Correlations in Circular Quantum Cascades

Miguel Ángel Palomo Marcos,[1] Eduardo Zubizarreta Casalengua,[2] Elena del Valle,[1,3,4] and Fabrice P. Laussy[5,*]

[1] *Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain*
[2] *Walter Schottky Institute, School of Computation, Information and Technology and MCQST, Technische Universität München, 85748 Garching, Germany*
[3] *Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, 28049 Madrid, Spain*
[4] *Institute for Advanced Study, Technische Universität München, 85748 Garching, Germany*
[5] *Instituto de Ciencia de Materiales de Madrid ICMM-CSIC, 28049 Madrid, Spain*

(Dated: August 6, 2024)

We introduce a one-way, one-quantum cascade, whereby a single excitation proceeds one-directionwise in a ladder of energy levels. This makes a variation from more famous two-way cascades where the excitation can go up and down following its excitation or relaxation in the ladder. We provide closed-form solutions for two-photon correlation functions between any transitions in such circular cascades. We discuss how the rich correlations that result from what appears to be an extremely simple implementation, are essentially those which have been entertained from complex architectures relying on strongly-correlated, many-body physics or cavity QED effects.

arXiv:2408.02068v1 [quant-ph] 4 Aug 2024

I. INTRODUCTION

Cascading is a widespread phenomenon that can amplify features of a system, as is best illustrated by the domino effect. With bosons, it led to the idea of the quantum cascade laser [1], whose realization [2] opened new perspectives and regimes of operation for coherent light [3]. The variation where the active medium itself is also bosonic [4] led to new regimes of superbunched emission [5]. Here, we consider cascades of a single quantum of excitation, down a ladder of N levels. In contrast to the previous cascades, both the active medium and the radiation field thus remain at the level of single quanta. There is a rich variety of platforms where this can take place [6–12] and such cascading is therefore not new, being in fact basically intrinsic to the way optical emitters release their excitation [13]. Phenomena like quantum cutting, whereby one quantum of excitation results in the production of several photons, i.e., with quantum efficiency larger than 1, have been long known [14, 15]. The main variation here will come from bringing such cascades in a stationary regime. When a cascade can be maintained in a steady state, it may result in a chain reaction that gives rise to new dynamical regimes, as in the aforementioned quantum-cascade and bosonic lasing. Under Continuous Wave (CW) excitation, cascades have been particularly studied by the semiconductor community to characterize spectral lines from complex multi-excitonic states [16] through the study of their correlations. This allows to identify the order of transitions [17, 18] and measure their radiative lifetimes [19]. The technique has been for instance demonstrated for the characterization of the tri-exciton, with photon cascades involving up to $N = 5$ excitonic levels, with three radiative steps [20] or four photon transitions from quadexcitons [21]. In the latter case and in other striking examples (e.g., with the tri-exciton again, but correlating all transitions simultaneously [22]), this was under pulsed excitation so that correlations were reduced to coincidences (in particular, of the bunching type), and thus deprived from the time dynamics, which had to be complemented by time-resolved photoluminescence. The reason for pulse excitation is that such cascades balance two types of transitions: downward as the system releases its excitation, and upward as it gets re-excited by the constant driving. As a result, depending on the pumping power, one gets stuck at more or less high stages of the ladder. Consequently, correlations are more easily obtained between consecutive steps, where the system is pinned, although photon correlations being so robust to low signal, they have also been successfully demonstrated between far-apart transitions even in the CW regime [23]. Nevertheless, this two-way option for the excitation which can hop up and down at any stage, weakens the cascade as a whole and effectively turns it into a succession of two-photon cascades. Here, we draw attention to spectacular features present in one-way cascades where the excitation can only relax downward, until it reaches the ground state, at which point it gets excited again to the top, as sketched in Fig. 1(a). Recently, there has been a surge of interest for such unidirectional flow of various types of excitations [24–26]. Notably, both quantum-cascade lasers and bosonic cascades maximize their properties when the flow is unidirectional. Here, we focus on the simplest possible one-way cascading: incoherent pumping initiates the cascade at random times by resetting the single excitation to the highest level N from the bottomest one. There needs not be an actual top and bottom levels, and the structure could really be circular, as sketched in Fig. 1(b). Given that this better captures the structure of the transitions, we shall refer to such cascades as “circular”. We will briefly discuss ways to achieve them but we first motivate their interest, for the correlations between the photons they produce.

* fabrice.laussy@gmail.com

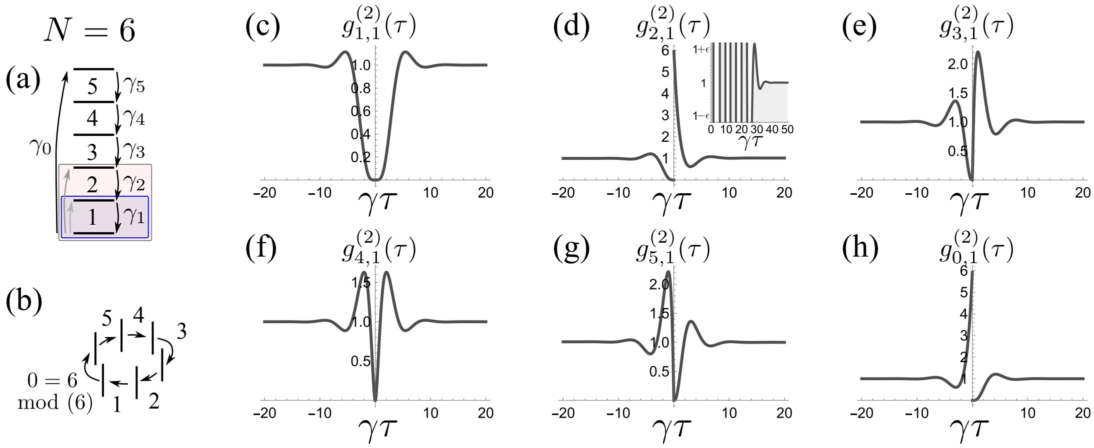

FIG. 1. (a) N-level cascade for $N = 6$, with five radiative transitions at rates γ_i $(1 \leq i \leq 5)$ plus one reloading γ_0. The two-level system is in the dark-shaded box ($N = 2$) and the two-photon cascade from a three-level system in the light-shaded box ($N = 3$), with their own γ_0 transition. The generalization to any N is obvious. (b) The equivalent circular representation, where pumping becomes a regular transition and could also be monitored. (c–h) All the possible correlation functions for $N = 6$, when all $\gamma_i = \gamma$. There are four different traces, modulo time mirror symmetry, that account for all the 36 possible correlations. The inset of $g^{(2)}_{2,1}$ in (d) makes a zoom in the window $1 \pm \epsilon$ where $\epsilon = 10^{-6}$, allowing to resolve five of the oscillations from the cascade, which carry on forever and are present in all correlation functions for $N \geq 3$.

II. LIQUID LIGHT AND WIGNER CRYSTALS

Our motivation is the recent report that such (circular) photon cascades with a large number of intermediate steps endow autocorrelations in time that are reminiscent of those found spatially in liquids [27], and that familiar single-photon correlations from two-level systems are a particular case of this broader cascading scheme. This shows that cascading is interesting even if one step only of the cascade is radiative. Interestingly, in the limit of a large number of levels, this approaches perfect single-photon sources with opening of a time gap [28]. Although stationary, such sources exhibit features of pulsed emission, but without any external synchronization. We became latterly aware that such peculiar correlations had been previously predicted for strongly-interacting Rydberg atoms maintained in their electromagnetically-induced transparency configuration [29]. Such correlations are not available to Kerr-type, point-interacting photon blockade. Instead, they follow from a quite dramatic phase-transition from the optically dense active medium itself, interacting strongly and nonlocally [30] and clustering into small self-avoiding regions, related to Wigner crystallization [29]. Switching off the driving field transfers the spatial correlations to temporal photons. This reinforce one's feeling that such correlations imprinted into the optical field correspond to a new phase (here we cannot write "of matter" since that is now more general than that). A remarkable point is that such correlations—which have been described as "quantum optics in its extreme, in which individual photons behave as impenetrable particles" [31]—are straightforwardly sculpted by a mere cascade mechanism, itself excited incoherently and thus with no external coherence, order or synchronicity. This produces an infinitely-long stream of quasi-crystalline order (to take the terminology of the Rydberg effect) as opposed to a finite size, quenched pulse in most configurations (Zeuthen *et al.* [32] discuss the CW driving converting the Poisson input into a regular pulse train of single photons). Photon liquefaction (to take the cascade terminology) thus appears to be more general and fundamental than one could think since it occurs in completely unrelated platforms from completely different mechanisms. It seems clear that it should be present in still other configurations or in disguise, and represents a fundamental phase of the optical field, which should be further scrutinized, especially as it better corresponds to what one understands as a single-photon source [28]. For instance, this type of correlations is also possible in coherently-driven few-step cascades [33]. The cascade implementation might thus be a privileged platform to realize and investigate it, given its simplicity in both conceptual, theoretical and applied aspects, at least when contrasted to the Rydberg blockade version, which is a highly sophisticated platform, replete with complications in real-laboratory implementations (such as "pollutants", i.e., Rydberg atoms unduly excited by their peers in the propagating blockade [34]).

In the following, we generalize our previous treatments of such circular cascades by studying all the two-photon correlations (not only one transition in isolation [27]) as well as correlations from any subset of transitions. These are other degrees of freedom from our scheme not available from the Rydberg gas. Since large cascades are naturally more difficult to realize than small ones, and because two-photon cascade from a three-level system has already enjoyed considerable popularity and attention, we also focus on this particular case.

III. FORMALISM

Our system of study is simple. If one contents with correlations of the transitions alone, then all the results can be obtained through rate equations, as was also the case for the other types of cascades [17, 35]. Such equations are best written in their matrix form for the probabilities p_i of each level $|i\rangle$ to be excited at time t for $1 \leq i \leq N$:

$$\partial_t \begin{pmatrix} p_N \\ p_{N-1} \\ \vdots \\ p_2 \\ p_1 \end{pmatrix} = \begin{pmatrix} -\gamma_{N-1} & \cdots & & & \gamma_0 \\ \gamma_{N-1} & -\gamma_{N-2} & & & \\ \vdots & \ddots & \ddots & & \\ & & \gamma_2 & -\gamma_1 & \\ 0 & & & \gamma_1 & -\gamma_0 \end{pmatrix} \vec{p}\,, \tag{1}$$

where $\vec{p}$ is the vector on the lhs. This makes for j transitions, from level $|j+1\rangle$ to level $|j\rangle$ for $1 \leq j \leq N-1$, occuring at rate γ_j, while the reloading brings level $|1\rangle$ back to level $|N\rangle$ at rate γ_0. Consequently, there are as many transitions as levels, although the reloading one is usually not regarded as a transition. This is an indication of the conceptual difference between the model (of levels) and the observable (transitions). In a quantum treatment, p_i would be obtained as $\langle i|\,\rho\,|i\rangle$ from the density matrix ρ of the system, but in absence of genuinely quantum observables, we do not need a master equation. Such a quantum description is eventually desirable, but even there, it would be especially because one should correlate the emitted photons instead of the transition operators $|j\rangle\langle j+1|$ from which such photon supposedly originate. We write “supposedly” because there is some inherent quantum uncertainty as to the identity of a *detected* photon, whose attributes (such as frequencies, time of detection, etc.) might not be sufficient to distinguish it from other measured events. By correlating transition operators, one makes the assumption that photons are clearly distinguishable, which can be the case, at least in some approximation, for instance when different transitions differ greatly in frequencies. Note that frequencies do not even appear explicitly at our current level of description. A physical photodetection description which does not make such approximations requires a theory of frequency-resolved photon correlations [36], and we postpone this to a future work. Here, at the level of transition operators, we can solve Eq. (1) with standard linear algebra, expressing the general solution as “basically a sum of exponentials with different time constants” [20]

$$\vec{p}(t) = \sum_{j=0}^{N-1} C_j \vec{A}_j e^{\lambda_j t} \tag{2}$$

where $\vec{A}_j$ and λ_j are the eigenvectors and eigenvalues of the relaxation matrix in Eq. (1). The eigenvalue problem leads to the following equation:

$$\prod_{i=0}^{N-1} (\lambda + \gamma_i) = (-1)^{N-1} \prod_{i=0}^{N-1} \gamma_i \tag{3}$$

with the convention that $\gamma_0 \equiv P$. For low N, we can assume all these variables to be independent, but for large N, since we do not know a closed-form solution for this equation, to simplify our discussion and stick to the main points, we will assume all rates (including the pumping rate), to be the same, equal to γ. This is with some loss of generality, and there are probably important qualitative effects to be found in the more general configurations. In this $\gamma_i = \gamma$ approximation for all i, the eigenvalues can be found as $\lambda_j = \gamma(1 - z_N^j)$ for $0 \leq j \leq N-1$ while the elements of the eigenvectors are given by $A_{jk} = z_N^{jk}$ where

$$z_N \equiv \exp\left(\frac{2i\pi}{N}\right) \tag{4}$$

is the Nth root of unity. The steady state is proportional to the eigenvector with eigenvalue 0, which always exists since the determinant of the matrix is zero. Thus, with $\lambda_0 = 0$ and $A_{0k} = 1$, also imposing $\sum_{i=0}^{N-1} p_i = 1$, then $p_k^{\mathrm{SS}} = 1/N$ for all k. This is the expected result on physical grounds under the given approximations. To compute correlation functions—the quantities of interest in this text—we rely on the stationarity of the signal, making $\vec{p}$ independent of t and thus providing the correlations as

$$g_{m,n}^{(2)}(\tau) = \frac{p_{n|m-1}(\tau)}{p_n^{\mathrm{ss}}} \tag{5}$$

where $p_{n|m-1}(t) = p_n(t)$ as given by Eq. (2) with the initial condition $p_i(0) = \delta_{i,m-1}$, i.e., expressing the probability of finding the system in state n at time $t+\tau$ given that it started in state $m-1$ at time t, i.e., it just underwent the transition m at this time. To compute the constant C_j, we thus have to solve the corresponding Eq. (2) at $t = 0$, i.e.,

$$\sum_{j=0}^{N-1} C_j A_{jn} = \delta_{n,m-1}\,. \tag{6}$$

The solution can be found by noting that the Nth roots of unity sum up to zero, or, using the formula for the sum of the first N terms of a geometric series, we find $C_j = z_N^{-j(m-1)}/N$. This provides us with the general correlation function for $\tau \geq 0$ between any two transitions:

$$g_{m,n}^{(2)}(\tau) = 1 + \sum_{j=1}^{N-1} z_N^{j(n-m+1)} \exp\left(-\gamma\tau(1 - z_N^j)\right)\,. \tag{7}$$

We restricted to positive τ, while for negative τ:

$$g_{m,n}^{(2)}(\tau) = g_{n,m}^{(2)}(-\tau)\,. \tag{8}$$

This is a generalization of our previous autocorrelation function [27] when $m = n$ (which was obtained with the waiting time distribution), in which case the level indices disappear and all transitions feature the same correlations, that are also symmetric in time. The case $m \neq n$ describes cross-correlations, between contiguous, or not, transitions. This describes the density of probability that if a photon is detected from transition m, then another will be (or was if $\tau < 0$) detected from transition n at time τ. The case $N = 1$ washes out all correlations and correspond to classical (Poisson) emission (of the ideal gas in the perspective of Ref. [27]). The case $N = 2$ is the incoherently-pumped two-level system, that describes the paradigmatic CW single-photon source. Although a cascading scheme, it is not traditionally regarded as such but as a single-transition quantum emitter, although its statistics, in particular its failure to suppress efficiently multiphoton emission, is best understood from the circular cascade [28], as we will further discuss. The case $N = 3$ corresponds to the two-photon cascade from a three-level system, which also received a fair share of attention, and to which we return in more details in Section V. The case $N = 6$ is shown in Fig. 1 as a particular case of the obvious general N. We cover this case first to provide results of general validity.

IV. MULTILEVEL CASCADE

From the N^2 possibilities for correlating any two transitions from a N-level cascade, assuming equal rates, Eq. (7) reduces to N different positive τ traces for the correlation functions:

$$g^{(2)}_{k\{N\}}(\tau) \equiv 1 + \sum_{j=1}^{N-1} z_N^{jk} \exp\left(-\gamma\tau(1 - z_N^j)\right) . \quad (9)$$

By cyclicity of the roots of unity, $g^{(2)}_{k\{N\}} = g^{(2)}_{(k+N)\{N\}}$. All display maximum antibunching

$$g^{(2)}_{k\{N\}}(0) = 0 \quad (10)$$

except if $k = 0$ (or a multiple of N) in which case, $z_N^0 = z_N^N = 1$ and the bunching is equal to the number of levels:

$$g^{(2)}_{0\{N\}}(0) = N . \quad (11)$$

For convenience, we use the terminology of "bunching" and "antibunching" for both auto and cross-correlations, although this properly describes autocorrelations only. These functions further combine to provide the full τ (positive or negative) correlations according to Eq. (8), so there are finally $\lfloor N/2 \rfloor + 1$ different all-τ traces for a N-level circular cascade, taking into account time mirror symmetry (the $+1$ to account for the autocorrelation). The relationship between all possible transitions and the backbone structure (9) is obtained by rotation of the cascade as, for any $1 \leq k, l \leq N$:

$$g^{(2)}_{k,l} = g^{(2)}_{(l-k+1)\{N\}} \quad (12)$$

where on the left-hand side, the two indices now correspond to which transitions are correlated, while on the right-hand side, one has Eq. (9) where the curly bracket index serves to track the number of transitions. This is valid for all τ. This means that it is enough to know the correlation between one transition (say the bottom one) and the successive ones over halfway trough the ladder to know all the correlations (involving a time mirror symmetry for the remaining transitions). For instance, in Fig. 1, all autocorrelations are identical to $g^{(2)}_{1,1}$ in panel (c) and $g^{(2)}_{2,1}$ is the time-mirror of $g^{(2)}_{0,1}$, or, alternatively, is identical to $g^{(2)}_{1,0}$. The physical interpretation of such symmetries is clear: $g^{(2)}_{2,1}$ shows the density of probability to detect a photon from the transition 1 after detecting a photon from transition 2. Given that these transitions are contiguous, the chances are high as directly related to the transition rate. The probability is further boosted by the knowledge of the current state of the system, which could otherwise be in any of the N states prior to effectuating a transition, but removing this uncertainty makes it N time more likely that it will now emit at the sought transition, thus explaining the integer factor (11). There is also a notable revival of probabilities, in the form of an elbow at $\tau \approx N/\gamma$. This corresponds to the second-order detection where the system went around through the whole cascade again after emitting the first photon, before detecting the second one. There are similar peaks at $\tau \approx kN/\gamma$ for all k, corresponding to going round the whole cascade k times before detection. One can resolve them by zooming in the correlation function, as shown in the inset of $g^{(2)}_{2,1}$ in Fig. 1(d), or by plotting $g^{(2)} - 1$ in log-scale. Such oscillations occur in all correlation functions for $N > 2$ and have been described in more details for the autocorrelation function [27]. Because they occur without any coherence and/or periodic driving, they can be seen as self-oscillations [37], although of correlations from a system that is itself stationary. Their magnitude increases slowly but surely with N, as shown in Fig. 2 through the values of $g^{(2)}_{i,i}(\tau)$ for the successive peaks as a function of N, i.e., the successive maxima of $g^{(2)}_{1\{N\}}$. It requires $N = 6$ to get a bunching of the first oscillation of $g^{(2)} \approx 1.1$ (10% deviation, cf. Fig. 1(c)) and $N = 13$ for the second peak to reach that value. At $N = 50$, othe seventh peak has $g^{(2)} \approx 1.13$ and the eight one $g^{(2)} \approx 1.09$ so at least eight-order cascades can be considered neatly resolvable, i.e., from the steady stream, strong correlations are maintained between any given photon and its eight descendant round the cascade, each generation undergoing fifty transitions. More precise experiments could track down such correlations arbitrarily down the stream for any N. A similar dynamics occurs for negative times: the chances

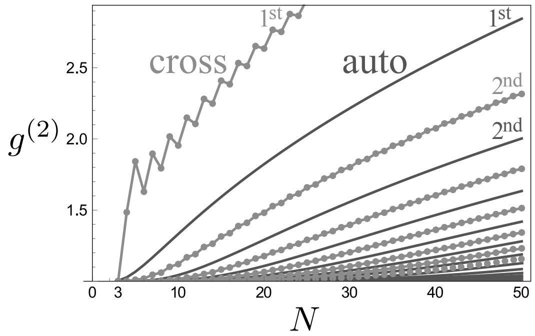

FIG. 2. Magnitudes of the successive peaks of $g^{(2)}_{1\{N\}}$ (autocorrelations, dark lines) and $g^{(2)}_{1\{\lfloor (N+3)/2\rfloor\}}$ (cross-correlations between opposite transitions in the ladder, dotted lines) as a function of N. The cross-correlations oscillate because of their asymmetry for odd N (the largest peak is taken). For clarity, only the first seven peaks of cross-correlations are retained.

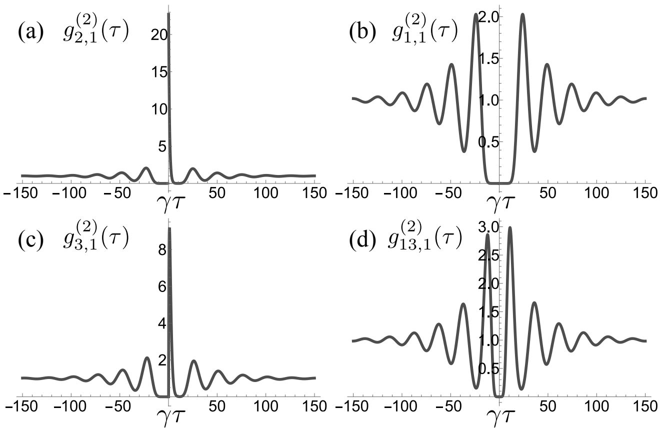

FIG. 3. N-level cascade with $N = 25$, showing four illustrative of the thirteen different correlations out of the 625 possible ones, namely, (a) contiguous or (c) next-to-contiguous transitions as well as (b) autocorrelations and (d) mid-ladder transitions. Long τ oscillations are now neatly resolved, manifesting a strong local time-ordering. The correlation function is discontinuous in (a) but not in the other panels, where it exhibits a more or less flat plateau around $\tau = 0$ as specified by Eq. (13).

get strongly suppressed that a photon from the transition 1 be observed before ($\tau < 0$) one from transition 2, which constitutes a transition in the wrong time order. In a physical model, such a process becomes possible due to detector uncertainties. Here, it is strongly suppressed because it requires to go round the full cascade to get another photon to pause a wrong-order one. This is impeded by all the intermediate steps. The succession of antibunching at negative τ followed by bunching at positive τ defines the archetype of a photon cascade, although this is one particular step only of the general dynamics, but an important one to which we come back in Section V. The identity between $g^{(2)}_{2,1}$ and $g^{(2)}_{1,0}$ is also clear as we consider the same processes in different positions of the ladder, as is obvious on the circular picture Fig.1(b) of the cascade, in contrast to the linear one (a). Similar dynamics explain non-contiguous transitions, e.g., $g^{(2)}_{3,1}$ in panel (e) shows the bunching at positive τ of the almost contiguous cascade, with only one intermediate transition to make, whose interruption also explains the $\tau = 0$ antibunching in this and all other non-contiguous correlations. Importantly, this flattens in time as the distance between transitions increases in the ladder. This is the principle of photon liquefaction, which endows the correlations with an intrinsic, spontaneous mechanism for local time-ordering. The small τ behaviors can be characterized through a series expansion of Eq. (9), but since this removes the modularity of k, one must ensure that $1 \le k \le N$:

$$g^{(2)}_{k\{N\}}(\tau) = \frac{N}{(N-k)!}(\gamma\tau)^{N-k} + o(\tau^{N-k+1})\,. \tag{13}$$

The case $k = N$ (which identifies to $k = 0$ modulo N in Eq. (12) and corresponds to contiguous transitions) is expanded not around zero (as per Eq. (10)) but around N (as per Eq. 11) and one must thus treat this case separately, to find:

$$g^{(2)}_{N\{N\}}(\tau) = N\exp(-\gamma\tau) + o(\tau^{N+1}) \tag{14}$$

so that to next-leading order in small times, $g^{(2)}_{N\{N\}}(\tau) = N(1-\tau) + o(\tau^2)$, in agreement with Eq. (13). All together, this shows that high-order corrections account for the long-term behaviour of these functions, following the plateau, or exponential decay for $k = N$, when oscillations kick in before converging to $g^{(2)}_{k,N}(\infty) = 1$. The small-time approximations also confirm that the farther the transitions in the cascade, the more suppressed is the possibility of a coincidence in the way accounted for by cumulative random events [28]: the probability to detect photons from two given transitions k-steps away is given by the compound probabilities of the intermediate steps in the sequence. This also “crystallizes” the chance of the coincidence to occur at the kth trial, so one has a steeper and larger bunching for small k. Once such a one-cascade loop has been completed, the process can then repeat, diluting the features as they become both less strong and less well-resolved in time due to accumulations of time uncertainties, but still resulting in all-time oscillations, already for the $N = 3$ cascade (but not for $N = 2$, showing that the effect is not trivial and some type of minimum cooperativeness is required for it to occur). An illustrative case is shown in Fig. 3 for the case $N = 25$. In this case, there are $\lfloor 25/2\rfloor + 1 = 13$ different traces but it is enough to consider a few illustrative cases, namely, the contiguous or almost contiguous transitions, shown in Panels (a) and (c), which one can contrast with their lower N case counterparts in Fig. 1(d) and (e), to see how the liquefaction phenomenology superimposes itself to the immediate cascading. The traces in (a) and (c) are very similar, except at small τ, where the strong bunching is curbed by the impossibility of a concidence from the in-betweener, cf. Eq. (10). As a result, the curve in panel (a) is discontinuous at $\tau = 0$ while that in (c) is not. They differ more from the autocorrelation function $g^{(2)}_{1\{25\}}$ in panel (b) as well as the mid-ladder case $g^{(2)}_{13\{25\}}$ in (d) that correlates the most distant transitions in the ladder. Since N is, in this case, odd, there is one more transition on one side of the cascade and the function is not exactly symmetric (cf. Fig. 1(f), which is symmetric). Such opposite-transition correlations look qualitatively like autocorrelations (especially when exactly symmetric) although they are more correlated, with steeper and higher bunching peaks, except that their temporal gap is shorter, being $N/(2\gamma)$ or half the gap of the N/γ autocorrelation, since it takes that time only to connect the two events. This shows how auto-correlations bear more constrains as applying within the same stream as opposed to cross-correlations that involve two signals. This relates to an important concept, which has accompanied photon cascades since their early days, namely, that of their violation of classical inequalities [38], predominantly, Cauchy-Schwarz inequalities of the type $g^{(2)}_{nn}(0)g^{(2)}_{mm}(0) \geq [g^{(2)}_{nm}(\tau)]^2$, which classical signals must satisfy. At our current level of description, the violation is trivially enforced for small times, in the form (from Eq. (13))

$$0 \geq \frac{N^2(\gamma\tau)^{2(N-m+n-1)}}{[N-m+n-1]!^2}\,, \tag{15}$$

which can never be satisfied (i.e., Cauchy–Schwarz inequalities are always violated). The violation is larger, the farther away the transitions. Such violations, which indicate that one is dealing with a quantum signal with no classical equivalent, were in fact first observed by Burnham and Weinberg [39] with parametric down conversion and provided the earliest experiment evidence of non-classicality of light, well ahead of the more famous antibunching experiment of Kimble *et al.* [40], which is the degenerate $n = m$ version. Circular cascades thus provide a platform to evidence even more forcefully non-classical correlations from light, even though from a more mundane relaxation, with no photon splitting.

Now that we characterized all possible transitions, we can take the next engineering step afforded by such cascades, by collecting light from a given subset $\mathcal{S} = \{i_1, \cdots, i_n\}$ of transitions. This could be enforced by having such transitions have the same frequency and filtering out the undesired ones, whose participation to the cascade is still necessary but not their contribution to the emitted light. One could also think of selection by polarization, non-radiative process or any other trick whose end result is to collect only photons from the subset. In this case, the correlation function follows from the basic correlation functions (7) as

$$g^{(2)}_{\mathcal{S}}(\tau) = \frac{1}{(n_{\mathcal{S}})^2}\sum_{i,j\in\mathcal{S}} g^{(2)}_{i,j}(\tau) \tag{16}$$

where $n_{\mathcal{S}} \equiv \#\mathcal{S}$ is the cardinality of $\mathcal{S}$, i.e., the number of transitions contributing to the emission. Thanks to Eq. (8), $g^{(2)}_{\mathcal{S}}$ is τ-symmetric, as should be for a correlation function. Some examples are shown in Fig. 4, this time for $N = 50$. Of particular interest is to select k successive transitions, in which case, as long as $k \ll N$, one has essentially the same autocorrelation but pierced through by a central superbunching peak, of magnitude

$$g^{(2)}_{[\![1,k]\!]}(0) = \frac{N(n_{\mathcal{S}}-1)}{{n_{\mathcal{S}}}^2}\,, \tag{17}$$

as shown in Fig. 4(a). This corresponds to a liquid of k-photon bundles, where each emission consists of a so-called bundle (group) of k photons, i.e., closely-packed photons in time as compared to the photons from the other bundles. Thanks to the cascaded emission process, those bundles do not have the harmonic progression of their cavity QED counterpart [41], but have instead a more flexible temporal structure which could be further engineered through the decay rates of the transitions. Similarly to the case of single photons, the cascaded regime optimizes their correlations through temporal liquefaction [27], showing that the previous proposals [41, 42] correspond to a temporal gas of bundles. Interestingly, as an incoherent process, the bundle purity (percentage of events featuring exactly the sought k number of photons in each bundle) is not fundamentally limited and could thus be arbitrarily close to 1 regardless of the size of the bundle. This is therefore another example of the great flexibility and versatility of the circular quantum cascade. For bundling as for Wigner crystallization, it provides much simpler, robust and tuneable implementations.

V. TWO-PHOTON CASCADE IN THE THREE-LEVEL SYSTEM

The case $N = 3$ is both that of the simplest and most studied radiative cascade under incoherent pumping. Largely, this is due to the rich bi-exciton/exciton cascade in semiconductors [43, 44] and its potential for generating entanglement [45]. We can expect the circular implementation to be also more easily realized with fewer steps. The two steps with $N = 3$ (if not counting the excitation one) realize the paradigm of a photon cascade. For that reason, we can indulge into a more general study where we relax the degeneracy between the parameters. This will allow us to get an idea of how much has just been overlooked in such an approximation. Before that, however, it is instructive to pause and consider the case $N = 2$ which, as an incoherently pumped system, describe the archetypal single-photon source (red box in

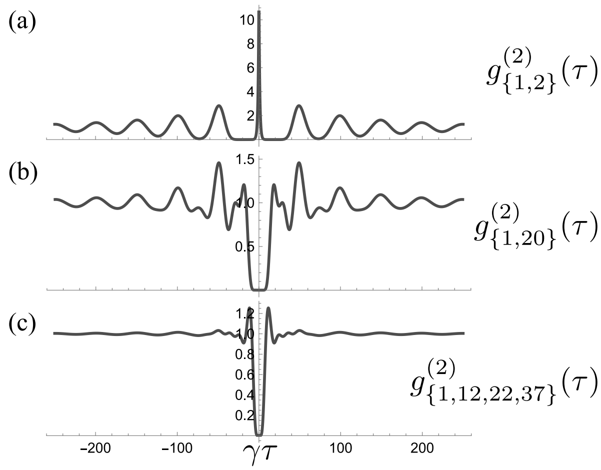

FIG. 4. Many-transition correlations where all the labelled transitions in the subscript set are jointly detected, here for an $N = 50$-level cascade. (a) Correlating two consecutive transitions realizes a two-photon bundle liquid, with the appearance of a superbunching peak (with $g^{(2)}_{\{1,2\}}(0) = 12.5$ from Eq. (17)). (b) Correlating distant transitions bring chaotic looking patterns. (c) A random subset of four transitions wash out the oscillations while still maintening the temporal gap, realizing a dense temporal gas. Many other combinations can be realized and remain to be investigated.

Fig. 1(a)). Our formalism shows, however, that it is the special case of the "one-photon cascade in the two-level system". This adds to the well-known and standard autocorrelation function

$$g_{1,1\{N=2\}}(\tau) = 1 - \exp\big(-(\gamma_0+\gamma_1)|\tau|\big) \tag{18}$$

the cross-correlation between emission and excitation:

$$g_{1,0\{N=2\}}(\tau) = 1 + \left(\frac{\gamma_0}{\gamma_1}\right)^{\varsigma(\tau)} \exp\big(-(\gamma_0+\gamma_1)|\tau|\big) \tag{19}$$

where $\varsigma(\tau) \equiv 2\Theta(\tau) - 1$ is $+1$ if $\tau \geq 0$ and -1 otherwise (Θ is the Heavyside function). By circular symmetry, one gets $g^{(2)}_{0,0}(\tau) = g^{(2)}_{1,1}(\tau)$ and $g_{0,1}(\tau) = g_{1,0}(-\tau)$. This solution is notable for having been apparently overlooked for so long. One of its merit is to clarify why incoherent pumping of a two-level system fails to provide a good single photon source: behind the antibunching (18) lurks the strong bunching (19) of the re-excitation, which counteracts from its cascading nature, the suppression of coincidences in the autocorrelations. This is the basic fact whose understanding and mitigation should drive the design of efficient CW single-photon sources. The $N = 3$ steady-state solution of Eq. (1) can now be found in full generality:

$$\vec{p}_{\mathrm{ss}} = \Bigg(\sum_{\substack{i,j=0\\ i>j}}^{2} \gamma_i\gamma_j\Bigg)^{-1} \begin{pmatrix}\gamma_0\gamma_1\\ \gamma_0\gamma_2\\ \gamma_1\gamma_2\end{pmatrix}. \tag{20}$$

The autocorrelation function is the same for both transitions, which is also a feature of Eq. (7) as already observed. Therefore, for $i = 1, 2$:

$$g^{(2)}_{i,i}(\tau) = 1 + \\ + \frac{\gamma_{012}-\zeta}{2\zeta} e^{-\frac{\gamma_{012}+\zeta}{2}|\tau|} - \frac{\gamma_{012}+\zeta}{2\zeta} e^{-\frac{\gamma_{012}-\zeta}{2}|\tau|} \tag{21}$$

where we defined

$$\zeta \equiv \sqrt{\sum_{i=0}^{2}\gamma_i^2 - \sum_{\substack{i,j=0\\ i\neq j}}^{2}\gamma_i\gamma_j} \tag{22}$$

as well as $\gamma_{i_1\cdots i_k} = \gamma_{i_1} + \cdots + \gamma_{i_k}$ with as many indices as there are terms in the sum. We will use a bar to denote a negative term, $\gamma_{i_1\cdots\overline{i_k}} = \gamma_{i_1} + \cdots - \gamma_{i_k}$, so that, e.g., $\gamma_{0\bar{1}2} \equiv \gamma_0 - \gamma_1 + \gamma_2$. We remind that $\gamma_0 \equiv P$ is the pumping rate. For the transition correlations in this case, we find:

$$g^{(2)}_{2,1}(\tau) = 1 + \\ \begin{cases} \frac{1}{4\zeta\gamma_2^2}\Big[(\gamma_{0\bar{1}2}-\zeta)\big(\gamma_1^2+\gamma_2^2-(\gamma_0+\zeta)\gamma_{12}\big)e^{-\frac{\zeta+\gamma_{012}}{2}\tau} - (\gamma_{0\bar{1}2}+\zeta)\big(\gamma_1^2+\gamma_2^2-(\gamma_0-\zeta)\gamma_{12}\big)e^{\frac{\zeta-\gamma_{012}}{2}\tau}\Big] & \text{if } \tau<0\,,\\ \frac{1}{4\zeta\gamma_0\gamma_1}\Big[(\gamma_{\bar{0}12}+\zeta)\big((\gamma_{0\bar{1}}+\zeta)\gamma_1+(2\gamma_0+\gamma_1)\gamma_2\big)e^{\frac{\zeta+\gamma_{012}}{2}\tau} - (\gamma_{\bar{0}12}-\zeta)\big((\gamma_{0\bar{1}}-\zeta)\gamma_1+(2\gamma_0+\gamma_1)\gamma_2\big)e^{-\frac{\zeta-\gamma_{012}}{2}\tau}\Big] & \text{if } \tau>0\,, \end{cases} \tag{23}$$

while, following Eq. (8), $g^{(2)}_{2,1}(\tau) = g^{(2)}_{1,2}(-\tau)$.

A first interesting observation from this general solution is to consider the range of parameters that result in oscillations or, on the other hand, in damped relaxations. This follows from the sign of the radicand of Eq. (22), which makes ζ real or imaginary and its exponential in Eqs. (21) and (23) an additional damping or an oscillation, respectively. Spelling out the condition for oscillations, we must have $2\gamma_0\gamma_1 + 2\gamma_0\gamma_2 + 2\gamma_1\gamma_2 > \gamma_0^2 + \gamma_1^2 + \gamma_2^2$ with all $\gamma_i > 0$. Rearranging for γ_2 as a function of γ_0

and γ_1, this leads to $\gamma_2^2 - 2\gamma_2(\gamma_0+\gamma_1) - (\gamma_0-\gamma_1)^2 < 0$. The zeros of this quadratic equation thus give us the range of γ_2 that results in oscillations of the correlation functions for a given γ_0 and γ_1 as

$$(\sqrt{\gamma_0} - \sqrt{\gamma_1})^2 < \gamma_2 < (\sqrt{\gamma_0} + \sqrt{\gamma_1})^2\,. \tag{24}$$

The same relations hold for any permutations of the indices. This shows that if two rates are equal, oscillations occur as long as the third (nonzero) rate is less than four times the common decay rate. Clearly, the case where all three decay rates are equal satisfy the condition for oscillations. While we do not provide an exact statement for the general case, it seems clear that a small spread of the decay rates favour oscillations while at the same time accommodating large deviations for some of the parameters. With $N = 3$, such oscillations are not resolvable from the correlation function in a linear scale, since their amplitude is too small (they would be visible in log-scale for $g^{(2)} - 1$), but this serves to illustrate the different structural regimes that the mechanism supports and how they survive or not different parameters. Extrapolated to larger N, liquid light should thus be accessible even with significant inhomogeneities of the cascading steps.

The exact form (23) for the two-photon cascade in a three-level system admits in regimes of interest simpler expressions, that one can compare to the phenomenological two-photon cascade correlations that assumes an uncorrelated (Poissonian) stream of photons heralding another stream [46]:

$$g^{(2)}(\tau) = 1 + \begin{cases} (1-p)\dfrac{\gamma_2}{\gamma_1}\exp(\gamma_2\tau) & \text{if } \tau < 0, \\ p\dfrac{\gamma_2}{\gamma_1}\exp(-\gamma_2\tau) & \text{if } \tau > 0. \end{cases} \tag{25}$$

where p describes the probability of good time ordering, γ_1 the emission rate of the uncorrelated heralding (first) photon and γ_2 that of the heralded (second) photons. Namely, in the limit of small excitation, i.e., when $\gamma_0 \ll \gamma_1, \gamma_2$, so that $\zeta \approx \sqrt{\gamma_1^2 + \gamma_2^2 - 2(\gamma_1+\gamma_2)} = |\gamma_1 - \gamma_2|$, in which case Eq. (23) takes the form:

$$g^{(2)}_{2,1}(\tau) = 1 + \begin{cases} -\exp(\gamma_1\tau) & \text{if } \tau < 0\,, \\ \frac{\gamma_2}{\gamma_0}\exp(-\gamma_2\tau) & \text{if } \tau > 0\,, \end{cases} \tag{26}$$

On the other hand, in the limit of high-pumping $\gamma_0 \gg \gamma_1, \gamma_2$, one finds:

$$g^{(2)}_{2,1}(\tau) = 1 + \begin{cases} \frac{\gamma_1}{\gamma_2}\exp([\gamma_1+\gamma_2]\tau) - \left(1 + \frac{\gamma_1}{\gamma_2}\right)\exp(\gamma_0\tau) & \text{if } \tau < 0\,, \\ \frac{\gamma_2}{\gamma_1}\exp(-[\gamma_1+\gamma_2]\tau) & \text{if } \tau > 0\,., \end{cases} \tag{27}$$

The second (negative) term for $\tau < 0$ is negligible for most τ except very close to zero where it forces the correlations to be exactly antibunched. This is the main deviation of the cascading scheme as compared to Eq. (25) which assumes uncorrelated heralders. The cascade, on the other hand, requires the same excitation to go up and down the ladder and thus demands that

$$\lim_{\substack{\tau\to 0\\ \tau<0}} g^{(2)}_{2,1}(\tau) = 0 \tag{28}$$

which is the counterpart of Eq. (10) satisfied also by Eq. (23) and the low-driving approximation (25). Since, on the other hand

$$\lim_{\substack{\tau\to 0\\ \tau>0}} g^{(2)}_{2,1}(\tau) = 1 + \left(\frac{1}{\gamma_0} + \frac{1}{\gamma_1}\right)\gamma_2 \tag{29}$$

(with limits $1 + \frac{\gamma_2}{\gamma_0}$ for low (26) and $1 + \frac{\gamma_2}{\gamma_1}$ for high (27) pumping, respectively), there is again the discontinuity at $\tau = 0$, as is also the case in the phenomenological model and in the more general cascade (11). This would be resolved with a photo-detection theory [36]. Lifting the degeneracy of the relaxation rates show that the discontinuity (11) is the smallest that can be, and that large γ_2 or small γ_0 and/or γ_1 result in strong discontinuities, as the cascade is rarefied as compared to its Poisson occurence. It is interesting, in this regard, to consider the opposite limit which tames down the correlations, as shown in Fig. 5, where one sees that the decorrelation of the heralded photon in the cascade, comes at the cost of strong correlations in the wrong order, i.e., of the heralding one instead, despite the perfect suppression of coincidences from Eq. (28). This realizes a "backward-cascade" where the detection of photons from the supposedly "heralding" transition tells us nothing about the subsequent transition of an heralded one, which are thus detected randomly in time, while such a transition effectively "heralds its heralder", i.e., the photon from the prior transition is in fact emitted shortly and strictly afterwards (in the wrong time order and never at the same time). That is because it is, of course, the next heralder, but this fact does not transpire in the emission and such a "feature" would not be easy to achieve otherwise. Reversing the detectors with Eq. (8), one thus has uncorrelated heralders heralding slightly delayed photons. We thus have a recipe to implement closely the phenomenological cascade of Eq. (25), would that be requested.

Finally, would we consider the $N = 3$ case not as a two-photon cascade from the three-level system, but as the generic circular cascade of Fig. 1(b) where all transitions can be correlated, then $g^{(2)}_{0,0}$ is given by Eq. (21) and the cross-correlations are similarly given by Eq. (23) with rotation of the parameters, namely, $g^{(2)}_{1,0}$ is obtained from the substitutions $(\gamma_0, \gamma_1, \gamma_2) \to (\gamma_2, \gamma_0, \gamma_1)$ and $g^{(2)}_{0,2}$ from the substitutions $(\gamma_0, \gamma_1, \gamma_2) \to (\gamma_1, \gamma_2, \gamma_0)$, while the other orders follow from Eq. (8). These correlations behave qualitatively as discussed in Fig. 1 but lifting the degeneracy of the transition rates and with similar enhancement and distortions of their correlations. This shows that non-degenerate decay rates do not enlarge the number of possible traces.

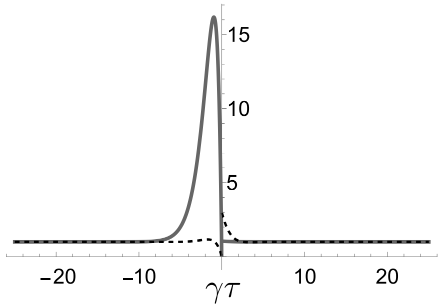

FIG. 5. Two-photon cascade in a three-level system with unbalanced rates (thick solid line, with $(\gamma_1, \gamma_2)/\gamma_0 = (1.1, 0.025)$), resulting in a markedly different correlation function (compare with all rates equal to the mean $\gamma \approx 0.71\gamma_0$ as the dashed thin line). The heralded photon is uncorrelated with the heralding one, while the opposite transition is strongly bunched at small negative delays, though still obeying the constrain (28) at $\tau = 0$. Similar strong departures are expected for the general N case. These parameters satisfy Eq. (24) so the functions are forever oscillating, although this is not visible on a linear scale.

VI. DISCUSSION AND CONCLUSIONS

We have provided a theory of circular quantum cascades, whereby a level can be excited from one direction only. While this seems to differ little from widespread and thoroughly studied two-way cascades, where the state can be excited from "above" (by relaxation) or from "below" (by excitation), this actually presents a considerably richer dynamics, able to endow the system with strong correlations that are washed away in other cases by breaking down the ladder into a succession of pairwise cascades. Indeed, although the formalism to describe circular cascades brings little novelty from those developped to describe other types, the nature of the solutions—developing all-time oscillations under incoherent excitation of the system—make a dramatic departure from the mere relaxation regimes previously discussed. From the comprehensive description of all the possible traces for the various combinations of transitions that can be correlated (which to the best of our knowledge remains to be similarly classified for traditional cascades), we highlighted how autocorrelations surprisingly recover highly-sought regimes of photon phase transitions, such as those realized in large ensemble of coupled optical cavities [47–49]. These implementations have been described as a "complex architecture [that] represents a considerable experimental challenge" by an EIT Rydberg group seeking similar correlations [31], also praising the Rydberg out-of-equilibrium character and its facility to transfer them to the optical field. Our mechanism can make the same remarks on the EIT Rydberg platform: the cascaded system is much more straightforward experimentally, being in principle available with a single multi-level emitter, it also needs no equilibrium, in fact not even coherent driving, and cares little about underlying details of the excitation, and it is directly and intrinsically built into the optical field. This challenges the previously held view that the "formation of a Tonks–Girardeau gas of photons is fundamentally a collective many-body effect" [31]. This in fact emerges as a much more fundamental and universal feature of the optical field itself, regardless of the underlying mechanism which produces it, that we identify as that of good single-photon emission. Nevertheless, the coupled cavities and Rydberg EIT physics being deeply rooted indeed in strongly-correlated many-body physics—whose photon correlations are directly linked to the pair correlation function of the Lieb-Liniger gas [50], where they are interpreted as Friedel oscillations [51]—this gives further credence to our earlier suggestions of a condensed-matter, thermodynamically inspired description of the optical field [27]. In our case, photons are not interacting, they merely inherit or imprint correlations that are those of a liquid, so even the denomination of "liquid light" might not be entirely correct and one should instead speak of *liquid time*, since this is properly, and maybe exclusively, the ticks in time that exhibit the features, not its carriers. Another striking demonstration of the might of circular cascades is their built-in ability to generate CW N-photon bundles, for any integer N and with no fundamental restriction on the bundle purity, which appear to also provide considerable improvement on their cavity QED counterparts. At any rate, proper attention to such potentialities and thus the exploitation of the wonders they promise, will require the experimental feasibility of such cascades. It is beyond the scope of this text to devise a microscopic system to achieve that, although we reiterate that in our view, despite the physical constraints on relaxation rates in N-level quantum systems [52], this seems a much simpler task than those involving strongly-correlated many-body platforms driven in extreme regimes. We can imagine several "poor man"'s solutions to the problem, including pulsed excitation to avoid re-excitation or coherent driving to isolate the γ_0 transition from the others. In such cases, however, there would be the risk of mistaking the oscillations for those imparted by the driving itself [53, 54]. With the onset of chiral quantum optics [55] and topological photonics [56], we have no doubt that resourceful inventors can find a faithful implementation of a genuine one-way one-photon (or one-quantum, in other platforms) cascade, with the result of producing not one, but a variety of stationary quantum light, yet so strongly correlated that they locally appear to be pulsed.

From our side, rather than indulging into such designs, we wish to conclude with the reiterated observation that the current theory of circular cascades, as well as, incidentally, those describing other types of cascades, rely on correlating transitions, as opposed to correlating detected photons. We believe that much physics and further engineering await to be revealed by applying the sensor formalism [36] to photon cascades—circular or traditional—with the effect of removing undesirable artifacts such as discontinuities in the correlation functions, taking more seriously photon indistinguishability as well as interferences, and, not least, providing a complete leapfrog (off-peak) picture of the transitions [57], also placing the observation at the heart of the process, thereby showing that quantum jumps and quantum cascades are two faces of the same coin. We hope to provide such a complete description soon.

ACKNOWLEDGMENTS

We thank Drs. Arturo Camacho Guardian, Asaf Paris Mandoki, Giuseppe Pirucci and Hugo Alberto Lara Garcia for attracting our attention to the Rydberg blockade correlations, at the occasion of the first Etic Tlahuilli school. EdV and FPL thank Guillermo del Valle and Lotte Crombaghs for their hospitality on their wedding week, during which this work was finalized. EdV acknowledges support from the TUM-IAS Hans Fischer Fellowship, the CAM Pricit Plan (Ayudas de Excelencia del Profesorado Universitario), Sinérgico CAM project Y2020/TCS-6545 (NanoQuCo-CM) and the Spanish Ministry of Science, Innovation and Universities under grant AEI/10.13039/501100011033 (2DEnLight) and through the "María de Maeztu" Programme for Units of Excellence in R&D (CEX2023-001316-M). FPL acknowledges support from the HORIZON EIC-2022-PATHFINDERCHALLENGES-01 HEISINGBERG project 101114978.

[1] Kazarinov, R. F. & Suris, R. Possibility of amplification of electromagnetic waves in a semiconductor with a superlattice. *Fizika i Tekhnika Poluprovodnikov* **5**, 797 (1971).
[2] Faist, J. *et al.* Quantum cascade laser. *Science* **264**, 553 (1994). URL 10.1126/science.264.5158.553.
[3] Gao, L., Feng, C. & Zhao, X. Recent developments in terahertz quantum cascade lasers for practical applications. *Nanotechnol. Rev.* **12**, 20230115 (2023). URL doi:10.1515/ntrev-2023-0115.
[4] Liew, T. C. H. *et al.* Proposal for a bosonic cascade laser. *Phys. Rev. Lett.* **110**, 047402 (2013). URL doi:10.1103/PhysRevLett.110.047402.
[5] Liew, T. C. H. *et al.* Quantum statistics of bosonic cascades. *New J. Phys.* **18**, 023041 (2016). URL doi:10.1088/1367-2630/18/2/023041.
[6] Crubellier, A. & Pavolini, D. Superradiance and subradiance. IV. atomic cascades between degenerate levels. *J. Phys. B.: At. Mol. Phys.* **20**, 1451 (1987). URL doi:10.1088/0022-3700/20/7/014.
[7] Briegel, H., Meyer, G. M. & Englert, B. Correlated atomic excitation in multilevel lasers. *Proc. SPIE* **43**, 2792 (1996). URL doi:10.1117/12.239183.
[8] Rodnyĭ, P. A. Cascade photon emission in luminophores. *Opt. Spectrosc.* **89**, 556 (2000). URL doi:10.1134/bf03356020.
[9] Bünzli, J. G., Comby, S., Chauvin, A. & Vandevyver, C. D. New opportunities for lanthanide luminescence. *J. Rare Earths* **25**, 257 (2007). URL doi:10.1016/s1002-0721(07)60420-7.
[10] Zhou, J., Teng, Y., Zhou, S. & Qiu, J. Quantum cutting in luminescent glasses and glass ceramics. *Int. J. Appl. Glass Sci.* **3**, 299 (2012). URL doi:10.1111/ijag.12000.
[11] Peng, D. *et al.* Lanthanide-doped energy cascade nanoparticles: Full spectrum emission by single wavelength excitation. *Chem. Mater.* **27**, 3115 (2015). URL doi:10.1021/acs.chemmater.5b00775.
[12] Fritzsche, S., Palmeri, P. & Schippers, S. Atomic cascade computations. *Symmetry* **13**, 520 (2021). URL doi:10.3390/sym13030520.
[13] Fontana, P. R. *Atomic Radiative Processes* (Academic Pr, 1982).
[14] Sommerdijk, J., Bril, A. & d. Jager, A. Two photon luminescence with ultraviolet excitation of trivalent praseodymium. *J. Lumin.* **8**, 341 (1974). URL doi:10.1016/0022-2313(74)90006-4.
[15] Piper, W., DeLuca, J. & Ham, F. Cascade fluorescent decay in Pr^{3+}-doped fluorides: Achievement of a quantum yield greater than unity for emission of visible light. *J. Lumin.* **8**, 344 (1974). URL doi:10.1016/0022-2313(74)90007-6.
[16] Bayer, M., Stern, O., Hawrylak, P., Fafard, S. & Forchel, A. Hidden symmetries in the energy levels of excitonic 'artificial atoms'. *Nature* **405**, 923 (2000). URL doi:10.1038/35016020.
[17] Dekel, E. *et al.* Cascade evolution and radiative recombination of quantum dot multiexcitons studied by time-resolved spectroscopy. *Phys. Rev. B* **62**, 11038 (2000). URL doi:10.1103/physrevb.62.11038.
[18] Regelman, D. V. *et al.* Semiconductor quantum dot: A quantum light source of multicolor photons with tunable statistic. *Phys. Rev. Lett.* **87**, 257401 (2001). URL doi:10.1103/PhysRevLett.87.257401.
[19] Camhy-Val, C., Dumont, A., Dreux, M. & Vitry, R. Absolute measures of mean lifetimes of excited states using the method of correlated photons in cascade. *Phys. Lett. A* **32**, 233 (1970). URL doi:10.1016/0375-9601(70)90295-1.
[20] Persson, J., Aichele, T., Zwiller, V., Samuelson, L. & Benson, O. Three-photon cascade from single self-assembled inp quantum dots. *Phys. Rev. B* **69**, 233314 (2004). URL doi:10.1103/physrevb.69.233314.
[21] Arashida, Y., Ogawa, Y. & Minami, F. Four-photon cascade from quadexcitons in a single gaas quantum dot. *Phys. Rev. B* **84**, 125309 (2011). URL doi:10.1103/physrevb.84.125309.
[22] Schmidgall, E. R. *et al.* Deterministic generation of a quantum-dot-confined triexciton and its radiative decay via three-photon cascade. *Phys. Rev. B* **90**, 241411 (2014). URL doi:10.1103/physrevb.90.241411.
[23] Molas, M. R., Nicolet, A. A. L., Babiński, A. & Potemski, M. Quadexciton cascade and fine-structure splitting of the triexciton in a single quantum dot. *Europhys. Lett.* **113**, 17004 (2016). URL doi:10.1209/0295-5075/113/17004.
[24] Corzo, N. V. *et al.* Waveguide-coupled single collective

excitation of atomic arrays. *Nature* **566**, 359 (2019). URL doi:10.1038/s41586-019-0902-3.

[25] Liedl, C. *et al.* Observation of superradiant bursts in a cascaded quantum system. *Phys. Rev. X* **14**, 011020 (2024). URL doi:10.1103/physrevx.14.011020.

[26] Pasharavesh, A. & Bajcsy, M. Deterministic single photon subtraction with a cascade of waveguide-coupled atoms. *Opt. Express* **32**, 26740 (2024). URL doi:10.1364/oe.529257.

[27] Zubizarreta Casalengua, E., del Valle, E. & Laussy, F. P. Photon liquefaction in time. *APL Quant.* **1**, 026117 (2024). URL doi:10.1063/5.0206213.

[28] Khalid, S. & Laussy, F. P. Perfect single-photon sources. *Sci. Rep.* **14**, 2684 (2024). URL doi:10.1038/s41598-023-47585-9.

[29] Otterbach, J., Moos, M., Muth, D. & Fleischhauer, M. Wigner crystallization of single photons in cold Rydberg ensembles. *Phys. Rev. Lett.* **111**, 113001 (2013). URL doi:10.1103/physrevlett.111.113001.

[30] Sevinçli, S., Henkel, N., Ates, C. & Pohl, T. Nonlocal nonlinear optics in cold Rydberg gases. *Phys. Rev. Lett.* **107**, 153001 (2011). URL doi:10.1103/physrevlett.107.153001.

[31] Chang, D. E. *et al.* Crystallization of strongly interacting photons in a nonlinear optical fibre. *Nature Phys.* **4**, 884 (2008). URL doi:10.1038/nphys1074.

[32] Zeuthen, E., Gullans, M. J., Maghrebi, M. F. & Gorshkov, A. V. Correlated photon dynamics in dissipative rydberg media. *Phys. Rev. Lett.* **119**, 043602 (2017). URL doi:10.1103/physrevlett.119.043602.

[33] Ooi, C. H. R. & Scully, M. O. Two-photon correlation in a cascade amplifier: Propagation effects via a simple model, nonclassical regimes, and validity of neglecting langevin noise. *Phys. Rev. A* **76**, 043822 (2007). URL doi:10.1103/physreva.76.043822.

[34] Bienias, P. *et al.* Photon propagation through dissipative Rydberg media at large input rates. *Phys. Rev. Res.* **2**, 033049 (2020). URL doi:10.1103/physrevresearch.2.033049.

[35] Pegg, D. T., Loudon, R. & Knight, P. L. Correlations in light emitted by three-level atoms. *Phys. Rev. A* **33**, 4085 (1986). URL doi:10.1103/physreva.33.4085.

[36] del Valle, E., González-Tudela, A., Laussy, F. P., Tejedor, C. & Hartmann, M. J. Theory of frequency-filtered and time-resolved n-photon correlations. *Phys. Rev. Lett.* **109**, 183601 (2012). URL doi:10.1103/PhysRevLett.109.183601.

[37] Jenkins, A. Self-oscillation. *Phys. Rep.* **525**, 167 (2013). URL doi:10.1016/j.physrep.2012.10.007.

[38] Loudon, R. Non-classical effects in the statistical properties of light. *Rep. Prog. Phys.* **43**, 913 (1980). URL doi:10.1088/0034-4885/43/7/002.

[39] Burnham, D. C. & Weinberg, D. L. Observation of simultaneity in parametric production of optical photon pairs. *Phys. Rev. Lett.* **25**, 84 (1970). URL doi:10.1103/PhysRevLett.25.84.

[40] Kimble, H. J., Dagenais, M. & Mandel, L. Photon antibunching in resonance fluorescence. *Phys. Rev. Lett.* **39**, 691 (1977). URL doi:10.1103/PhysRevLett.39.691.

[41] Sánchez Muñoz, C. *et al.* Emitters of N-photon bundles. *Nature Photon.* **8**, 550 (2014). URL doi:10.1038/nphoton.2014.114.

[42] Bin, Q., Lü, X.-Y., Laussy, F. P., Nori, F. & Wu, Y. n-phonon bundle emission via the stokes process. *Phys. Rev. Lett.* **124**, 053601 (2020). URL doi:10.1103/PhysRevLett.124.053601.

[43] Moreau, E. *et al.* Quantum cascade of photons in semiconductor quantum dots. *Phys. Rev. Lett.* **87**, 183601 (2001). URL doi:10.1103/PhysRevLett.87.183601.

[44] Kiraz, A. *et al.* Photon correlation spectroscopy of a single quantum dot. *Phys. Rev. B* **65**, 161030(R) (2002). URL doi:10.1103/PhysRevB.65.161303.

[45] Akopian, N. *et al.* Entangled photon pairs from semiconductor quantum dots. *Phys. Rev. Lett.* **96**, 130501 (2006). URL doi:10.1103/PhysRevLett.96.130501.

[46] Zubizarreta Casalengua, E., del Valle, E. & Laussy, F. P. Two-photon correlations in detuned resonance fluorescence. *Phys. Scr.* **98**, 055104 (2023). URL doi:10.1088/1402-4896/acc89e.

[47] Hartmann, L., Dür, W. & Briegel, H.-J. Steady-state entanglement in open and noisy quantum systems. *Phys. Rev. A* **74**, 052304 (2006). URL doi:10.1103/PhysRevA.74.052304.

[48] Angelakis, D. G., Santos, M. F. & Bose, S. Photon-blockade-induced Mott transitions and XY spin models in coupled cavity arrays. *Phys. Rev. A* **76**, 031805 (2007). URL doi:10.1103/PhysRevA.76.031805.

[49] Greentree, A. D., Tahan, C., Cole, J. H. & Hollenberg, L. C. L. Quantum phase transitions of light. *Nature Phys.* **2**, 856 (2006). URL doi:10.1038/nphys466.

[50] Muth, D. & Fleischhauer, M. Dynamics of pair correlations in the attractive lieb-liniger gas. *Phys. Rev. Lett.* **105**, 150403 (2010). URL doi:10.1103/physrevlett.105.150403.

[51] Friedel, J. Metallic alloys. *Il Nuovo Cimento* **7**, 287 (1958). URL doi:10.1007/bf02751483.

[52] Schirmer, S. G. & Solomon, A. I. Constraints on relaxation rates for n-level quantum systems. *Phys. Rev. A* **70**, 022107 (2004). URL doi:10.1103/physreva.70.022107.

[53] Cheng, M. *et al.* Dynamics and the statistics of three-photon cascade emissions from single semiconductor quantum dots with pulse excitation. *J. Mod. Opt.* **53**, 2129 (2006). URL doi:10.1080/09500340600808915.

[54] Agarwal, G. S. Quantum beats in correlation functions of spontaneously emitted radiation and the transition rates of a multilevel atom. *Phys. Rev. A* **15**, 2380 (1977). URL doi:10.1103/physreva.15.2380.

[55] Lodahl, P. *et al.* Chiral quantum optics. *Nature* **541**, 473 (2017). URL doi:10.1038/nature21037.

[56] Ozawa, T. *et al.* Topological photonics. *Rev. Mod. Phys.* **91**, 015006 (2019). URL doi:10.1103/RevModPhys.91.015006.

[57] Sánchez Muñoz, C., Laussy, F. P., Tejedor, C. & del Valle, E. Enhanced two-photon emission from a dressed biexciton. *New J. Phys.* **17**, 123021 (2015). URL doi:10.1088/1367-2630/17/12/123021.