I. INTRODUCTION

Photonic[1] has been highly successful in the engineering of quantum sources since the proof-of-principle production of quantum light in the mid-seventies[2–4]. Befitting its status of the elementary brick of the light field, the single photon was the first type of genuinely non-classical type of light and gave rise to the concept of a single photon source (SPS)[5–9]. Nowadays, SPS abound with ever-increasing figures of merit[10–13] and are even commercially available. The motivations for SPS are many[14] from metrology[13,14] to input for quantum information processing, [15–17] passing by bio-technology, [18–20] sensing and detecting[21–26], etc. In most cases, the technology is still under development and quantum light is not yet deployed on the market. For this reason, the focus is still largely on the source itself rather than on its direct use as part of a technological component. There is however an increasing interest in using quantum light for actual applications. For instance, there have been recently converging propositions from independent groups to use quantum light for spectroscopy[22–26].

Thanks to the theory of frequency-resolved photon correlations[27], it was shown how quantum sources can be greatly tuned in their characteristics by selecting light in astute frequency windows, revealed by the theory in the form of so-called “two-photon correlations spectra”[28–32]. This can be used to identify and exploit unsuspected types of quantum correlations[33–36] and/or enhance them by learned combination of timescales and frequencies[37]. Merely by spectral filtering of a quantum emitter[38], beyond optimizing parameters to find the best compromises for sought applications, the theory also reveals that interesting quantum signal is typically not found at the expected spectral locations such as peaks, that correspond to classical-like de-excitation of the emitter between real states at the one-photon level. Instead, strong quantum correlations arise from, e.g., two-photon de-excitation involving intermediate virtual states. Such channels of de-excitation were termed “leapfrog processes”[39,40]. They are emitted in unremarkable frequency windows at the photo-luminescence level. At the quantum-optical level, however, they are the quintessence of quantum emission. This picture has been confirmed experimentally by A. Müller’s group[33] A closer look at, and proper selection of, the photons emitted by quantum sources thus appears fundamental for state-of-the-art quantum applications, their correlations being otherwise averaged over often competing types. This viewpoint of frequency-resolved photon correlations therefore poses in a new light the problem of the excitation of optical targets with such inside-knowledge of the features of the sources. This revives a question put to close scrutiny by Gardiner[34] and Carmichael[35] in 1993 following the emergence of sources of squeezed light[36] which appeared sufficiently more elaborate as compared to SPS to warrant a direct investigation of how they would affect optical targets as compared to conventional types of excitation (classical fields, possibly stochastic). Resonance fluorescence in the squeezed vacuum has in fact been just recently reported[37] Gardiner and Carmichael’s (independent) treatment of the problem of quantum excitation in two consecutive Letters in the Physical Review[38,39] achieved the setting up of a formalism—named the “cascaded formalism” by Carmichael—that allows to excite a system (which we will call the “target”) by an other (the “source”) without back-action from the target to the source. This permits to think separately of the quantum source, which properties can be first studied (through the two-photon spectrum, for instance) and then directed onto a target. For historical accuracy, let us mention that the problem was first contemplated by Kolobov and Sokolov[40] who tackled it by providing all the correlations of the exciting quantum field. This was recognized as an overkill by Gardiner and Carmichael[33,34] (Gardiner had made prior attempts along these lines). They proposed instead to model the quantum source dynamics as well as the response of the target, even if only the latter is of interest. From our point of view, such a treatment is essential since the frequency correlations of the source are too complicated to be treated otherwise than fully and explicitly by solving the complete problem. Also, such correlations are dynamical in character, and cannot

Exciting with Quantum Light. I. Exciting an harmonic oscillator.

J. C. López Carreña and F. P. Laussy

1 Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain
2 Russian Quantum Center, Novaya 100, 143025 Skolkovo, Moscow Region, Russia

(Dated: September 6, 2016)

We start a series of studies of the excitation of an optical target by quantum light. In this first part, we introduce the problematic and address the first case of interest, that of exciting the quantum harmonic oscillator, corresponding to, e.g., a single-mode passive cavity or a non-interacting bosonic field. We introduce a mapping of the Hilbert space that allows to chart usefully the accessible regions. We then consider the quantum excitation from single photon sources in the form of a two-level system under various regimes of (classical) pumping: incoherent, coherent and in the Mollow triplet regime. We close this first opus with an overview of the material to be covered in the subsequent papers.
be well approximated by quantum states as initial conditions. Instead, they must be dealt with through the full apparatus of dissipative quantum optics, feeding the target with the complete treatment of virtual states and other types of strongly correlated quantum input. The cascaded formalism is therefore particularly amenable for exciting optical systems with the knowledge of the two-photon correlations spectrum of an emitter.

Despite the conceptual importance of quantum excitation, there have been a moderate follow-up of this cascaded formalism, which we believe is a deep and far-reaching contribution to the problem of light-matter interaction. Even though it became textbook material (see the last chapter from one of the pioneering authors) and generated a sizable amount of citations, few texts do actually fully exploit the idea. Gardiner and Parkins (the formalism is sometimes also named after these two authors) undertook a more thorough analysis of various types of non-standard statistics of the source and used it to describe perfect transmission in their proposal for a quantum network but overall, the core of the literature using the formalism focuses on specialized particular cases, such as driving with squeezed light. Typically, the discussion is then held at the level of correlations from a quantum state (namely a squeezed state), as opposed to dynamical correlations from a quantum source. The other studies, already evoked, turned to approximate or indirect approaches, quite similar to the earlier attempts before the cascaded formalism was set up. The reasons for this is certainly a mix between convenience of using well-known and established formalisms and the as-yet unclear advantages of the alternative one.

In this series of texts, we make an extensive study of exciting with quantum light (with no feedback of the target to the source), and show that some new features of light-matter interaction emerge, making the overall problem in need of scrupulous attention. The formalism itself needs little further development and we will mainly adapt it to new cases but with the additional knowledge provided by frequency-resolved correlations (we will extend the formalism in the following papers to consider sequences of cascades and multiple sources). We briefly introduce its core machinery in a self-contained way in Section as a convenience for the reader, and refer to the original works for details of the derivation or to the Supplementary Material of Ref. where it is cast in the problematic of the present text. Section gives an overview of the many possibilities one can study as well as details of the configurations we focus on in the following of the series. After what can be seen as a long introduction, Section introduces the first important results by providing a way to characterize quantum states of the Harmonic oscillator in a space of quantum-optical diagonal correlators, which will be helpful to later characterize the sources, and with effect to disprove a popular criterion for single-photon states. Section and characterize the incoherent and coherent SPS, respectively. As the latter will prove to be more interesting, we devote most of our attention to this case. Section shows that the advantages of the cascaded coupling over the conventional Hamiltonian coupling (cavity QED) remain present even when considering alternative descriptions of coupling between the source and the target. Section draws the conclusions for the cases studied here while Section does so for the wider picture of cascaded coupling and introduces the other cases to be investigated in follow-up papers.

II. THEORETICAL BACKGROUND

The coupling between two quantum systems is typically given by an interaction Hamiltonian. In the second quantization formalism, such a Hamiltonian reads in its
most simple form (we take $\hbar = 1$ along the paper):

$$H_I = g c_i^\dagger c_2 + c_2^\dagger c_1,$$  \hspace{1cm} (1)

where $c_1$, $c_2$ are annihilation operators describing the particles of the coupled system, and $g$ is their interaction strength. If the particles described by $c_i$ have a decay rate $\gamma_i$, and are freely evolving with a Hamiltonian $H_i$, $i = 1, 2$, the master equation describing the dynamics of this system is:

$$\partial_t \rho = i[\rho, H_1 + H_2 + H_I] + \frac{\gamma_1}{2} L_{c_1} \rho + \frac{\gamma_2}{2} L_{c_2} \rho,$$  \hspace{1cm} (2)

where $L_{c_i} \rho = (2 c_i c_i^\dagger - c_i^\dagger c_i - \rho).$ Depending on whether the coupling $H_I$ or the dissipation $\gamma_i$ dominates, one speaks of strong or weak coupling, respectively. If one of the two systems, say, 1, is itself excited externally, for instance being driven by a laser, or merely being given a non-vacuum initial state, then it has a crucial picture of system 1 exciting system 2. This is a fairly accurate description in the weak-coupling limit where the dynamics seems irreversible, simply because excitations are dissipated before they can cycle back (we discuss in Section VII when this becomes exact).

The coupling between quantum systems does not have to be reversible: it can instead correspond to the scenario of a source and its target. In this case, there is a deep asymmetry between the coupled systems. For instance, one can remove the target from the beam of the source, which leaves the latter unaffected while the former passes from being irradiated to the vacuum. Note that such an asymmetry does not have to hold on logical grounds. In fact, in electronics, while an ideal source should not be affected by the circuit it powers, in reality, there is a load and every component affects all the others to some extent. In photonics, the picture of a flying qubit, left to propagate long enough before it meets its target, makes it intuitively clear that it should be possible to forbid back-action. This could also be realized by taking advantage of the fast-growing field of chiral optics.\cite{18181818, 18181818} Theoretically, this asymmetry is achieved by the cascaded coupling\cite{18181818}, where the equations of motion are expressed in the quantum Langevin form, thus allowing to set the output field of one of the systems (the source) as the input field for the other (the target). This can be brought to a master equation type of description, with both coherent and Lindblad terms that contrive to direct the flow of excitation from the source to target only. This makes all the operators of the source independent from those of the target, while in turn those depend on operators of the source. The generic case where the source (resp. target) is described by the Hamiltonian $H_1$ (resp. $H_2$) and has a decay rate $\gamma_1$ (resp. $\gamma_2$) is ruled by the following master equation:\cite{18181818}

$$\partial_t \rho = i[\rho, H_1 + H_2] + \frac{\gamma_1}{2} L_{c_1} \rho + \frac{\gamma_2}{2} L_{c_2} \rho -$$

$$- \sqrt{\gamma_1 \gamma_2} \left[ (c_2^\dagger c_1) \rho + [\rho c_1^\dagger, c_2] \right],$$  \hspace{1cm} (3)

The source must also be excited, which can be done either by an incoherent or by a coherent (classical) type of excitation. The incoherent excitation is described simply by adding the Lindblad term ($P_{c_1}/2) L_{c_1} \rho$ to Eq. (3). The coherent excitation, however, requires a subtler description, for which one uses the input–output formalism. The coupling between a coherent field and the system (that latter will be used as the source of quantum excitation) happens through an input channel for the said system. If such an input channel is the only one available to excite the source, then it follows that the only output channel from the source also contains the coherence of the driving field. In this case, the target of the quantum excitation is also driven by the coherent field, and its dynamics is given by Eq. (3), setting $H_1 = \omega_1 c_1^\dagger c_1 - i \sqrt{\gamma_1 \gamma_2} (c_1^\dagger - c_1),$ and $H_2 = \omega_2 c_2^\dagger c_2 - i \sqrt{\gamma_2} (c_2^\dagger - c_2),$ i.e., the dynamics is ruled by the master equation:

$$\partial_t \rho = i \left[ \rho, \omega_1 c_1^\dagger c_1 + \omega_2 c_2^\dagger c_2 - i \sqrt{\gamma_1 \gamma_2} (c_1^\dagger - c_1) - \right.$$

$$\left. - i \sqrt{\gamma_2} (c_2^\dagger - c_2) \right] + \frac{\gamma_1}{2} L_{c_1} \rho + \frac{\gamma_2}{2} L_{c_2} \rho -$$

$$\sqrt{\gamma_1 \gamma_2} \left( [c_2^\dagger, c_1] \rho + [\rho c_1^\dagger, c_2] \right),$$  \hspace{1cm} (4)

where $\mathcal{E}$ is the amplitude of the coherent field driving the source. Note that the effective driving intensity, i.e., $\Omega_1 = \sqrt{\gamma_1 \gamma_2}$, depends on the decay rate of the system that is being excited, in agreement with the fact that a system that cannot emit cannot be excited either. To prevent the target to be also driven by the coherent field, one can use other input (and their corresponding output) channels to excite the source (and also the target). Each of these channels couples with an amplitude $\epsilon_k \leq 1$, with the condition that $\sum_k \epsilon_k = 1$, the sum being over all the input channels. In this case, and considering only two input channels (with amplitudes $\epsilon_1$ and $\epsilon_2 = 1 - \epsilon_1$) as well as only one input channel for the target (with amplitude 1), the dynamics of the system is given by Eq. (3) with $H_1 = \omega_1 c_1^\dagger c_1 - i \sqrt{\gamma_1 \gamma_2} (c_1^\dagger - c_1),$ $H_2 = \omega_2 c_2^\dagger c_2,$ and replacing the coupling strength $\sqrt{\gamma_1 \gamma_2}$ by $\sqrt{(1 - \epsilon_1)} \gamma_1 \gamma_2$ in the second line of Eq. (3), i.e., the dynamics is now ruled by the master equation:

$$\partial_t \rho = i \left[ \rho, \omega_1 c_1^\dagger c_1 + \omega_2 c_2^\dagger c_2 - i \sqrt{\gamma_1 \gamma_2} (c_1^\dagger - c_1) \right] +$$

$$+ \frac{\gamma_1}{2} L_{c_1} \rho + \frac{\gamma_2}{2} L_{c_2} \rho - \sqrt{\epsilon_2} \sqrt{\gamma_2} \left( [c_2^\dagger, c_1] \rho + [\rho c_1^\dagger, c_2] \right),$$  \hspace{1cm} (5)

where $\epsilon_2 = 1 - \epsilon_1$. The additional input channel to the source makes the coupling between the coherent field not as efficient as when there is only one input channel, thus reducing the effective driving intensity. For the target, although the coupling strength is also reduced, now the driving is uniquely due to the emission from the quantum source.

Putting Eq. (3) in the Lindblad form contributes a Hamiltonian part. The formalism thus corresponds to a
quantum coherent coupling, allowing the description of continuous wave (cw) and resonant excitation of quantum states. Importantly, in contrast to the Hamiltonian coupling in Eq. (1), the coupling strength is now fixed by the decay rate of the source and of the target. An infinitely-lived target cannot be excited. The stronger one wishes to make the coupling between a source and its target, the stronger has to be their (geometric) mean dissipation. This imposes some fundamental constraints on external driving (or driving without feedback). In contrast, Hamiltonian coupling sets the coupling strength and decay rates independently. While it would therefore appear that the Hamiltonian coupling has the upper hand, and that one should strive for the standard strong-coupling regime, we will show in the following that the cascaded architecture can be superior to the other types of coupling in some cases.

III. QUANTUM SOURCES AND OPTICAL TARGETS

The general problem of excitation with quantum light has obviously numerous ramifications. To the already large variety of optical targets, one now has to combine a much enlarged set of quantum sources. This literally opens a new dimension to optics. Indeed, classical excitation could arguably be limited to a rather small set of categories:

- (a) Coherent excitation (driving with a laser)
- (b) Incoherent excitation (incoherent pumping, Boltzmann dynamics, thermal baths, equilibrium, etc.)

(we postpone to part V the discussion of time-dependent and pulsed excitation and consider until then the case of continuous wave excitation, cf. Section IX). Quantum light, on the other hand, encompasses not only the above—if only because it is a more general case that includes classical excitation as a particular case—but also comes with many more and higher degrees of freedom. This did not lead so far, to the best of our knowledge, to a classification. Tentatively, this could be provided in a first approximation by $g^{(2)}$ (antibunching, uncorrelated, bunching, superbunching). Since squeezed states have the correlations of coherent states, however, and they are precisely the type of input that motivated a new formalism, it is clear that this is still far from appropriate. We decided to approach this general problem by considering:

- the same optical target (here an harmonic oscillator, in the next paper a two-level system),
- the same type of quantum source (here and in the next paper, a SPS, in the third paper, an $N$-photon emitter),
- both coherent and incoherent regimes,
- both low and high pumping regimes.

The SPS is a good starting point because it is the paradigm of quantum light and is the most common light of this type in the laboratory. Exciting the harmonic oscillator makes the problem both simple and fundamental, so this is also a good starting point. While we have considerably restrained the possibilities, there still remains much to be explored and the following will only address the main results. The two types of excitations aforementioned (a–b) yield quite different types of SPS, that are introduced in the beginning of their respective Sections, and are summarized in Table I. Note that in the table, the last row also includes the second one (which is the limit of small pumping). They are however so distinct qualitatively that it is helpful to think of them as separate sources. As we will show, the second row (small coherent pumping) leads to the best antibunching in the target.

IV. CHARTING THE HILBERT SPACE

Prior to considering which quantum states of the harmonic oscillator one can access by exciting it with various types of SPS, one needs a roadmap to characterize all of the states at a glance. The Hilbert space is a big place. Human’s capacity of abstraction gives a deceivingly simple picture of it. The canonical basis of Fock states $|n\rangle$ with $n \in \mathbb{N}$ provides a comprehensive and concise map of the states in the harmonic oscillator’s Hilbert space $\mathcal{H}_{\infty}$:

$$|\psi\rangle = \sum_{n=0}^{\infty} \alpha_n |n\rangle ,$$

(6)

with $\alpha_n \in \mathbb{C}$ such that $\sum_{n=0}^{\infty} |\alpha_n|^2 = 1$. This is as precise as it is misleading, since this fails to provide a qualitative map of the possible states. Instead, one uses families of particular cases. Beyond Fock states per se, one makes a great use of the coherent states of classical physics, for which $\alpha_n = \alpha^n \exp(-|\alpha|^2)/\sqrt{n!}$ for some complex number $\alpha$ that defines the amplitude and phase of a classical field. The next family of important states requires the concept of micity that involves statistical averages and upgrades the wavefunction to a density matrix:

$$\rho = \sum_{n,m=0}^{\infty} \beta_{nm} |n\rangle \langle m| .$$

(7)

This allows the introduction of thermal states, for which $\beta_{nm} = (1 - \theta)\theta^n \delta_{nm}$ for some reduced temperature $\theta \in [0,1]$. These two families can be united into a larger class of “cothermats” states that interpolate between the two and describe, e.g., single-mode Bose condensates not too far from threshold. From there on, one essentially deals with quantum states of light with popular examples such as squeezed states, cat states, etc., and other less popular such as binomial states or displaced Fock states. Such a zoology of states is familiar to every quantum physicist, but it fails to provide the sought mapping of
the Hilbert space. In particular, it is restricted to known (and popular) cases and leaves most of Eqs. (6–7) uncharted.

We introduce a map of the Hilbert space based on important observables for the quantum state, namely, the $n$-th order correlation functions:

$$g^{(n)} = \frac{\langle a_n^\dagger a_n \rangle}{\langle a^\dagger a \rangle^n}.$$  \hspace{1cm} (8)

Since we consider only the quantum states and not their dynamics, normalization makes $g^{(1)}$ trivially unity. We therefore use instead the normalization itself, which is an important observable, the population of the state (average number of excitations), for which we introduce the notation:

$$n_a \equiv \langle a^\dagger a \rangle.$$ \hspace{1cm} (9)

Note that only the diagonal elements $p(m) = |a_m|^2$ are needed so there is no distinction between pure and mixed states in this discussion. The $n$-th order correlator reads:

$$g^{(n)} = \frac{\sum_{m=0}^\infty m!p(m)/m!}{\sum_{m=0}^\infty m!p(m)/m!}.$$ \hspace{1cm} (10)

A convenient mapping of the Hilbert space is to chart it with the $n_a$ and $g^{(2)}$ “rulers”, that is, tag the possible quantum states through their joint statistical properties and population. The value of $g^{(2)}$ allows to tell apart classical from quantum states depending on whether $g^{(2)}$ is larger (bunching) or smaller (antibunching) than unity. This is meant in the sense of whether a classical, possibly stochastic, description is possible, or whether some “quantum” features such as violation of Cauchy–Schwarz inequalities or negative probabilities in the phase space are manifest. The population gives another meaning to quantumness, in the sense of few-particle effects vs macroscopic occupation. It is therefore particularly interesting to contrast these two attributes and consider, e.g., highly occupied genuinely quantum states. There is a physical limit to such states and for populations $n_a > 1$, some values of antibunching are out of reach. This is expected on physical grounds from familiar features of the Fock states popularly known as being the “most quantum” states, with their $g^{(2)}$ tending to one as the number of excitations increases. Also, macroscopic quantum states, such as a BEC, are essentially coherent states in most formulations. Here we make this notion precise by providing the complete picture, in Fig. 2 along with the closed-form expression for the boundary that separates accessible combinations from impossible ones (this is proved in Appendix A):

$$g^{(2)} = \frac{n_a}{n_a^2}.$$ \hspace{1cm} (11)

This boundary is provided by superpositions of contiguous Fock states, i.e., of the type:

$$\sqrt{p(n)} |n\rangle + \sqrt{1 - p(n)} e^{i\theta} |n + 1\rangle,$$ \hspace{1cm} (12)

for $n \in \mathbb{N}$, $p(n) \in [0, 1]$ and $\theta \in [0, 2\pi]$ an (irrelevant) phase. (As already commented, this includes also mixed states of the type $p(n) |n\rangle + (1 - p(n)) |n + 1\rangle$ and all others with the same diagonal but different off-diagonal elements. We will not make this distinction anymore in the following.) We call these states, Eq. (12), “Fock duos”. They set the continuous lower limit of the space of available quantum states in our charted Hilbert space. Their antibunching (11) is a generalization to non-integer $n_a$ of the formula $g^{(2)} = 1 - 1/n_a$ for the Fock state $|n\rangle$ to which it reduces for $n_a \in \mathbb{N}$. It shows that the popular “single-particle criterion” that asserts that $g^{(2)} < 0.5$ ensures a one particle state [61, 62] is wrong, as demonstrated, e.g., for the case $n_a = 1.5$ for which $g^{(2)} = 4/9 \approx 0.44$. The generalization to higher orders is straightforward. Namely, the $n$th-order correlation frontier for real $n_a$ is given by:

$$g^{(n)} = \frac{|n_a|!}{(|n_a| - n)!n_a^n} \left[ 1 + \frac{n(n_a - |n_a|)}{|n_a| + 1 - n} \right],$$ \hspace{1cm} (13)

that generalizes the Fock state formula $g^{(n)} = (1/n_a^n)m!/(m! - n)!$ valid for integer $n_a$ where it agrees with Eq. (14). Interestingly, the lower limit is also set, for all $n$, by Fock duos (not “n-plets”).

One can also consider other charts of the Hilbert space, such as $(g^{(2)}, g^{(3)})$, this time contrasting two- and three-particle correlations together. This time there are no boundaries in this space (one can find state with any joint values of $g^{(2)}$ and $g^{(3)}$). The proof of this statement is given in Ref. [62]. Since the excitation of an harmonic oscillator by the SPS leads to strong correlations between various $g^{(n)}$, we will focus on the $(n_a, g^{(2)})$ space.

| Source | Pumping | Population $n_a$ | $g^{(2)}(\tau)$ | Lineshape | Linewidth |
|--------|---------|------------------|-----------------|-----------|-----------|
| Incoherent SPS | $P_n$ | $P_n/(\gamma_n + P_n)$ | $1 - \exp \left(-\gamma_n P_n/\gamma_n P_n\right)$ | Lorentzian | $\gamma_n + P_n$ |
| Coherent SPS | small $\Omega_\sigma$ | $4\Omega_\sigma^2/\gamma_\sigma^2$ | $\left(1 - \exp(-\gamma_n \tau/2)\right)^2$ | Lorentzian | $\gamma_\sigma$ |
| Coherent SPS | large $\Omega_\sigma$ | $4\Omega_\sigma^2/\gamma_\sigma^2 + 8\Omega_\sigma^2$ | $1 - e^{-\gamma_\sigma^2/8\Omega_\sigma^2}$ | Triplet | $\gamma_\sigma$ (central) | $3\gamma_\sigma^2/2$ (satellites) |

TABLE I. Characteristic of a SPS driven incoherently or coherently. In the latter case, for clarity, we also consider separately the low-driving intensity (middle row) from the general case (bottom row) that covers all pumpings, to highlight the strong qualitative features of the Mollow regime.
from Eqs. (14), this gives the equation for the trajectory and the emission rate of the 2LS, system: i) the ratio between the decay rate of the cavity density (or pumping rate) of the 2LS, $P$, incoherently, as sketched in Fig. 1(c). Namely, there is a level system (2LS) that acts as the source, is itself driven excitation with an incoherent SPS, i.e., where the two-level system (2LS) is put in its excited state, and is otherwise left to decay. The system is thus described by the master equations (2–3), with $c_1 = \sigma$ the 2LS and $c_2 = a$ the harmonic oscillator operators, respectively. The 2LS pumping is described by a Lindblad term $(P_\sigma/2)\mathcal{L}_{\sigma \rho}$. Thanks to the absence of feedback, the dynamics is ruled by closed equations which allows us to obtain exact solutions for the observables of interest. Namely, we find that the cavity population $n_\sigma$ and statistics $g^{(2)}$ of the target are given by (cf. Table I for the source):

$$n_\sigma = \frac{4P_\sigma \gamma_\sigma}{(\gamma_\sigma + P_\sigma)(\gamma_\sigma + P_\sigma + \gamma_\sigma)}, \quad (14a)$$

$$g^{(2)} = \frac{2(\gamma_\sigma + P_\sigma)}{\gamma_\sigma + P_\sigma + 3\gamma_\sigma}. \quad (14b)$$

Only two parameters are required to describe fully the system: i) the ratio between the decay rate of the cavity and the emission rate of the 2LS, $\gamma_\sigma/\gamma_\sigma$, and ii) the intensity (or pumping rate) of the 2LS, $P_\sigma$, also normalized to $\gamma_\sigma$ to keep the parameters dimensionless. Eliminating $P_\sigma$ from Eqs. (14), this gives the equation for the trajectory in the $(n_\sigma, g^{(2)})$ space as function of the parameter $\gamma_\sigma/\gamma_\sigma$:

$$n_\sigma = \frac{2}{3} \frac{(2 - g^{(2)})(3g^{(2)}(\gamma_\sigma/\gamma_\sigma) - (2 - g^{(2)}))}{g^{(2)}(1 + g^{(2)})(\gamma_\sigma/\gamma_\sigma)}. \quad (15)$$

V. EXCITING WITH AN INCOHERENT SPS

Now that we can conveniently characterize the states of the harmonic oscillator that one can excite, we come back to the dynamical problem of the quantum driving of an harmonic oscillator and how close can one get to the limit set by Eq. (11). We first consider the case of an harmonic oscillator and how close can one get to the ideal antibunching, which is zero. All the states, and only these states, above this line and below $g^{(2)} = 2$, are accessible with an incoherent SPS.

These trajectories in the Hilbert space are plotted in Fig. 3. The curves start from the point:

$$(n_\sigma = 0, g^{(2)} = 2 \frac{2}{1 + 3\gamma_\sigma/\gamma_\sigma}), \quad (16)$$

at vanishing pumping, reach a turning point with coordinates:

$$(n_\sigma = \frac{4(2 + (\gamma_\sigma/\gamma_\sigma) - 2\sqrt{1 + (\gamma_\sigma/\gamma_\sigma)})}{(\gamma_\sigma/\gamma_\sigma)^2}, \quad g^{(2)} = \frac{2}{3\sqrt{1 + \gamma_\sigma/\gamma_\sigma} - 2}) \), \quad (17)$$

when $P_\sigma = \sqrt{\gamma_\sigma(\gamma_\sigma + \gamma_\sigma)}$ and converge at $(n_\sigma = 0, g^{(2)} = 2)$ at large pumpings, where the source gets quenched. For each value of $\gamma_\sigma/\gamma_\sigma$, there are two values of pumping that result in the same population but two values of $g^{(2)}$. The curve of Eq. (15) is fairly constant till the turning point and, from Eq. (16), leads to a genuine quantum state (i.e., featuring antibunching) as long as $\gamma_\sigma/\gamma_\sigma > 1/3$, which means that, with a SPS, one can imprint antibunching in a system that has a substantially longer lifetime. The optimum antibunching/population for a given $\gamma_\sigma/\gamma_\sigma$ is achieved slightly earlier than the turning point, namely, when $P_\sigma = \gamma_\sigma$. The envelope of all the curves in Fig. 3 is the closest one can get to the Fock-duos limit, and is given by $g^{(2)} = 2n_\sigma/(3 - 2n_\sigma)$. This is the thick dark green line in Fig. 3.

The above solution gives an already substantial description of the response of an harmonic oscillator to an incoherent SPS. We can complement it with alternative descriptions that approach the solution from different viewpoints and manifest the advantage of cascaded coupling over other types of excitation. A natural comparison is with the standard Hamiltonian coupling, that...
is the usual way to couple different systems. For instance, it is convenient to lay out the observables as function of the parameters in 2D density plots, as shown in the upper row of Fig. 4. Panels (a–c) show the photon statistics (color code) and the population of the cavity (isolines) using the (a) cascaded coupling and (b, c) the Hamiltonian coupling. For the later case, we consider two configurations: (b) when the 2LS has the same decay term as when acting as a source in the cascaded scheme, and (c) when the 2LS has no decay term, so that all its excitation is directed towards the cavity, corresponding to the ideal excitation of the cascaded scheme. Interestingly, (a) and (b) types of excitation present a qualitatively similar photon statistics layout, but the topography of the resulting population yields largely differing states, as evident once the (\( \sigma_n, \sigma_g^{(2)} \)) space, panel (d). The color circles along the isolines mark the points with strongest antibunching (i.e., smallest \( g^{(2)} \)) for a given population. It is seen that for a given signal, the antibunching is significantly larger with the cascaded coupling. In both cases the antibunching is marred before the cavity population reaches one photon on average. At larger populations, the photon statistics of the cavity become bunched, i.e., \( g^{(2)} = 2 \). On can contemplate other configurations but they result in worse results: detuning the cavity from the 2LS leaves the photon statistics unchanged, but reduces the population of the cavity, so the antibunching in lost at even smaller cavity populations. Dephasing the 2LS increases the broadening of the emission peak, but also drives the cavity towards a coherent state faster. Hamiltonian coupling with no decay of the 2LS, case (c), differs qualitatively from the former cases and provides a substantially better antibunching than Hamiltonian coupling with decay of the 2LS. It even becomes better than the cascaded coupling when \( n_a \gtrsim 2/3 \). This situation can be much improved with the coherent SPS.

VI. EXCITING WITH A COHERENT SPS

We now consider excitation by a coherent SPS, i.e., when the 2LS that acts as the source is driven coherently by an external laser, as sketched in Fig. 4(d). This driving is described by the Hamiltonian:

\[
H_\sigma = \omega_\sigma \sigma^\dagger \sigma + \Omega_\sigma \sqrt{\epsilon_1} (\sigma^\dagger e^{-i\omega_L t} + \sigma e^{i\omega_L t}),
\]

where \( \omega_\sigma \) is the energy of the 2LS, and \( \omega_L \) and \( \Omega_\sigma = \sqrt{\gamma_\sigma \epsilon L} \) are the frequency and intensity of the driving laser (which drives a 2LS of decay rate \( \gamma_\sigma \) with a coherent field of intensity \( \epsilon_1 \)), respectively. The coefficient \( \sqrt{\epsilon_1} \) is put here so that the cavity is driven only by the emission of the 2LS and not by the external laser. The system is otherwise still described by the master equations (2) and (5), with \( c_1 = \sigma \) the 2LS (source) and \( c_2 = \hat{a} \) the harmonic oscillator (target), and reducing the coupling strength by a factor \( \sqrt{1-\epsilon_1} \), i.e., the coupling is given by \( \sqrt{1-\epsilon_1} \gamma_\sigma \sigma^\dagger \). Here again, one can obtain the cavity observables in closed-form, although it takes slightly more cumbersome expressions. At reso-
FIG. 5. (Color online). Same as Fig. 3 but for the excitation from a coherent SPS. In (a,c) the implicit parameter is pumping and the green line shows the closest one can get from the Fock-duos limit from the case at resonance. In (d) the implicit parameter is the detuning between the source and its target. (a) Exciting at resonance. The optimum antibunching is the envelope in this configuration of the family of curves varying with $\gamma_a/\gamma_\sigma$. Highlighted are the cases $\gamma_a/\gamma_\sigma = 10^{-2}$ (blue), $10^{-1}$ (green), 1 (chartreuse), 10 (yellow) and $10^2$ (red). (b-d) Exciting in other configurations, namely (b) with the leapfrog processes, (c) with the satellite and (d) as a function of frequency. Highlighted are the cases $\gamma_a/\gamma_\sigma = 10^{-1}$ (blue), 0.5 (chartreuse), 1 (green), 5 (orange) and 10 (red). The dashed-dotted line in panel (b) and the dashed line in panel (c) show the limit of large intensities and are merely a visual aid rather than a physical boundary. They are also shown in panel (d), along with the black solid line for the envelope of the curves at $\omega_a = \omega_\sigma$.

The PL of the source is still a single line as shown...
in the bottom inset, while the upper branch corresponds to high pumping where the PL has split into a Mollow triplet, shown in the upper inset. Each curve starts from the point:

\[
    n_a = 0, g^{(2)} = \frac{1}{(1 + \gamma_a/\gamma_\sigma)^2},
\]

showing that coherent SPS provide a much stronger antibunching than their incoherent counterpart, Eq. (16), already at vanishing pumping. At high pumping, in contrast to the incoherent case that vanishes the population of its target, the coherent SPS quenches it to a nonzero value and featuring bunching:

\[
    n_a = \frac{1 - \epsilon_1}{1 + \gamma_a/\gamma_\sigma}, g^{(2)} = \frac{3 + \gamma_a/\gamma_\sigma}{1 + 3\gamma_a/\gamma_\sigma}.
\]

Eliminating \(\gamma_a/\gamma_\sigma\) here leads to the curve \(g^{(2)} = 3(1 - \epsilon_1)/[3(1 - \epsilon_1) - 2n_a]\), which for \(\epsilon_1 = 1/2\) simplifies to \(g^{(2)} = 3/(3 - 4n_a)\) as shown in thick black in Fig. 5(a). The expression for the lower envelope of maximum antibunching, in blue, is too complicated to be derived fully in closed-form but we can find simple asymptotic expressions by series expansion of Eqs. (19), namely, \(g^{(2)} \approx 5n_a^2\) when \(n_a < 1\) and \(g^{(2)} \approx 1 - 1/(3(n_a + 5))\) when \(n_a \gg 1\). The states thus get close to the ideal Fock limit for large populations, approaching unity as \(1/(3n_a)\) rather than \(1/n_a\). There is no limit in the population that can be excited in the target by the coherent SPS, in stark contrast to the incoherent SPS that is bounded by unity. Although the accessible states in the \((n_a, g^{(2)})\) space remain quite some distance away from the ideal limit, the coherent SPS provides a much better antibunching than its incoherent counterpart, as can be seen by comparing Figs. 3 and 5. Namely, it is still antibunched when population is unity (with a maximum antibunching slightly over 1/2) and we have already commented how it allows for arbitrary large populations, that still feature antibunching. This is achieved by exciting at resonance targets of very long lifetimes as compared to the source.

This describes the resonant situation. Since the coherent SPS has a rich spectral structure, it opens new configurations of excitation in the Mollow triplet regime beyond the central peak, such as exciting with a satellite peak (as shown in Fig. 5(c)) or with the leapfrog window (Fig. 5(b)). In these cases we show the results in log-log plots as only small populations are within reach (unlike the case of resonance) and, in the case of leapfrog excitation, also a huge bunching can be imparted to the target. The envelope of optimum antibunching as obtained in resonance is reported in these panels (also in green) for comparison, showing that these alternative schemes do not enhance antibunching. The excitation with leapfrog processes allows to access new regions of the Hilbert space related to superbunching, so it is a configuration that presents its own interest. This is achieved at the price of small populations. Exciting with a satellite peak also conquers new territories in the \((n_a, g^{(2)})\) space not accessible through either the central peak or leapfrog emission in a small region \((n_a \approx 0.6, g^{(2)} \lessgtr 2)\). Panel (d) shows the trajectories when varying the frequency of emission, where the cases just discussed—resonance, leapfrog and satellites—appear as boundaries of the complete picture, thus showing that these configurations already give access to all the accessible states.

We can also compare in the parameter space the coherent SPS with its Hamiltonian counterpart, as we did for the incoherent SPS. This is shown in the bottom row of Fig. 4 that is also usefully compared with the upper row (incoherent SPS excitation). For the coherent SPS as well, both cascaded and Hamiltonian coupling are qualitatively similar in their statistical layout when the 2LS has a decay term, but also differ notably from the topology of the associated intensities. This is, again, clear on the \((n_a, g^{(2)})\) space, with the maximum antibunching that can be obtained for a given cavity population with cascaded coupling (green line) and Hamiltonian coupling (red line). The Hamiltonian coupling with no decay of the 2LS also results in qualitative differences and an enhanced antibunching. However, in this case, the Hamiltonian coupling never comes to surpass the cascaded scheme. Instead, in all the range of population, the antibunching obtained through the cascaded coupling is larger than the one obtained with any type of normal (Hamiltonian) coupling, for a given population. When the cavity is detuned from the 2LS, the antibunching is lost before the cavity’s population reaches one photon. Also, when the cavity is in resonance with the leapfrog process emission, the photon statistics of the cavity is superbunched but its population remains well below unity. Including a dephasing rate to the dynamics of the 2LS increases the broadening of each emission line in the triplet, and even a small dephasing rate spoils the antibunching. Thus, the best antibunching with a coherent SPS is obtained exciting the cavity at low pumping and in resonance with the 2LS, using the cascaded coupling.

One of the departing features of the coherent SPS excitation, is that it allows to populate its target with more than one excitation on average, therefore triggering Bose stimulation effects. There is a phase locking (of 0, as determined by the Hamiltonian) of the incident photons from the quantum source that accumulate while still exhibiting some features of Fock states, in particular being antibunched. A complete picture of the resulting quantum states, beyond the diagonal elements only, is given Fig. 6 for the states marked with black circles in Fig. 4(f), both through the Wigner representation, Fig. 6(a-c), and through their matrix representation, Fig. 6(d-f). The Wigner representation exhibits negative values at the origin, a mark of a genuine quantum state with no classical counterpart, for the states with \(n_a = 1\) and \(n_a = 1.5\) (the blue spot at the center in Fig. 6(b)). It is, however, positive everywhere for \(n_a = 3\), although the state is still antibunched. Also, one can see that the phase uncertainty decreases as the population increases, in agreement with the “classical limit”. Moreover, the matrix elements in these cases also satisfy the
relation $\rho_{22} > \max(\rho_{11}/2, \rho_{33})$, which means that fluctuations are smaller than could be expected from rate equation arguments, since two excitations are twice as much likely to decay than one excitation, making the probability to find one excitation only in principle at least twice as large than double occupation.

We conclude this Section on driving a harmonic oscillator with the coherent SPS by considering states in the $(g^{(2)}, g^{(3)})$ space. This is shown in Fig. 7 for the most notable configurations of exciting with the central peak of the Mollow triplet or the leapfrog region between the central peak and a satellite. All points are physical on this map, including cases with $g^{(2)} \ll 1$ and $g^{(3)} \gg 1$ (and vice-versa), although the accessible regions are strongly confined along curves that we can fit in monomial form, leading in good approximation to $g^{(3)} \approx 0.2|g^{(2)}|^2$ at low pumping (in the antibunching corner) and $g^{(3)} \approx 4.5g^{(2)}$ at large pumpings (in the superbunching corner). We indicate positions of popular quantum states such as Fock states, coherent states, thermal states and their combinations. Driving with a SPS spans over great distances in this space, especially when exciting with leapfrog processes, as seen in panel (a) of the figure. This shows, again, how the central peak remains confined essentially to the antibunched corner of the map (excursions to bunching cases are up to $g^{(2)} = 3$). Panel (b) is a zoom around small values in linear scale. The leapfrog processes, on the contrary, allow one to reach large joint values of $g^{(2)}$ and $g^{(3)}$. Interestingly, while the points accessible in the $(n_a, g^{(2)})$ space extend over 2D areas, in the $(g^{(2)}, g^{(3)})$ space they are confined to 1D curves, showing how these correlators are strongly inter-dependent, and thus making the $(n_a, g^{(2)})$ representation more prominent.

VII. VARIATIONS IN THE TYPE OF COUPLING

The comparison between both rows of Fig. 4 shows that the coherent SPS is a much better quantum drive to generate bright antibunching than the incoherent one, even in the region where $n_a < 1$. The coherent SPS exciting the cavity still remains some distance away from the Fock duos boundary, but it gets significantly closer than with the Hamiltonian coupling. We complete our juxtaposition between the two types of coupling, cascaded and Hamiltonian, by comparing with other variations in the way the source couples to its target.

First, we consider another source of decay for the source in the cascaded coupling scheme, i.e., with an extra term $(\gamma^*_{a}/2)\mathcal{L}_\sigma\rho$ in the master equation, leaving the coupling strength constant and equal to $\sqrt{\alpha/\gamma_a}/2$. This describes the situation where not all the light that is lost from the source is redirected to the target. Figure 8-
Coherent excitation
Incoherent excitation

Source (when source) enhances the antibunching until it reaches the one state (for the coherent SPS) or to a thermal state (for the cascaded coupling) shows how this other source of decay spoils the antibunching. (c, d) Increasing the coupling strength can only be smaller or at most equal to $\gamma_{a}^{*}$, the antibunching is worse than that obtained using the Hamiltonian coupling (without any additional dissipative channel). This shows that it is better to minimize losses from the quantum emitter, but that unless these are drastic, the impact is moderately detrimental. Note that the cascaded coupling strength can only be smaller or at most equal to $\sqrt{\gamma_{a}^{*}/2}$, unlike some statements in the literature that refer to “arbitrary coupling strengths”. This leads, otherwise, to unphysical states for the target. Second, in the Hamiltonian coupling, we increase the coupling strength and bring the system into the strong-coupling regime. This is one considerable degree of freedom of the Hamiltonian coupling that is lost when considering some external excitation, since photons that drive the system from outside can couple to it in the first place because it is dissipative. In fact, the more efficient is the source–target coupling, the more dissipative the target has to be. But a dissipative target cannot sustain quantum effects for long, as the states do indeed decay. Therefore there is a compromise for the target’s lifetime to allow simultaneously an efficient driving from the source while still allowing to store long enough the imprinted quantum attributes. In the Hamiltonian coupling, however, coupling and decay are independent and one can consider a pure Hamiltonian picture with no decay whatsoever. We will see later in this series how one can remedy to this vicious circle to some extent by considering other types of quantum sources. For now, we show that, surprisingly, cascaded coupling indeed provides a better quantum driving than the Hamiltonian coupling brought in the strong-coupling regime. This is because in the latter case, the reversible process from the “target” back to the “source” acts like an effective decay. This is shown in Fig. 8(c,d), where we compare again the maximum antibunching obtained in the configuration of Sec. V(b) (solid green lines) with a magnified Hamiltonian coupling $g^{*} = N \sqrt{\gamma_{a}/\gamma_{a^{*}}}/2$ for various $N$. For both the coherent and incoherent excitation cases, the antibunching is improved by raising the coupling strength. However, this improvement is still not large enough to overcome the cascaded coupling. As seen in the figure, even for large Hamiltonian coupling strengths, the antibunching obtained with the cascaded coupling (solid green line) is always significantly better. Increasing even further the value of $N$ does not improve the antibunching, but makes it occur at larger ($\Omega_{a}, \gamma_{a}$) values. At this limiting case, the antibunching obtained for the Hamiltonian coupling with the decay of the source coincides with the antibunching for the Hamiltonian without the decay of the source. Therefore, Fig. 8(c) shows the optimum antibunching that can be obtained using both the cascaded and Hamiltonian couplings.

Increasing the coupling has a much stronger effect in the case of incoherent excitation, as shown in Fig. 8(d). Even a small variation of the coupling strength, $N = 1.5$, changes significantly the curvature of the antibunching curve. Increasing the value of $N$ makes the system enter the “one-atom laser” regime and the state in the cavity becomes coherent. As for the coherent excitation, the best antibunching saturates with increasing coupling (converging at roughly $g^{*} = 10g$), with larger values of the coupling strength merely occurring at larger ($P_{a}, \gamma_{a}$) values. Nevertheless, although raising the coupling strength improves the antibunching, and at large $g^{*}$ the $g^{(2)}$ of the cavity at large population goes to 1, the best antibunching is still obtained with the cascaded coupling in the coherent excitation regime (green line in Fig. 8[d]). Overall, it is therefore established that optimum antibunching is obtained in the cascaded architecture. This could have important consequences in the design of future quantum-optical devices.

FIG. 8. (Color online). Variations in the model. (a, b) Adding a dissipative channel to the source in the cascaded coupling spoils the antibunching. (c, d) Increasing the coupling strength $g$ in the Hamiltonian coupling (with decay in the source) enhances the antibunching until it reaches the one obtained for the Hamiltonian coupling without decay of the source (when $g^{*} \approx 10g$). However, the enhancement is never enough to top the antibunching obtained using the coherently driven 2LS and the cascaded coupling.

VIII. SUMMARY AND CONCLUSIONS

Our analysis of the excitation by a SPS of a quantum harmonic oscillator can be summarized by Fig. 8. Here we show again the charting of the Hilbert space of the quantum harmonic oscillator that we have introduced, along with the various areas that can be accessed with a SPS. First, the region I of physical states, shown in light blue, delimited by Fock duos (Eq. (12)) according to Eq. (11). No state exists that can provide the corresponding joint population and antibunching in the white region. In contrast to popular belief, states do exist in the region above this boundary that satisfy $g^{(2)} < 1/2$ and $n_{a} > 1$ which shows that on mathematical grounds, the criterion $g^{(2)} < 1/2$ cannot be used to exclude states with more than one particle. In this region of physi-
FIG. 9. (Color online). Summary of our results: the Hilbert space of an harmonic oscillator charted by the $n_a$ (population) and $g^{(2)}$ (second-order correlation function) is accessible for any point in the blue region I (the white region is forbidden). Of this region, the wider span is achieved by exciting with a coherent SPS, covering the green region. This region breaks down into II, the region achieved at resonance between the source and the target, III, that achieved with the excitation from a satellite of the Mollow triplet and IV that achieved with the leapfrog emission. The incoherent SPS covers a smaller region, delimited by the red dotted curve. The hamiltonian coupling, between the two dotted-dash yellow curves, also covers a large span of the Hilbert space and is neither included nor fully encloses the SPS area. In particular, the excitation by a SPS allows to provide a stronger antibunching for any given population as compared to a Hamiltonian coupling, although it still remains some distance away from the ideal limit.

FIG. 10. (Color online). Exciting with Quantum Light: the basic configurations we propose to study in this series of texts. Part I is the current work, exciting an harmonic oscillator (single cavity mode) with a SPS. Part II addresses the excitation of a 2LS with a SPS. Part III is exciting coupled light-matter systems, or polaritons, still with a SPS. Following the comprehensive treatments of these several targets under the cw excitation from a SPS, we then trade this simple quantum source for, IV, a $N$–photon emitter (bundler) and, V, a pulsed quantum source, impinging on all the targets previously considered.

IX. PERSPECTIVES

We have considered only one aspect of the general picture of quantum excitation of optical targets. Namely, various types of SPS exciting an harmonic oscillator. In the laboratory, this can take the form of exciting a passive cavity mode, or a non-interacting boson field. This is the configuration sketched in Fig. 10 I. and that we addressed in this text, the opening one of a series that will carry on similar studies to other configurations of interest, as sketched in the rest of Fig. 10 and summarized here. In the next text, II., we consider the problem...
of exciting a 2LS rather than the harmonic oscillator. In part III, we increase one step more the level of complexity of the target by exciting polaritons, i.e., coupled light–matter systems. This focuses on different targets subjected to the (quantum) light of the same type of source. In part IV, we revisit all these targets together but now excited by an even “more quantum” source than the SPS, namely, $N$-photon emitters. In Section V., we come back to all the combinations dealt with so far but turning to pulsed excitation rather than cw, addressing the problem of quantum state preparation. Even with the five texts taken together, we do not exhaust the topic and much remains to be studied even in the configurations put to scrutiny. For instance, we considered one particular type of SPS only, the simplest one as realized by a single emitter, while implementations from pairs of photons with one heralding the other (mainly through parametric down conversion (PDC) and four-wave mixing (FWM)) are extremely popular and with advantages of their own, such as determinism. It is clear that there is much engineering to be done with such sources as well, and certainly some improvements in their operating conditions to be gained from the cascaded architecture, e.g., helping to fix their principal drawback so far: the nonzero probability to emit more than a single photon. Also, this extended exploration of the general problem of quantum excitation will give us many occasions to revisit the same aspects from different angles. Already in part II, we will come back to the question of what regions of the Hilbert space are within reach. As another example, in part V, we will provide a clear physical picture of why the cascaded architecture allows such an enhancement as compared to the Hamiltonian coupling by considering the time-resolved excitation of the target by the spontaneous emission of an initial condition prepared in the source, which can be solved exactly. This brings much clarification into the nature of the target and the source and their interrelationship in a quantum context.

ACKNOWLEDGMENTS

We thank C. Sánchez Muñoz and E. del Valle for constant discussions. Funding by the POLAFLOW ERC project No. 308136 and by the Spanish MINECO under contract FIS2015-64951-R (CLAQUE) is acknowledged.

Appendix A: Proof that Fock duos bound the $(n_a,g^{(2)})$ space

We prove mathematically that there are no states below the boundary set by Eq. (11) but at least one for every point above, thus showing that our charting of the Hilbert space in the $(n_a,g^{(2)})$ space as defined by this boundary is complete.

Proposition: For a given population $n_a \in \mathbb{R}$, the maximum antibunching is given by a superposition of at most two consecutive Fock states. □

First, for any $n_a \in \mathbb{R}$, there exists a superposition of at most two consecutive Fock states that provides this population, namely:

$$\sqrt{|n_a|-n_a+1} |n_a\rangle + \sqrt{n_a-[n_a]} |[n_a]+1\rangle. \tag{A1}$$

We can write without loss of generality the generic state $|n\rangle$ as:

$$\sum_{k=-\lfloor n\rfloor}^{\infty} \sqrt{p([n]+k)} |[n]+k\rangle, \tag{A2}$$

where we remind that $p(n) = |\alpha_n|^2$ and is such that $\sum_{n=0}^{\infty} p(n) = 1$. Restricting ourselves to states with the same population implies:

$$\sum_{k=-\lfloor n\rfloor}^{\infty} p([n]+k)(|[n]+k) = n_a. \tag{A3}$$

The second order correlation function for such states is then found as:

$$g^{(2)} = \sum_{k=-\lfloor n\rfloor}^{\infty} ([n]+k)(|[n]+k-1)p([n]+k)/n_a^2. \tag{A4}$$

Since $n_a$ is the same for both states (A1) and (A2) when Eq. (A3) is satisfied, it is enough for the comparison of their antibunching to consider the difference of their $n_a^2g^{(2)}$ (effectively getting rid of the denominator, which simplifies the notations). We call this difference $\Delta G^{(2)}$ and find, from Eq. (11) and (A4):

$$\sum_{k=-\lfloor n\rfloor}^{\infty} (([n]+k)(|[n]+k-1)p([n]+k)/n_a^2 - \sum_{k=-\lfloor n\rfloor}^{\infty} (([n]+k)(|[n]+k-1)p([n]+k)/n_a^2)\Delta G^{(2)} = 0.$$

We prove mathematically that there are no states below the boundary set by Eq. (11) but at least one for every point above, thus showing that our charting of the Hilbert space in the $(n_a,g^{(2)})$ space as defined by this boundary is complete.
\[ \Delta G^{(2)} = \sum_{k=-\lfloor n_a \rfloor}^{\infty} (\lfloor n_a \rfloor + k)(\lfloor n_a \rfloor + k - 1)p(\lfloor n_a \rfloor + k) - |n_a|(\lfloor n_a \rfloor - 1)(\lfloor n_a \rfloor - n_a + 1) - |n_a|(\lfloor n_a \rfloor + 1)(n_a - |n_a|) + , \]

where we have separated the terms \( k = 0, 1 \) from the sum and re-arranged the factors. Now, since \( p(\lfloor n_a \rfloor + 1) = 1 - p(\lfloor n_a \rfloor) - \sum_{k \neq \{0, 1\}} p(\lfloor n_a \rfloor + k) \) from the probability normalization, we arrive to:

\[
\Delta G^{(2)} = \left[ \sum_{k \neq \{0, 1\}} (k-1)(2\lfloor n_a \rfloor + k)p(\lfloor n_a \rfloor + k) \right] - 2\lfloor n_a \rfloor(p(\lfloor n_a \rfloor) - |n_a| + n_a - 1). \tag{A5}
\]

On the other hand, from Eq. (A5), we can isolate:

\[ p(\lfloor n_a \rfloor) = |n_a| - n_a + 1 + \sum_{k \neq \{0, 1\}} (k-1)p(\lfloor n_a \rfloor + k), \tag{A6} \]

which, injected back in Eq. (A5), yields:

\[ \Delta G^{(2)} = \sum_{k \neq \{0, 1\}} k(k-1)p(\lfloor n_a \rfloor + k). \]

This is the final result, since all the terms in the sum are non-negative, we can conclude that \( \Delta G^{(2)} \geq 0 \).

The inequality is maximized when all the probabilities \( p(\lfloor n_a \rfloor + k) = 0 \) for \( k \neq \{0, 1\} \), i.e., for the states of type \( \{1\} \). When \( n_a \) is an integer, i.e., \( \lfloor n_a \rfloor = n_a \), the maximum antibunching is obtained with the Fock state \( |n_a \rangle \).

QED.

**Proposition:** For a given population \( n_a \in \mathbb{R} \) and any positive real number \( \delta \), there is at least one state such that \( \Delta G^{(2)} = \delta \).

Let us consider the set of states with population \( n_a = \sum_n np(n) \). We can construct a state \( \rho_0 \) with population \( n_a \) in the following way:

\[ \rho_0 = p(0)|0 \rangle \langle 0| + p(|n_a \rangle)[|n_a \rangle \langle n_a|] + (1 - p(0) - p(|n_a \rangle))|k \rangle \langle k|. \]

Imposing the population constraint, we obtain the following condition:

\[ p(0) = \left( \frac{k-n_a}{k} \right) - p(|n_a \rangle) \left( \frac{k-n_a}{k} \right), \]

where we have assumed that \( |n_a| < k \in \mathbb{N} \). The \( g^{(2)} = g^{(2)}_s + \delta \) of this state is given by:

\[ g^{(2)}_a = |n_a|(|n_a| - 1)p(|n_a \rangle) + k(k-1)(1 - p(0) - p(|n_a \rangle)), \]

from which we can obtain \( p(|n_a \rangle) = n_a \left( k - 1 - n_a g^{(2)} \right) \left( n_a(k - |n_a|) \right). \]

If \( \delta > 0 \) we can find an integer number \( k \geq \max(n_a g^{(2)}, |n_a|) \) such that \( 0 \leq p(|n_a \rangle) \leq 1 \). Considering that this state is not made of two consecutive Fock states, the accessible \( g^{(2)}_s \) are larger than \( g^{(2)}_s \). Of course, when \( p(0) = 0 \) and \( k = |n_a| + 1 \), \( \rho_0 \) is the maximally antibunched state and \( g^{(2)} = g^{(2)}_s \). When \( n_a < 1 \), \( |n_a| = 0 \), and the procedure above fails. In such case, we can build another state,

\[ \rho_1 = p(0)|0 \rangle \langle 0| + p(1)|1 \rangle \langle 1| + (1 - p(0) - p(1))|k \rangle \langle k|, \]

such that \( n_a = p(1) + k(1 - p(0) - p(1)) \). From this constraint we obtain the following condition:

\[ p(1) = \frac{k(1 - p(0)) - n_a}{k - 1}. \]

The \( g^{(2)}_a \) of this state is given by:

\[ g^{(2)}_a = k(k-1)(1 - p(0) - p(1)), \]

\[ = k(n_a + p(0) - 1), \]

from which we conclude that:

\[ p(0) = \frac{g^{(2)}_a n_a^2 + k(1 - n_a)}{k}. \]

Again, we can always find an integer \( k > 1 \), such that \( 0 \leq p(0) \leq 1 \).
1 Saleh, B. E. A. & Teich, M. C. *Fundamentals of Photonics* (Wiley-Interscience, 2007), 2nd edn.
2 Clauser, J. F. Experimental distinction between the quantum and classical field-theoretic predictions for the photoelectric effect. *Phys. Rev. D* 9, 853 (1974).
3 Kimble, H. J., Dagenais, M. & Mandel, L. Photon antibunching in resonance fluorescence. *Phys. Rev. Lett.* 39, 691 (1977).
4 Michler, P. et al. A quantum dot single-photon turnstile device. *Science* 290, 2262 (2000).
5 Shields, A. J. Semiconductor quantum light sources. *Nat. Photon.* 1, 215 (2007).
6 Eisaman, M. D., Fan, J., Migdall, A. & Polyakov, S. V. Single-photon sources and detectors. *Rev. Sci. Instrum.* 82, 071101 (2011).
7 Kuhlmann, A. V. et al. Transform-limited single photons from a single quantum dot. *Nat. Commun.* 6, 8204 (2015).
8 Portalupi, S. L. et al. Bright phonon-tuned single-photon source. *Nano Lett.* 15, 6290 (2015).
9 Müller, K. et al. Coherent generation of nonclassical light on chip via detuned photon blockade. *Phys. Rev. Lett.* 114, 233601 (2015).
10 Ding, X. et al. On-demand single photons with high extraction efficiency and near-unity indistinguishability from a resonantly driven quantum dot in a micropillar. *Phys. Rev. Lett.* 116, 020401 (2016).
11 Somaschi, N. et al. Near-optimal single-photon sources in the solid state. *Nat. Photon.* 10, 340 (2016).
12 Chunnilall, C. J., Degiovanni, I. P., Kück, S., Müller, I. & Sinclair, A. G. Metrology of single-photon sources and detectors: a review. *Opt. Eng.* 53, 081910 (2014).
13 Taylor, M. A. & Bowen, W. P. Quantum metrology and its application in biology. *Phys. Rep.* 615, 1 (2016).
14 Beveratos, A. et al. Single photon quantum cryptography. *Phys. Rev. Lett.* 89, 187901 (2002).
15 Kiraz, A., Atatüre, M. & İnanməxoju, A. Quantum-dot single-photon sources: Prospects for applications in linear optics quantum-information processing. *Phys. Rev. A* 69, 032305 (2004).
16 Tamma, V. & Laibacher, S. Multiboson correlation interference with arbitrary single-photon pure states. *Phys. Rev. Lett.* 114, 243601 (2015).
17 Lambrecht, N. et al. Quantum biology. *Nat. Phys.* 9, 10 (2013).
18 Tinsley, J. N. et al. Direct detection of a single photon by humans. *Nat. Comm.* 7, 12172 (2016).
19 Hadfield, R. H. Single-photon detectors for optical quantum information applications. *Nat. Photon.* 3, 696 (2009).
20 Reithmaier, G. et al. On-chip time resolved detection of quantum dot emission using integrated superconducting single photon detectors. *Scientific Report* 3, 1901 (2013).
21 Kira, M. & Koch, S. W. Quantum-optical spectroscopy of semiconductors. *Phys. Rev. A* 73, 013813 (2006).
22 Affmann, M. & Bayer, M. Nonlinearity sensing via photon-statistics excitation spectroscopy. *Phys. Rev. A* 84, 053806 (2011).
23 Affmann, M. & Bayer, M. Stochastic pumping of a polariton fluid. *Phys. Rev. A* 91, 053835 (2015).
24 Mukamel, S. & Dorfman, K. E. Nonlinear fluctuations and dissipation in matter revealed by quantum light. *Phys. Rev. A* 91, 053844 (2015).
25 López Carreño, J. C., Sánchez Muñoz, C., Sanvitto, D., del Valle, E. & Laussy, F. P. Exciting polaritons with quantum light. *Phys. Rev. Lett.* 115, 196402 (2015).
26 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).
27 del Valle, E. Distilling one, two and entangled pairs of photons from a quantum dot with cavity QED effects and spectral filtering. *New J. Phys.* 15, 025019 (2013).
28 Gonzalez-Tudela, A., Laussy, F. P., Tejedor, C., Hartmann, M. J. & del Valle, E. Two-photon spectra of quantum emitters. *New J. Phys.* 15, 033036 (2013).
29 Sánchez Muñoz, C., del Valle, E., Tejedor, C. & Laussy, F. Violation of classical inequalities by photon frequency filtering. *Phys. Rev. A* 90, 052111 (2014).
30 González-Tudela, A., del Valle, E. & Laussy, F. P. Optimization of photon correlations by frequency filtering. *Phys. Rev. A* 91, 043807 (2015).
31 Kameide, K., Iwamoto, S. & Arakawa, Y. Eigenvalue decomposition method for photon statistics of frequency-filtered fields and its application to quantum dot emitters. *Phys. Rev. A* 92, 033833 (2015).
32 Peiris, M. et al. Two-color photon correlations of the light scattered by a quantum dot. *Phys. Rev. B* 91, 195125 (2015).
33 Gardiner, C. W. Driving a quantum system with the output field from another driven quantum system. *Phys. Rev. Lett.* 70, 2269 (1993).
34 Carmichael, H. J. Quantum trajectory theory for cascaded open systems. *Phys. Rev. Lett.* 70, 2273 (1993).
35 Walls, D. F. Squeezed states of light. *Nature* 306, 141 (1983).
36 Toyli, D. M. et al. Resonance fluorescence from an artificial atom in squeezed vacuum. *Phys. Rev. X* 6, 031004 (2016).
37 Kolobov, M. & Sokolov, I. Interference of a physical radiation field and of vacuum fluctuations at a light-source output. *Opt. Spectrosk.* 62, 112 (1987).
38 Gardiner, G. W. & Zoller, P. *Quantum Noise* (Springer-Verlag, Berlin, 2000), 2nd edn.
39 Gardiner, C. W. & Parkins, A. S. Driving atoms with light of arbitrary statistics. *Phys. Rev. A* 50, 1792 (1994).
40 Cirac, J. I., Zoller, P., Kimble, H. J. & Mabuchi, H. Quantum state transfer and entanglement distribution among distant nodes in a quantum network. *Phys. Rev. Lett.* 78, 3221 (1997).
41 Smyth, W. S. & Swain, S. Complete quenching of fluorescence from a two-level atom driven by a weak, narrow-band, nonclassical light field. *Phys. Rev. A* 50, R2579(R) (1999).
42 Messikh, A., Tanaš, R. & Ficek, Z. Response of a two-level atom to a narrow-bandwidth squeezed-vacuum excitation. *Phys. Rev. A* 61, 033811 (2000).
43 Grundmann, M., Stier, O. & Bimberg, D. InAs/GaAs pyramidal quantum dots: Strain distribution, optical phonons, and electronic structure. *Phys. Rev. B* 52, 11969 (1995).
44 Petersen, J., Volz, J. & Rauschenbeutel, A. Chiral...
nanophotonic waveguide interface based on spin-orbit interaction of light. *Science* **346**, 67 (2014).

46 Gonzalez-Ballestero, C., Gonzalez-Tudela, A., García-Vidal, F. J. & Moreno, E. Chiral route to spontaneous entanglement generation. *Phys. Rev. B* **92**, 155304 (2015).

47 Pichler, H., Ramos, T., Daley, A. J. & Zoller, P. Quantum optics of chiral spin networks. *Phys. Rev. A* **91**, 042116 (2015).

48 Coles, R. J. et al. Chirality of nanophotonic waveguide with embedded quantum emitter for unidirectional spin transfer. *Nat. Comm.* **7**, 11183 (2016).

49 Mahmoodian, S., Lodahl, P. & Sorensen, A. S. Quantum networks with chiral light-matter interaction in waveguides. *arXiv:1602.07054* (2016).

50 Guimond, P., Pichler, H., Rauschenbeutel, A. & Zoller, P. Chiral quantum optics with v–level atoms and coherent quantum feedback. *arXiv:1608.00446* (2016).

51 Lodahl, P. et al. Chiral quantum optics. *arXiv:1608.00446* (2016).

52 Gonzalez-Ballestero, C., Moreno, E., García-Vidal, F. J. & Gonzalez-Tudela, A. Non-reciprocal few-photon devices based on chiral waveguide-emitter couplings. *arXiv:1608.04928* (2016).

53 Laussy, F. P., Shelykh, I. A., Malpuech, G. & Kavokin, A. Effects of Bose–Einstein condensation of exciton polaritons in microparticles on the polarization of emitted light. *Phys. Rev. B* **73**, 035315 (2006).

54 Lvovsky, A. I. *Fundamentals of Photonics and Physics*, chap. 5. Squeezed light (Wiley-Blackwell, 2015).

55 Haroche, S. & Raimond, J.-M. *Exploring the Quantum: Atoms, Cavities, and Photons* (Oxford University Press, 2006).

56 Stoler, D., Saleh, B. & Teich, M. Binomial states of the quantized radiation field. *Opt. Acta* **32**, 345 (1985).

57 Nieto, M. M. Displaced and squeezed number states. *Phys. Lett. A* **229**, 135 (1997).

58 Dong, S., an Y. Liu, T. H., Wang, J., G. Zhang, L. X. & Jia, S. Fast recognition of single molecules based on single-event photon statistics. *Phys. Rev. A* **76**, 063820 (2007).

59 Verma, V. B. et al. Photon antibunching from a single lithographically defined InGaAs/GaAs quantum dot. *Opt. Express* **19**, 4182 (2011).

60 Martino, G. D. et al. Quantum statistics of surface plasmon polaritons in metallic stripe waveguides. *Nano Lett.* **12**, 2504 (2012).

61 Leifgen, M. et al. Evaluation of nitrogen- and silicon-vacancy defect centres as single photon sources in quantum key distribution. *New J. Phys.* **16**, 023021 (2014).

62 Zabizarreta Casalengua, E., López Carreño, J. C., del Valle, E. & Laussy, F. P. Structure of the harmonic oscillator Hilbert space. *arXiv:1607.03976* (2016).

63 Flayac, H. & Savona, V. Heralded preparation and readout of entangled phonons in a photonic crystal cavity. *Phys. Rev. Lett.* **113**, 143603 (2014).

64 del Valle, E. & Laussy, F. P. Regimes of strong light-matter coupling under incoherent excitation. *Phys. Rev. A* **84**, 043816 (2011).

65 Sánchez Muñoz, C. et al. Emitters of N-photon bundles. *Nat. Photon.* **8**, 550 (2014).