one naturally expect various transitions to come with possibly largely varying parameters), the number of steps in the cascade could also be a random variable, and the possibility for various carriers to undergo such cascades simultaneously be included. Our basic picture of liquefaction should however remain and even apply to more general cases, e.g., that of interactions between several emitters, that display curves similar to Eq. (6) [34].

Our findings come with several conclusions. One is that the quest for perfect single-photon sources has been so far driven by technological improvements to reduce $g^{(2)}(0)$ as much as possible from the basic structure of a two-level system. This is a quantitative and asymptotic race that is doomed to imperfection as the limitations are fundamental: photodetection as a physical process will always detect simultaneous photons from a two-level system [19]. To obtain perfect single-photon emission, one must open a gap somewhere [16]. We have

provided a straightforward mechanism, furthermore of relevance in solid-state platforms and that could otherwise be engineered, of a cascade process that results in strong repulsions between photons with the effect of imprinting strong correlations between them, similarly to how interactions order and correlate molecules in a fluid. The number N of cascades rules the magnitude of the effect and we have given a general closed-form analytical expression for all N. A related conclusion is that $g^{(2)}(0)$ itself is not the most relevant measure for two-photon suppression. One must instead consider $g^{(2)}(\tau)$ locally around $\tau = 0$, and consider both the power dependence of the short-time correlations as well as the presence of oscillations or at least bunching (elbows) past the first coherence time, as these mark the onset of short-time photon ordering. This also suggest that considerably more types of quantum lights are awaiting to be discovered and classified through such a perspective. Finally, maybe the most far-reaching suggestion of our approach, is that thermodynamic concepts that are central to describe condensed matter could also provide more systematic and deeper descriptions of quantum light, possibly relying on equations of states to describe the various phases, as opposed to n-th order coherence (almost always truncated to $n = 2$ anyway). Bunching, for instance, might be related to plasma. Intriguingly, this condensed-matter perspective would require to trade space for time.

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