Optimal generation of indistinguishable photons from non-identical artificial molecules

E. Cancellieri\textsuperscript{1,2}, F. Troiani\textsuperscript{2} and G. Goldoni\textsuperscript{2,3}

NEST CNR-INFM and Scuola Normale Superiore, Pisa, Italy\textsuperscript{1}
S3 CNR-INFM, Modena, Italy\textsuperscript{2}

Dipartimento di Fisica, Università di Modena e Reggio Emilia, Italy\textsuperscript{3}

emiliano.cancellieri@uam.es

Abstract: We show theoretically that nearly indistinguishable photons can be generated with non-identical semiconductor-based sources. The use of virtual Raman transitions and the optimization of the external driving fields increases the tolerance to spectral inhomogeneity to the meV energy range. A trade-off emerges between photon indistinguishability and efficiency in the photon-generation process. Linear (quadratic) dependence of the coincidence probability within the Hong-Ou-Mandel setup is found with respect to the dephasing (relaxation) rate in the semiconductor sources.

© 2009 Optical Society of America

OCIS codes: (140.3948) Microcavity devices; (230.5590) Quantum-well, -wire and -dot devices; (270.5565) Quantum communications; (270.5585) Quantum information and processing.

References and links
1. Cirac, Zoller, Kimble, and Mabuchi, “Quantum State Transfer and Entanglement Distribution among Distant Nodes in a Quantum Network”, Phys. Rev. Lett. 78, 3221 (1997).
2. Kok, Munro, Nemoto, Ralph, Dowling, and Milburn, “Linear optical quantum computing with photonic qubits”, Rev. Mod. Phys. 79, 135 (2007).
3. Keller, Lange, Hayasaka, Lange, and Walther, “Continuous generation of single photons with controlled waveform in an ion-trap cavity system”, Nature 431, 1075 (2004).
4. McKeever, Boca, Boozer, Miller, Miller, Buck, Kuzmich, and Kimble, “Deterministic Generation of Single Photon from One Atom Trapped in a Cavity”, Science 303, 1992 (2004).
5. Darquié, Jones, Dingjan, Beugnon, Bergamini, Sortais, Messin, Browaeys, and Grangier, “Controlled Single-Photon Emission from a Single Trapped Two-Level Atom”, Science 309, 454 (2005).
6. Beugnon, Jones, Dingjan, Darquié, Messin, Browaeys, and Grangier, “Quantum interference between two single photons emitted by independently trapped atoms”, Nature 440, 779 (2006).
7. Martini, Giuseppe, and Marrocco, “Single-Mode Generation of Quantum Photon States by Excited Single Molecules in a Microcavity Trap”, Phys. Rev. Lett. 76, 900 (1996).
8. Brunel, Lounis, Tamarat, and Orrit, “Triggered Source of Single Photons based on Controlled Single Molecule Fluorescence”, Phys. Rev. Lett. 83, 2722 (1999).
9. Lounis and Moerner, “Single photons on demand from a single molecule at room temperature”, Nature 407, 491 (2000).
10. Kurtsiefer, Mayer, Zarda, and Weinfurter, “Stable Solid-State Source of Single Photons”, Phys. Rev. Lett. 85, 290 (2000).
11. Michler, Mason, Carson, Strouse, Buratto, and Imamoglu, “Quantum correlation among photons from a single quantum dot at room temperature”, Nature 406, 968 (2000).
12. Moreau, Robert, Manin, Thierry-Mieg, Gerard, and Abram, “Quantum Cascade of Photons in Semiconductor Quantum Dots”, Phys. Rev. Lett. 87, 183601 (2001).
13. Zwiller, Blom, Jonsson, Panev, Jeppesen, Tsegaye, Goobar, Pistol, Samuelson, and Bjork, “Single quantum dots emit single photons at a time: Antibunching experiments”, Appl. Phys. Lett. 78, 2476 (2001).
14. Santori, Pelton, Solomon, Dale, and Yamamoto, “Triggered Single Photons from a Quantum Dot”, Phys. Rev. Lett. 86, 1502 (2001).
15. Santori, Fattal, Vuckovic, Solomon, and Yamamoto, “Indistinguishable photons from a single-photon device”, Nature 419, 594 (2002).
16. Zrenner, “A close look on single quantum dots”, J. Chem. Phys. 112, 7790 (2000).
17. Badolato, Hennessy, Atature, Dreyer, Hu, Petroff, and Imamoglu, “Deterministic Coupling of Single Quantum Dots to Single Nanocavity Modes”, Science 308, 1158 (2005).
18. Scully and Zubairy, in Quantum optics (Cambridge University Press, Cambridge, 1997).
19. Kiraz, Atature, and Imamoglu, “Quantum-dot single-photon sources: Prospects for applications in linear optics quantum-information processing”, Phys. Rev. A 69, 32305 (2004).
20. Troiani, Perea, and Tejedor, “Analysis of the photon indistinguishability in incoherently excited quantum dots”, Phys. Rev. B 73, 035316 (2006).
21. Sheng and Leburton, “Spontaneous localization in InAs/GaAs self-assembled quantum-dot molecules”, Appl. Phys. Lett. 81, 4449 (2002).
22. Bester, Shumway, and Zunger, “Theory of Excitonic Spectra and Entanglement Engineering in Dot Molecules”, Phys. Rev. Lett. 93, 47401 (2004).
23. The first excited charged-exciton state 4 has to be included, for its energy separation from 3 is typically of a few meV [29], comparable to the detunings \( \delta_1 \) and \( \delta_2 \).
24. Hong, Ou, and Mandel, “Measurement of subpicosecond time intervals between two photons by interference”, Phys. Rev. Lett. 59, 2044 (1987).
25. Michalewicz Genetic Algorithms + Data Structures = Evolutionary Programming (Springer-Verlag, Berlin, 1992).
26. For simplicity, we keep \( \omega_{41}^A = \omega_{41}^B = 5 \text{meV} \) and \( \omega_{12}^A = \omega_{12}^B = 25 \text{meV} \). Possible differences between the bonding-antibonding splittings of the two AMs can be trivially compensated by adjusting \( \omega_{2}^A \) and \( \omega_{2}^B \).
27. Zanardi and Rossi, “Quantum Information in Semiconductors: Noiseless Encoding in a Quantum-Dot Array”, Phys. Rev. Lett. 81, 4752 (1998).
28. Berton, Rontani, Goldoni, Troiani, and Molinari, “Field-controlled suppression of photon-induced transitions in coupled quantum dots”, Appl. Phys. Lett. 85, 4729 (2004).
29. Krenner, Clark, Nakaoka, Bichler, Scheurer, Abstreiter, and Finley, “Optically Probing Spin and Charge Interactions in a Tunable Artificial Molecule”, Phys. Rev. Lett. 97, 076403 (2006).
30. Fernée, Rubinsztein-Dunlop, and Milburn, “Improving single-photon sources with Stark tuning”, Phys. Rev. A 75, 043815 (2007).
31. Troiani, Wilson-Rae, and Tejedor, “All-optical nondemolition measurement of single hole spin in a quantum-dot molecule”, Appl. Phys. Lett. 90, 144103 (2007).
32. Flindt, Sorensen, Lukin, and Taylor, “Spin-Photon Entangling Diode”, Phys. Rev. Lett. 98, 240501 (2007).
33. Cortez, Krebs-Laurent, Senes, Marie, Vossis, Ferreira, Bastard, Gerard, and Amand, “Optically Driven Spin Memory in n-Doped InAs-GaAs Quantum Dots”, Phys. Rev. Lett. 89, 207401 (2002).

1. Introduction

Single-photon sources (SPSs) are fundamental devices in quantum communication [1] and linear-optics quantum computation [2]. Essentially, a SPS consists of an atomic-like system that can be deterministically excited and thus triggered to emit single-photon wavepackets into a preferential mode. In addition, all photons have to be emitted in the same quantum state, in order to maximize the visibility of two-photon interference.

So far, single-photon generation has been demonstrated both with atomic [3, 4, 5, 6], molecular [7, 8, 9], and solid-state systems [10, 11, 13, 12, 14, 15]. Amongst the latter, self-assembled semiconductor quantum dots (SAQDs) [16] are particularly promising, for they can be control-
lably coupled to optical microcavities [17], so as to greatly enhance both the photon emission rate and the collection efficiency. As a major downside, SAQDs come with a finite dispersion in terms of size, shape, and composition. This typically gives rise to spectral inhomogeneities in the meV range, i.e., orders of magnitude larger than the homogeneous linewidths (few \( \mu eVs \)) of the lowest exciton transitions at cryogenic temperatures. Photons spontaneously emitted by two distinct SAQDs therefore tend to be completely distinguishable; this could impede scalability and ultimately limit the potential of semiconductor-based SPSs.
In spite of its relevance to any solid-state approach, the problem of generating indistinguishable photons with non-identical emitters has still received limited attention. In this Letter, we assess the possibility of compensating the effects of such differences between two SPSs by suitably tuning the exciting laser pulses. To this aim, we combine a density-matrix approach [18] – to simulate the system dynamics and compute the coherence functions of the emitted radiation – with a genetic algorithm [25] – to optimize the external driving fields. As a crucial point, the photon generation results from a (virtual) Raman transition. Raman transitions have already been proposed as means to avoid the classical uncertainty on the initial time of the emission process (the so-called time-jitter) [19, 20] and to tune both the temporal profile and the central frequency of the emitted wavepacket [4]. Here we show that such flexibility can be exploited to generate two nearly indistinguishable photons, in spite of the spectral differences between their respective semiconductor sources.

2. Method

We specifically consider the case where each source is represented by two coherently coupled SAQDs (often referred to as artificial molecule, AM), embedded in an optical microcavity (MC) (Fig. 1). The AM is doped with an excess electron, whose levels 1 and 2 [Fig. 2(a)] define a pseudo-spin, corresponding to the hybridized – bonding and antibonding – states of the two dots [21, 22]; together with the lowest charged exciton states (3 and 4), these form a double Λ scheme [23]. There, a Raman transition can be induced by an off-resonant laser pulse (ω_L), that drives the AM from 1 to 2, while generating a cavity photon (ω_c).

We simulate the dynamics of the two sources (A and B) within a density-matrix approach. The state of each AM-MC system is denoted by |j, n⟩, where j = 1, ..., 4 specifies the AM eigenstate, whereas n represents the occupation number of the cavity mode. The time evolution of the density matrix within such Hilbert space is given by the master equation \( \dot{\rho} = i[H_{MC} + H_L] + \mathcal{L} \rho \) (rotating-wave approximation, \( \hbar = 1 \)) [18]. The coupling of the AM with the MC is accounted by

\[
H_{MC} = \sum_{j=1,2} \sum_{k=3,4} g_{jk}(\sigma_{jk} a^\dagger + a \sigma_{jk}^\dagger),
\]

where \( a \) is the annihilation operator of the cavity photon, \( \sigma_{jk} \equiv |j⟩⟨k| \) the ladder operator acting...
Fig. 2. (Color online) (a) Relevant level scheme for each negatively-charged AM. The levels 1 and 2 correspond to the bonding and antibonding states of the excess electron; these are optically coupled to the two lowest charged exciton states 3 and 4. The off-resonant laser pulse ($\omega_L$) induces a virtual Raman transition from 1 to 2; this process results in the creation and emission of a photon from the fundamental mode of the MC ($\omega_c$). (b) Schematics of the Hong-Ou-Mandel setup. The photons are emitted by the two sources A and B in the input modes of a balanced beam-splitter (BS), whose output modes are coupled to the photodetectors C and D.

on the AM state, and $g_{jk}$ the corresponding dot-cavity coupling constant. The coupling of the AM with the exciting laser is given by

$$H_L = \frac{1}{2} \sum_{p=1}^{N_L} \sum_{j=1,2} \sum_{k=3,4} \Omega_{jk}^p(t) \left( e^{-i\delta_{jk}^p} \sigma_{jk} + H.c. \right),$$

where $\Omega_{jk}^p(t)/g_{jk} = \Omega_0^p \exp\{(t - \tau_0^p)^2/2\sigma_p^2\}$ gives the time envelope of the $p$-th laser pulse, $\delta_{jk}^p = \epsilon_k - \epsilon_j - \omega_L^p$, and $\epsilon_j$ the AM levels. Having found no evidence of an increased indistinguishability arising from non-gaussian laser-pulse profiles, in the calculations presented hereafter we set $N_L = 1$. The interaction of the AM-MC system with the environment is given by the superoperator

$$\mathcal{L} = \sum_j L(\sqrt{\gamma_j} P_j) + \sum_{jk} L(\sqrt{\Gamma_{jk}} \sigma_{jk}) + L(\sqrt{\kappa} a),$$

where $L(A)\rho \equiv (2A\rho A^\dagger - A^\dagger A\rho - \rho A^\dagger A)/2$ and $P_j \equiv |j\rangle \langle j|$ (see [18]). The three terms in Eq. 3 account for pure dephasing (with rate $\gamma$), radiative ($jk = 13, 14, 23, 24$) and non-radiative ($jk = 12, 34$) relaxation of the AM, and cavity-photon emission, respectively. In the following, we assume for simplicity that $\Gamma_{jk} \equiv \Gamma_r$ for all the radiative relaxation processes of the AM, and $\Gamma_{jk} \equiv \Gamma_{nr}$ for the non-radiative ones.

The degree of indistinguishability between the photons generated by the two sources can be measured within the Hong-Ou-Mandel setup [24] [Fig. 2(b)]. There, if two indistinguishable photons enter the input ports (A and B) of a balanced beam splitter, two-photon interference results in a vanishing probability of a coincidence event ($P_{CD} = 0$) in the two photodetectors at the output modes (C and D). For distinguishable photons, instead, $P_{CD} = 1/2$. The coincidence probability can thus be regarded as a measure of the photon indistinguishability. Formally, $P_{CD}$
can be connected to the dynamics of the A and B sources through [19] $P_{CD} = F_1 - F_2$, where $F_{p=1,2} = k^2 \int dt \int d\tau f_p(t, \tau)$ and

$$f_1(t, \tau) = \left[ n_A(t + \tau) n_B(t) + n_B(t) n_A(t + \tau) \right] / 4,$$

$$f_2(t, \tau) = \text{Re} \left\{ \left[ G_A^{(1)}(t, t + \tau) \right]^* G_B^{(1)}(t, t + \tau) \right\} / 2.$$ 

Here, $G_A^{(1)}(t, t + \tau) = \langle a_A^\dagger(t) a_A(t + \tau) \rangle$ and $n_A(t) = \langle a_A^\dagger(t) a_A(t) \rangle$ are, respectively, the first-order coherence function and cavity-mode occupation corresponding to the sources $\chi = A, B$. 

In order to maximize the overlap between the wavepackets of the photons emitted by A and B, we optimize the driving laser pulses and the frequency of the cavity mode, i.e., the vector $X = (\Omega_B^0, \nu_B, \sigma_B, \omega_A)$. For two given sources [each characterized by the vector $Y = (\epsilon_j, g_{jk}, \gamma_j, \Gamma_{jk})$, with $Y_A \neq Y_B$], the suitability of each set of laser pulses for the generation of two indistinguishable photons is quantified by a fitness function $\mathcal{F}(X_A, X_B|Y_A, Y_B) \geq 0$, defined as

$$\mathcal{F} = (F_2/F_1) g(P_e^A, P_e^B).$$  

Here, $0 \leq F_2/F_1 \leq 1$ measures the degree of indistinguishability between the two photons, whereas $g$ accounts for the statistics of the two SPSs; more specifically, it imposes a penalty on the vectors $X, Y$ corresponding to a photon-emission probability $P_e^A = \int n_A(t) dt$ far from 1. We identify the best solution ($X_B^0$) with the vectors that correspond to the maximum value of the fitness function, for the given values of the physical parameters that characterize the SPSs ($Y$): $\mathcal{F}_M = \mathcal{F}(X_B^0, X_B^0|Y_A, Y_B)$. Hereafter, we take: $g = \theta(P_e^A - P_e^B) \cdot \theta(P_e^B - P_e^A)$, where $\theta$ is the Heaviside function and $P_e^j$ the threshold photon-emission probability. This functional dependence of $\mathcal{F}$ on $P_e^A$ and $P_e^B$ allows to preselect the fittest individuals before computing the time integrals of the correlation functions, thus speeding up the optimization procedure. Besides, provided that $P_e^A, P_e^B > P_e^0$, $1 - \mathcal{F}$ coincides with $P_{CD}/(P_e^A \cdot P_e^B)$, which has a clear physical interpretation: the overlap between the quantum states of the two emitted photons.

To this aim, we combine the density matrix approach with a genetic algorithm [25]; this allows to efficiently explore, within a large parameter space, the particularly complex landscape induced by the penalty function $g$.

3. Results

The distinguishability between the photons emitted by the two sources mainly arises from the differences between their transition energies ($\omega_A^0 \neq \omega_B^0$) and oscillator strengths ($g_{jk}^A \neq g_{jk}^B$). In Fig. 3 we show the dependence of the maximum fitness for optimized laser pulses, $\mathcal{F}_M$, on $g_{jk} \equiv g_{jk}^A / g_{jk}^B$ and $\Delta_{AB} = \omega_{12}^A - \omega_{12}^B$ [26], for a threshold emission probability $P_e^0 = 0.9$. The fact that $\mathcal{F}_M < 1$ even for the case of identical sources ($g_{AB} = 1$ and $\Delta_{AB} = 0$) is due to the presence of dephasing and non-radiative relaxation (see below). By adjusting the laser pulses, moderate differences between the oscillator strengths of the two sources ($g_{AB} \geq 0.9$) can be efficiently compensated, whereas decreases of $\mathcal{F}_M$ arising from mismatches between the optical-transition energies of A and B in the meV range can be limited to a few percent.

A couple of general comments are in order. On the one hand, stimulated Raman processes do increase the tolerance to inhomogeneities between two SPSs from the $\mu$eV – as is the case with spontaneous emission – to the meV range. In order for this to be possible, the two sources need to be excited by two different and suitably optimized laser pulses. On the other hand, such inhomogeneities cannot be completely compensated by properly tuning the frequencies of the driving laser pulses, as could be naively expected on the basis of a simple energy-conservation
Maximized fitness function $F_M$ as a function of the differences between the A and B sources in terms of frequencies ($\Delta_{AB} = \omega_A^{\lambda} - \omega_B^{\lambda}$) and oscillator strengths ($g_{AB} \equiv g_{jk}^B / g_{jk}^A$) of the optical transitions. The remaining parameters are: $\Gamma_{nr} = \Gamma_A / B = 10^{-3}$ ps$^{-1}$, $\gamma_A / B = 10^{-2}$ ps$^{-1}$, $\kappa_A / B = 10^{-1}$ ps$^{-1}$, $g_{jk}^A = 0.120$ meV.

These limitations are partially due to the effects of decoherence; besides, the requirement that the two sources emit photons with a high probability seems to conflict with that of maximizing their indistinguishability. In the following we investigate separately these two aspects.

The coupling between the carriers confined in each AM and the phonons gives rise to energy relaxation and dephasing. These two contributions are considered separately in Fig. 4(a), where we report $F_M$ as a function of $1/\gamma$ (with $\Gamma_{21} = 0$, squares), and of $1/\Gamma_{21}$ (with $\gamma = 0$, triangles), within realistic ranges of values for these rates. As in the case of two photons sequentially emitted by the same source [19], the coincidence probability increases linearly with $\gamma$ (dotted green curve). A stronger dependence is found with respect to the non-radiative relaxation between the two lowest states in the $\Lambda$-scheme (a fit by a quadratic function of $\Gamma_{21}$ is given by the dotted red line). We note, however, that the value of $\Gamma_{21}$ can be strongly reduced by suitably engineering the AM geometry and by the application of external fields [27, 28].

In order to investigate the relation between the emission probability of the A and B photons and their indistinguishability, we have computed $F_M$ as a function of $P_{te}$. In Fig. 4(b) we report two representative cases: $\Delta_{AB} = 0$ (orange squares) and $\Delta_{AB} = 1.5$ meV (gray triangles), with $g_{AB} = 1$. The photon emission probability and the degree of indistinguishability are clearly anti-correlated, thus demonstrating the existence of a trade-off between the two requirements. The dependence of $F_M$ on $P_{te}$, though evident even for identical sources ($\Delta_{AB} = 0$), is more pronounced in the presence of a spectral mismatch ($\Delta_{AB} = 1.5$). The above curves are relatively insensitive to differences in the oscillator strengths ($g_{AB} \geq 0.9$, not shown here).

We finally comment on the robustness of the solution with respect to small departures of the control parameters from their optimal values. As a representative example, we report the dependence of $P_D = 1 - P_{CD}$ on the cavity frequency $\omega_t \equiv \omega_t^A = \omega_t^B$, referred to its optimized value $\omega_t^0$ (Fig. 5 (a)). The robustness of the photon indistinguishability with respect to non-optimal parameters decreases for increasing spectral mismatches between the two sources. Besides, our simulations show a stronger dependence of $P_{CD}$ on the laser (solid and dotted lines) than on the cavity frequencies (dashed). This feature can be traced to the fact that the uncertainty on the cavity frequency ($\sim 1/\kappa = 10$ ps) is larger than that on the laser frequency (typical
duration of the optimized laser pulses are few tens of ps). As to $P_e^{A/B}$ [Fig. 5 (b)], we note that the value corresponding to the optimized parameter ($\Delta \omega_c = 0$) is close to the minimum allowed value ($P_e^0$): this provides a further indication on the existence of a trade-off between photon indistinguishability and emission probability. Besides, the existence of a spectral mismatch (gray lines) makes also the fulfillment of the requirement $P_e^A, P_e^B > P_e^0$ more sensitive to the tuning of the physical parameters. The required precision is of the order of a few $\mu$eV for $\Delta \omega_c = 1.5$ meV. Being $|\omega_c^A - \omega_c^B|$ of the same order of the spectral mismatch between the two sources, this implies that the emission of the two photons cannot be triggered by a same laser pulse.
4. Conclusions

In conclusion, we have investigated the degree of indistinguishability between single photons emitted by non-identical artificial molecules. We found that the use of virtual Raman transitions, combined with the optimal design of the driving laser pulses, increases the tolerance with respect to the spectral inhomogeneities between the sources up to the meV energy range. Defined power-law dependences of the coincidence probability on the dephasing and (non-radiative) relaxation rates are identified. Besides, unlike the case of identical sources, a trade-off emerges between the requirements of maximizing the emission efficiency and the photon indistinguishability. It is finally worth mentioning that the use of a pseudo-spin within the Λ-level scheme offers promising possibilities [31]. These include the use of quantum-confined Stark effect [29] for triggering the photon emission [30], and the entangling of carrier spins localized in remote (and thus non-identical) semiconductor systems [32]. The latter goal requires the combination of a spin-photon entangling [33] process with two-photon interference within the Hong-Ou-Mandel setup.

Acknowledgments

We acknowledge financial support from the Italian MIUR under FIRB Contract No. RBIN01EY74.