PHYSICAL REVIEW A **83**, 021802(R) (2011)

Origin of strong photon antibunching in weakly nonlinear photonic molecules

Motoaki Bamba,[1,*] Atac Imamoğlu,[2] Iacopo Carusotto,[3] and Cristiano Ciuti[1,†]

[1]*Laboratoire Matériaux et Phénomènes Quantiques, Université Paris Diderot-Paris 7 et CNRS, Bâtiment Condorcet, 10 rue Alice Domon et Léonie Duquet, F-75205 Paris Cedex 13, France*

[2]*Institute of Quantum Electronics, ETH Zürich, CH-8093 Zürich, Switzerland*

[3]*INO-CNR BEC Center and Dipartimento di Fisica, Università di Trento, I-38123 Povo, Italy*

(Received 9 July 2010; published 22 February 2011)

In a recent work [Liew and Savona, Phys. Rev. Lett. **104**, 183601 (2010)] it was numerically shown that a resonantly driven photonic "molecule" consisting of two coupled cavities can exhibit strong photon antibunching with a surprisingly weak Kerr nonlinearity. Here, we analytically identify the subtle quantum interference effect that is responsible for the predicted efficient photon blockade effect. We then extend the theory to the experimentally relevant Jaynes-Cummings system consisting of a single quantum emitter in a coupled-cavity structure and predict the strong antibunching even for single-atom cooperativity on the order of or smaller than unity. The potential of this quantum interference effect in the realization of strongly correlated photonic systems with only weak material nonlinearities is assessed by comparing on-site and inter-site correlations in a ring of three coupled photonic molecules.

DOI: 10.1103/PhysRevA.83.021802 PACS number(s): 42.50.Ct, 42.50.Ar, 42.50.Dv

The photon blockade is a quantum optical effect preventing the resonant injection of more than one photon into a nonlinear cavity mode [1], leading to antibunching in photon correlation measurements. Photon blockade have been observed by resonant laser excitation on a single atom [2], while small signatures have been observed so far in the case of a resonantly driven single quantum dot embedded in a strongly coupled cavity [3]. Arguably, the most convincing realization was based on a single atom coupled to a microtoroidal cavity [4]. Concurrently, on the theory side there have been a number of proposals investigating strongly correlated photons in coupled cavity arrays [5–7] or one-dimensional optical waveguides [8]. The specific proposals based on the photon blockade effect include the fermionization of photons in one-dimensional cavity arrays [9], the crystallization of polaritons in coupled array of cavities [10], and the quantum-optical Josephson interferometer in a coupled photonic mode system [11].

It is commonly believed that photon blockade necessarily requires a strong on-site Kerr nonlinearity U for a photonic mode, whose magnitude should well exceed the mode broadening γ. However, in a recent work [12] Liew and Savona used a numerical and semianalytical linear-fluctuation analysis to show that strong antibunching can be obtained with a surprisingly weak Kerr nonlinearity ($U \ll \gamma$) in a system consisting of two coupled zero-dimensional (0D) photonic cavities (boxes), as shown in Fig. 1(a) [12]. Such a configuration can be obtained, e.g., by considering two modes in two photonic boxes coupled with a finite mode overlap due to leaky mirrors; the corresponding tunnel strength will be designated with J. In Ref. [12] numerical evidence indicated that nearly perfect antibunching can be achieved for an optimal value of the on-site repulsion energy U and the detuning between the pump and mode frequency. However, a physical understanding of the mechanism leading to strong photon antibunching is needed to identify the limitations of the scheme in the context of proposed experiments on strongly correlated photons, as well as to determine the dependence of the optimal coupling and detuning on the relevant physical parameters J and γ.

In this Rapid Communication, we show analytically that the surprising antibunching effect is the result of a subtle destructive quantum interference effect which ensures that the probability amplitude of having two photons in the driven cavity is zero. A similar mechanism was invoked to explain the strong nonclassical effects in cavity systems with many atoms [13–15]. We show that a weak Kerr nonlinearity is required only for the auxiliary cavity that is not laser driven and whose output is not monitored, indicating that photon antibunching is obtained for a linear cavity that tunnel couples to a weakly nonlinear one. Armed with this observation, we extend the prediction of Liew and Savona [12] to the experimentally relevant system consisting of a single anharmonic quantum emitter (i.e., an atom or a quantum dot) embedded in a photonic molecule: In contrast to what is usually expected [2–4], we find that an analogous quantum interference effect allows for the observation of strong quantum correlations between single photons even when the single-atom cooperativity parameter is on the order of or even smaller than unity [13–15]. Finally, we demonstrate the potential of this quantum interference effect to realize strongly correlated photonic systems; in spite of the weak material nonlinearities, strong on-site antibunching can be observed in a ring of coupled photonic molecules simultaneously with a strong inter-site bunching.

We consider two coupled photonic modes with energy E_i and on-site photon-photon interaction strength U_i ($i = 1,2$). The Hamiltonian is written as

$$\hat{H} = \sum_{i=1}^{2}[E_i\hat{a}_i^\dagger\hat{a}_i + U_i\hat{a}_i^\dagger\hat{a}_i^\dagger\hat{a}_i\hat{a}_i] + J(\hat{a}_1^\dagger\hat{a}_2 + \hat{a}_2^\dagger\hat{a}_1) + Fe^{-\mathrm{i}\omega_\mathrm{p}t}\hat{a}_1^\dagger + F^* e^{\mathrm{i}\omega_\mathrm{p}t}\hat{a}_1, \tag{1}$$

*motoaki.bamba@univ-paris-diderot.fr

†cristiano.ciuti@univ-paris-diderot.fr

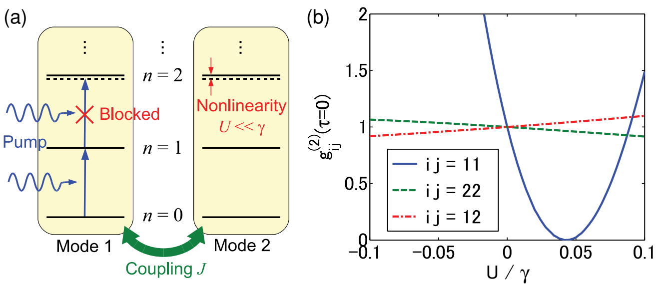

FIG. 1. (Color online) (a) Sketch of the two coupled photonic modes. The coupling strength is J, and the antibunching is obtained with a small nonlinear energy U compared to mode broadening γ. (b) Equal-time second-order correlation functions $g^{(2)}_{ij}(\tau = 0)$ plotted as functions of nonlinearity $U = U_1 = U_2$ normalized to γ. The nearly perfect antibunching is obtained at the pumped mode $[g^{(2)}_{11}(\tau = 0) \simeq 0]$ for $U = 0.0428\gamma$. The parameters are $\gamma_1 = \gamma_2 = \gamma$, $J = 3\gamma$, $E_1 = E_2 = \hbar\omega_p + 0.275\gamma$, and $F_1 = 0.01\gamma$.

where $\hat{a}_i$ is the annihilation operator of a photon in the ith mode, and F and ω_p are the pumping strength and frequency, respectively. Following Ref. [12], we first calculate the second-order correlation function $g^{(2)}_{ij}(\tau) = \langle \hat{a}_i^\dagger \hat{a}_j^\dagger(\tau)\hat{a}_j(\tau)\hat{a}_i\rangle / \langle \hat{a}_i^\dagger \hat{a}_i\rangle\langle \hat{a}_j^\dagger \hat{a}_j\rangle$ in the steady state using the master equation in a basis of Fock states [16]. The results are shown as functions of nonlinearity $U = U_1 = U_2$ in Fig. 1(b). As already demonstrated in Ref. [12], we can get strong antibunching of the pumped mode $[g^{(2)}_{11}(0) \simeq 0]$ for an unexpectedly small Kerr nonlinearity $U = 0.0428\gamma$.

In order to understand the origin of the strong antibunching, we use the ansatz

$$|\psi\rangle = C_{00}|00\rangle + e^{-i\omega_p t}(C_{10}|10\rangle + C_{01}|01\rangle) + e^{-i2\omega_p t}(C_{20}|20\rangle + C_{11}|11\rangle + C_{02}|02\rangle) + \cdots \quad (2)$$

to calculate the steady state of the coupled-cavity system. Here, $|mn\rangle$ represents the Fock state with m particles in mode 1 and n particles in mode 2. Under weak pumping conditions ($C_{00} \gg C_{10}, C_{01} \gg C_{20}, C_{11}, C_{02}$), we can calculate the coefficients C_{mn} iteratively [13]. For one-particle states, the steady-state coefficients are determined by

$$(\Delta E_1 - i\gamma_1/2)C_{10} + JC_{01} + FC_{00} = 0, \quad (3a)$$

$$(\Delta E_2 - i\gamma_2/2)C_{01} + JC_{10} = 0, \quad (3b)$$

where $\Delta E_i = E_i - \hbar\omega_p$ and we consider a broadening γ_j for each mode. Since we assume weak pumping, the contribution from the higher states (C_{20}, C_{11}, and C_{02}) to the steady-state values of C_{10}, C_{01} is negligible. From Eq. (3b), the amplitude of mode 2 can be written as

$$C_{01} = -\frac{J}{\Delta E_2 - i\gamma_2/2}C_{10}, \quad (4)$$

indicating that for strong photon tunneling ($J \gg |\Delta E_2|, \gamma_2$), the probability of finding a photon in the auxiliary cavity is much larger than in the driven cavity.

In the same manner, the coefficients of two-particle states are determined by

$$2(\Delta E_1 + U_1 - i\gamma_1/2)C_{20} + \sqrt{2}JC_{11} + \sqrt{2}FC_{10} = 0, \quad (5a)$$

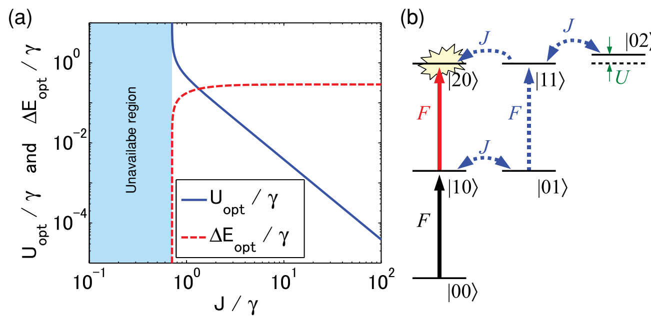

FIG. 2. (Color online) (a) Optimal nonlinearity U_{opt} and detuning ΔE_{opt} plotted as functions of inter-mode coupling strength J normalized to broadening γ ($\gamma_1 = \gamma_2 = \gamma$ and $E_1 = E_2 = E$). Nearly perfect antibunching is obtained for $J > \gamma/\sqrt{2}$. (b) Transition paths leading to the quantum interference responsible for the strong antibunching. One path is the direct excitation from $|10\rangle$ to $|20\rangle$, but it is forbidden by the interference with the other path drawn by dotted arrows.

$$(\Delta E_1 + \Delta E_2 - i\gamma_1/2 - i\gamma_2/2)C_{11} + \sqrt{2}JC_{20} + \sqrt{2}JC_{02} + FC_{01} = 0, \quad (5b)$$

$$2(\Delta E_2 + U_2 - i\gamma_2/2)C_{02} + \sqrt{2}JC_{11} = 0. \quad (5c)$$

When we simply consider $E_1 = E_2 = E$ and $\gamma_1 = \gamma_2 = \gamma$, the conditions to satisfy $C_{20} = 0$ are derived from Eqs. (4) and (5) as

$$\gamma^2(3\Delta E + U_2) - 4\Delta E^2(\Delta E + U_2) = 2J^2U_2, \quad (6a)$$

$$12\Delta E^2 + 8\Delta EU_2 - \gamma^2 = 0. \quad (6b)$$

For fixed J and γ, from these equations, the optimal conditions (those that lead to $C_{20} = 0$) are given by

$$\Delta E_{opt} = \pm\frac{1}{2}\sqrt{\sqrt{9J^4 + 8\gamma^2J^2} - \gamma^2 - 3J^2}, \quad (7a)$$

$$U_{opt} = \frac{\Delta E_{opt}(5\gamma^2 + 4\Delta E_{opt}{}^2)}{2(2J^2 - \gamma^2)}, \quad (7b)$$

and, if $J \gg \gamma$, they are approximately written as

$$\Delta E_{opt} \simeq \pm\frac{\gamma}{2\sqrt{3}}, \quad (8a)$$

$$U_{opt} \simeq \pm\frac{2}{3\sqrt{3}}\frac{\gamma^3}{J^2}. \quad (8b)$$

In Fig. 2(a), the optimal ΔE_{opt} and U_{opt} [Eq. (7)] are plotted as functions of J/γ. The strong photon antibunching can be obtained even if $U_2 < \gamma$, provided $J > \gamma/\sqrt{2}$. Remarkably, the required nonlinearity decreases with increasing tunnel coupling J obeying Eq. (8b).

In Fig. 2(b), we show a sketch of the quantum interference effect responsible for this counterintuitive photon antibunching. The interference is between the following two paths: (a) the direct excitation from $|10\rangle \xrightarrow{F} |20\rangle$ (solid arrow) and (b) tunnel-coupling-mediated transition $|10\rangle \overset{J}{\leftrightarrow} |01\rangle \xrightarrow{F} (|11\rangle \overset{J}{\leftrightarrow} |02\rangle) \xrightarrow{J} |20\rangle$ (dotted arrows). In order to show in detail the origin of the quantum interference, we

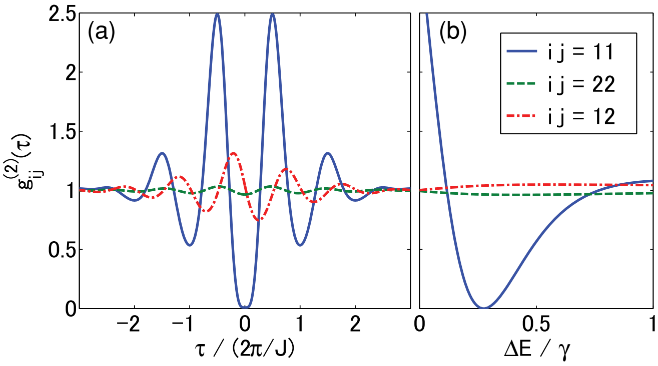

FIG. 3. (Color online) (a) The time evolution of the second-order correlation function, which oscillates with period $2\pi/J$ as the result of amplitude oscillation between $|01\rangle$ and $|10\rangle$. (b) Equal-time second-order correlation functions plotted as functions of $\Delta E_1 = \Delta E_2 = \Delta E$ normalized to $\gamma_1 = \gamma_2 = \gamma$. The spectral width of the antibunching resonance is $\approx 0.3\gamma$. The parameters are $J = 3\gamma$, $U_1 = U_2 = 0.0428\gamma$, and $F = 0.01\gamma$. $\Delta E = 0.275\gamma$ in panel (a).

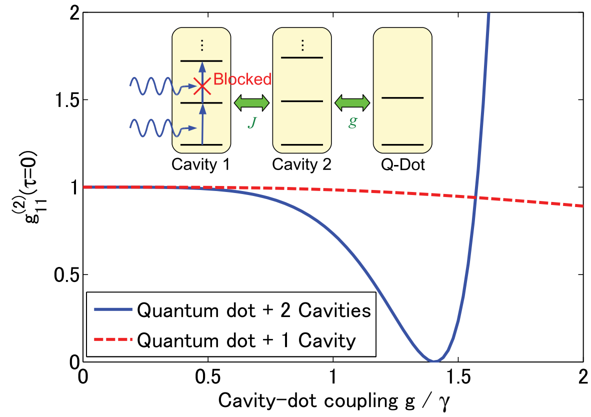

FIG. 4. (Color online) Equal-time correlation functions plotted as functions of coupling strength g between a cavity and a quantum dot. The solid line represents the results in the system sketched in the inset [Eq. (10)]. The parameters are $\gamma_1 = \gamma_2 = \gamma_{\text{ex}} = \gamma$, $J = 3\gamma$, $E_1 = \hbar\omega_{\text{p}} + 0.275\gamma$, $E_2 - E_1 = \gamma$, $E_{\text{ex}} - E_2 = 2\gamma$, and $F = 0.01\gamma$. The dashed line represents the result in the system with one quantum dot and one cavity [Jaynes-Cummings model]. $\gamma_1 = \gamma_{\text{ex}} = \gamma$, $E_{\text{ex}} - E_1 = 2\gamma$, $F = 0.01\gamma$, and $\hbar\omega_{\text{p}}$ is tuned to the lower one-particle eigenenergy of the Jaynes-Cummings ladder.

rederive Eqs. (6) for $C_{20} = 0$ as follows. First, we calculate C_{11} from Eqs. (4) and (5) neglecting C_{20} as

$$C_{11} = -2JFC_{10}(\Delta E + U_2 - i\gamma/2)(\Delta E - i\gamma/2)^{-1} \times [2J^2 - 4\Delta E(\Delta E + U_2) + \gamma^2 + i2\gamma(2\Delta E + U_2)]^{-1}. \tag{9}$$

This amplitude is the result of excitation from $|01\rangle$ to $|11\rangle$ and of the coupling between $|10\rangle$ and $|01\rangle$ and also between $|11\rangle$ and $|02\rangle$. From this amplitude, C_{20} is determined by Eq. (5a) as $C_{20} \propto JC_{11} + FC_{10}$, and we can derive Eqs. (6) for the condition $C_{20} = 0$. It is worth noting that the nonlinearity U_1 of the pumped cavity mode is irrelevant for the antibunching. This implies that a (weak) nonlinearity is required only in the auxiliary (undriven) photonic mode to achieve the perfect destructive quantum interference that leads to $C_{20} = 0$. Of course, a finite value of U_2 is required to see any antibunching; even if a destructive interference effect takes place in a linear system, $g^{(2)}_{11}(\tau = 0)$ remains strictly unity in the absence of a nonlinearity.

As seen in Fig. 1(b), while no more than one photon is present in the first cavity mode for the optimal condition, there can be more than one photon in the whole system. While there is nearly perfect antibunching in the driven mode [$g^{(2)}_{11}(\tau = 0) \ll 1$], the cross-correlation between the two modes exhibits bunching [$g^{(2)}_{12}(\tau = 0) > 1$]. The amplitude oscillation between $|10\rangle$ and $|01\rangle$ produces the time oscillation of $g^{(2)}_{11}(\tau)$ with period $2\pi/J$, as reported in Ref. [12] and shown in Fig. 3(a). A similar oscillation is also observed in the atomic case as a result of amplitude oscillation between the cavity and atoms [13–15].

The equal-time correlation function is plotted in Fig. 3(b) as a function of the pump detuning $\Delta E/\gamma$: While the optimal value of the detuning is at $\Delta E = 0.275\gamma$, strong antibunching is obtained in a range of about 0.3γ around the optimal value and the width of this window does not significantly depend on J/γ. This may suggest that pump pulses of duration Δt_p longer than $1/(0.3\gamma)$ could be enough to ensure strong antibunching. However, the time scale over which strong quantum correlations between the photons exist is on the order of $1/J < \sqrt{2}/\gamma$, as seen in Fig. 3(a). While weak nonlinearities do lead to strong quantum correlations, these correlations last for a time scale that scales with $1/J \propto \sqrt{U_{\text{opt}}}$ [see Eq. (8b)]. From a practical perspective, a principal difficulty with the observation of the photon antibunching with weak nonlinearities is that it requires fast single-photon detectors. Conversely, for a given detection setup, the required minimal value of the nonlinearity is ultimately determined by the time resolution of the available single-photon detector.

A large majority of experimental efforts aimed at demonstrating the photon blockade effect are based on the resonantly driven Jaynes-Cummings model. Having noticed that a weak nonlinearity is needed in only one of the cavities, we investigate if an analogous quantum interference effect can be exploited to lower the minimum value of the cavity-emitter coupling strength g that is required to achieve photon antibunching in an extended Jaynes-Cummings model consisting of a single quantum emitter embedded in a photonic molecule (see the inset in Fig. 4). The Hamiltonian is written as

$$\hat{H}_{\text{cav-JC}} = \sum_{i=1}^{2} E_i \hat{a}_i^\dagger \hat{a}_i + J(\hat{a}_1^\dagger \hat{a}_2 + \hat{a}_2^\dagger \hat{a}_1) + E_{\text{ex}}|\text{ex}\rangle\langle\text{ex}| + g(\hat{a}_2^\dagger |\text{g}\rangle\langle\text{ex}| + \text{H.c.}) + Fe^{-i\omega_{\text{p}}t}\hat{a}_1^\dagger + F^* e^{i\omega_{\text{p}}t}\hat{a}_1. \tag{10}$$

Here, $|\text{g}\rangle$ and $|\text{ex}\rangle$ represent the ground and excited states of the quantum dot, respectively, and E_{ex} is the excited state energy. We take the quantum dot exciton broadening γ_{exc} to be equal to the cavity decay rate γ for simplicity.

In most prior implementations with $\gamma \simeq \gamma_{\text{exc}}$, the requirement for observation of strong quantum correlations in the Jaynes-Cummings model [2] was assumed to be a large

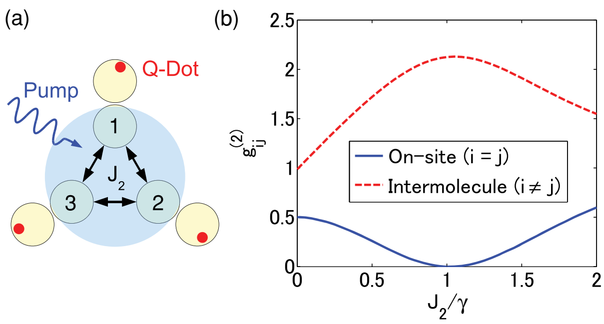

FIG. 5. (Color online) (a) Sketch of a triangular lattice of coupled photonic "molecules." The driven cavities ($i = 1$, 2, and 3) are coupled with strength J_2. (b) The equal-time second-order correlation functions in each mode (solid line) and between neighbors (dashed line) plotted vs J_2/γ. The parameters are $J = 3\gamma$, $\Delta E = 0.450\gamma$, $U = 0.0769\gamma$, and $F = 0.01\gamma$.

single-atom cooperativity parameter $C = g^2/(\gamma\gamma_{\mathrm{exc}}) \gg 1$ [17]. To see if this condition can be relaxed using quantum interference, we have solved numerically the master equation associated with the Hamiltonian in Eq. (10). Figure 4 shows $g^{(2)}_{11}(\tau = 0)$ of the pumped mode as a function of g/γ. The coupling energy between the two cavities is chosen to be $J = 3\gamma$, which gives $U_{\mathrm{opt}} = 0.0428\gamma$ from Fig. 2. In the present system, this U_{opt} is practically achieved at $g = 1.4\gamma$, which is an intermediate strength between the weak- and strong-coupling regimes of cavity mode and quantum emitter excitation. The dashed line in Fig. 4 represents the results in the system consisting of one quantum dot and one cavity: In this ordinary Jaynes-Cummings system, only a vanishingly small antibunching is obtained at $g \simeq \gamma$, and $C \gg 1$ is required for the observation of large photon antibunching [1,2]. In contrast, in the new scheme using quantum interference, nearly perfect antibunching can be obtained even for $g \simeq \gamma$ and significant quantum correlations between photons survive even for $g < \gamma$, i.e., $C < 1$.

The realization of strongly correlated driven and dissipative photonic systems in arrays of cavities in the presence of strong nonlinearities has emerged as a principal research direction in quantum optics [5–11]. Given the completely different physical mechanism underlying antibunching in weakly nonlinear photonic molecules, it is not *a priori* clear whether the predictions of this work may scale up to strong quantum correlations in an array of coupled photonic molecules. To address this most important question, we consider a ring of three molecules whose driven dots are coupled with each other by a tunnel coupling of amplitude J_2 [see Fig. 5(a)]. Also in this case nearly perfect antibunching can be observed in each driven mode, as shown in the plots of $g^{(2)}_{ii}(\tau = 0)$ as a function of J_2/γ that are shown as a solid line in Fig. 5(b). In order to optimize the antibunching at a finite value of $J_2 \simeq \gamma$, values of $U = 0.0769\gamma$ and $\Delta E = 0.450\gamma$ slightly different from the single-molecule optimal ones ($U_{\mathrm{opt}} = 0.0428\gamma$ and $\Delta E_{\mathrm{opt}} = 0.275\gamma$) had to be chosen. At the same time, a strong bunching effect is observed in the equal-time cross-correlation function between neighboring cavities, which shows a value of $g^{(2)}_{i\neq j}(0)$ significantly larger than the coherent field value of $g^{(2)}_{i\neq j}(0) = 1$. This remarkable combination of strong on-site antibunching and strong inter-site bunching suggests that this system may be a viable alternative to the realization of a Tonks-Girardeau gas of fermionized photons discussed in Ref. [9].

In summary, we have analytically determined that a destructive quantum interference mechanism is responsible for strong antibunching in a system consisting of two coupled photonic modes with a weak nonlinearity of either Kerr or Jaynes-Cummings type. This quantum interference effect is robust against small changes in system parameters and has the peculiar feature that the resulting quantum correlation between the generated photons survives for time scales much shorter than the photon lifetime. Nonetheless, we have shown that this quantum interference scheme has the potential to generate strongly correlated photon states in arrays of weakly nonlinear cavities.

C.C. acknowledges support from ANR (projects QPOL and SENOQI) and from the Institut Universitaire de France (IUF).

[1] A. Imamoğlu, H. Schmidt, G. Woods, and M. Deutsch, Phys. Rev. Lett. **79**, 1467 (1997).
[2] K. M. Birnbaum, A. Boca, R. Miller, A. D. Boozer, T. E. Northup, and H. J. Kimble, Nature (London) **436**, 87 (2005).
[3] A. Faraon, I. Fushman, D. Englund, N. Stoltz, P. Petroff, and J. Vuckovic, Nature Phys. **4**, 859 (2008).
[4] B. Dayan, A. S. Parkins, T. Aoki, E. P. Ostby, K. J. Vahala, and H. J. Kimble, Science **319**, 1062 (2008).
[5] M. J. Hartmann, F. G. S. L. Brandao, and M. B. Plenio, Nature Phys. **2**, 849 (2006).
[6] A. D. Greentree, C. Tahan, J. H. Cole, and L. C. L. Hollenberg, Nature Phys. **2**, 856 (2006).
[7] D. G. Angelakis, M. F. Santos, and S. Bose, Phys. Rev. A **76**, 031805 (2007).
[8] D. E. Chang, V. Gritsev, G. Morigi, V. Vuletic, M. D. Lukin, and E. A. Demler, Nature Phys. **4**, 884 (2008).
[9] I. Carusotto, D. Gerace, H. E. Tureci, S. De Liberato, C. Ciuti, and A. Imamoğlu, Phys. Rev. Lett. **103**, 033601 (2009).
[10] M. J. Hartmann, Phys. Rev. Lett. **104**, 113601 (2010).
[11] D. Gerace, H. E. Tureci, A. Imamoğlu, V. Giovannetti, and R. Fazio, Nature Phys. **5**, 281 (2009).
[12] T. C. H. Liew and V. Savona, Phys. Rev. Lett. **104**, 183601 (2010).
[13] H. J. Carmichael, R. J. Brecha, and P. R. Rice, Opt. Commun., **82**, 73 (1991).
[14] G. Rempe, R. J. Thompson, R. J. Brecha, W. D. Lee, and H. J. Kimble, Phys. Rev. Lett. **67**, 1727 (1991).
[15] G. T. Foster, S. L. Mielke, and L. A. Orozco, Phys. Rev. A, **61**, 053821 (2000).
[16] A. Verger, C. Ciuti, and I. Carusotto, Phys. Rev. B **73**, 193306 (2006).
[17] It is however important to recall that strong antibunching has been reported also for smaller C comparable to unity [13-15] and predicted to be perfect [$g^{(2)}_{11}(\tau = 0) = 0$] for $C = 1/2$ in the bad-cavity limit $\gamma \gg \gamma_{\mathrm{exc}}$ [18].
[18] H. J. Carmichael, Phys. Rev. Lett. **55**, 2790 (1985).