Ultrafast optical signature of quantum superpositions in a nanostructure

F. J. Rodríguez,[1] L. Quiroga,[1] and N. F. Johnson[2]

[1]*Departamento de Física, Universidad de Los Andes, A.A. 4976, Bogotá D.C., Colombia*
[2]*Physics Department, Clarendon Laboratory, Oxford University, Oxford OX1 3PU, United Kingdom*

We propose an unambiguous signature for detecting quantum superposition states in a nanostructure, based on current ultrafast spectroscopy techniques. The reliable generation of such superposition states via Hadamard-like quantum gates is crucial for implementing solid-state based quantum information schemes. The signature originates from a remarkably strong photon antibunching effect which is enhanced by non-Markovian dynamics.

The possibility of performing quantum information processing in nanostructure systems is of great interest from the perspectives of both fundamental science and future emerging technologies. Quantum dots (QDs) are solid-state nanostructures which are analogs of real atoms;[1] semiconductor QDs and macromolecules are seen as excellent candidates for performing quantum information processing tasks.[2–5] There are also many inorganic and organic structures that qualify as "nanostructures" and may therefore become good candidates: e.g., carbon buckyballs, and even microbiological molecular structures such as the photosynthetic complexes in purple bacteria.[6] Essential steps toward the implementation of standard quantum information schemes in such nanostructures include: (1) the identification of the basic qubit (quantum bit), and (2) the application of one- and two-qubit quantum gates in order to generate quantum superpositions and entanglement. An important example of a one-qubit gate is the Hadamard-like gate, since it generates a superposition state (e.g., $|0\rangle+|1\rangle$) from an unsuperposed initial state (e.g., $|0\rangle$). Reference 2 showed that (1) and (2) can be achieved with excitons generated using current techniques in ultrafast optical spectroscopy.[7] Several experimental groups are now actively pursuing this route. Such qubit control has already been achieved in neutral atoms[8] and ions in an ion trap,[9] however scalability issues may limit such non-solid-state devices to just a few qubits.

However there is a crucial third step, which is to verify the reliability of the quantum superpositions (entanglements) generated in the single nanostructure (pair of nanostructures) by the one-qubit (two-qubit) quantum gate. How can we show experimentally that we have generated such superposition (entanglement)? The present work addresses this question for the important first step of a single nanostructure (e.g., a single QD). Specifically, we consider the ultrafast second-order coherence function of the emitted light from the optically generated exciton in a single nanostructure (e.g., a QD). This quantity $g^{(2)}$ is calculated for the QD interacting with two baths: (i) photon environment and (ii) a phonon system. A strong antibunching effect is predicted in the resonance fluorescence response at very short times, *if and only if the initial exciton state comprises a quantum superposition*. This strong effect does not arise for initial states described by a statistical mixture, i.e., nonsuperposed states. We show numerically, that non-Markovian effects sig-

nificantly enhance the antibunching signal, hence demonstrating that temporal correlations *cannot* be neglected *a priori* in such ultrafast regimes. In addition to the quantum superposition test, our results may prove useful in designing photon-emitting devices with controllable and accurate emission rates.[10]

In atomic systems, resonance fluorescence experiments have already proved themselves to be extremely valuable.[11] Similar experiments in solid-state systems have only recently been performed. In particular it has been demonstrated that for a single CdSe QD at room temperature[12,13] and a single self-assembled InAs QD at cryogenic temperatures,[14,15] strong antibunching effects are observed in fluoresence experiments. This provides direct evidence that single QDs present the same kind of nonclassical light emission as a single two-level atom. The present paper provides a further stimulus to experimentalists to improve resolution times, with the potential payoff that antibunching measurements will then provide a direct probe of the initial quantum state.

The solid-state system of interest here comprises a nanostructure (e.g., a QD) of any shape, coupled to the electromagnetic field and to a heat bath, represented by a set of harmonic oscillators, which provides the basic source of temperature dependence. The light source is of low intensity hence the number of excitons generated is small. A single exciton in its ground state can be described by a two-level system.[2] The Hamiltonian is given by

$$H = \frac{\epsilon}{2}\sigma_z + \sum_k \omega_k a_k^\dagger a_k + \sum_k (g_k \sigma^+ a_k + g_k^* \sigma^- a_k^\dagger)$$
$$+ \sum_q \Omega_q b_q^\dagger b_q + \sum_q \sigma_z (f_q b_q + f_q^* b_q^\dagger)$$
$$+ E(t)\sigma^+ e^{i\omega t} + c.c. \quad (1)$$

where a_k and b_q represent the annihilation operators for photons and phonons respectively, ϵ is the total exciton energy, $\omega_k(\Omega_q)$ denotes the photon (phonon) frequencies, $g_k(f_q)$ the exciton-photon (exciton-phonon) coupling, and $E(t)$ describes the envelope of a classical source of light of frequency ω acting on the QD. The exciton population is described by $\sigma_z = |X\rangle\langle X| - |0\rangle\langle 0|$ where $|X\rangle$ stands for a one-

exciton state while $|0\rangle$ denotes the QD vacuum, i.e., no exciton. Similarly the raising and lowering pseudospin operators are $\sigma^+ = |X\rangle\langle 0|$ and $\sigma^- = |0\rangle\langle X|$, respectively. From Eq. (1) it is seen that the photon field is associated with the dissipative dynamics of the QD whereas the phonon field is responsible for dephasing effects. This phonon dephasing accounts for the temperature effects. The fundamental band gap is typically much larger than $k_B T_e$ (T_e is the temperature) hence the photon field can be taken as remaining at zero temperature. Non-Markovian effects are included for both exciton-photon and exciton-phonon couplings. One of the advantages of this model is that it is reasonably simple, yet sufficiently complex to manifest many important features of the ultrafast response of nanostructures. More refined models should consider multiexciton complexes if a strong excitation is applied. In that case, joint effects coming from non-Markovian processes as described here, together with strong particle correlations, should be observable. However, these kinds of effects are beyond the scope of the present work.

We employ a master equation of Lindblad form, since this kind of equation can properly account for the coupling of the QD system to its environment—it can also go beyond the Markov approximation using time dependent damping coefficients.[16] We stress that master equations with time-independent damping coefficients are unable to account for the evolution of an open system on very short time scales. At resonance, i.e., $\omega = \epsilon$, and using the rotating wave approximation, the Liouvillian acting on any QD operator O is given by

$$L(t)O = -\frac{i}{\hbar}[E\sigma^+ + E^*\sigma^-, O]$$
$$+ \gamma_{relax}(t)\left[\sigma^- O\sigma^+ - \frac{1}{2}O\sigma^+\sigma^- - \frac{1}{2}\sigma^+\sigma^- O\right]$$
$$+ \gamma_{dph}(T_e,t)[\sigma_z,[\sigma_z,O]], \qquad (2)$$

where the coupling to photons $\gamma_{relax}(t)$, and the coupling to phonons $\gamma_{dph}(T_e,t)$, include non-Markovian effects through their time dependences. Temperature effects do not need to be included for γ_{relax} in the coupled light-QD subsystem, since $\hbar\epsilon \gg k_B T_e$. Solving the Liouville equation for the QD density matrix ρ, and for different Rabi frequencies $\Omega = \mu|E|/\hbar$ (μ is the dipole moment), expectation values for any QD operator may be evaluated and the characteristics of the emitted photon field hence obtained.

The coherence properties of the emitted photon field can be properly accounted for by the second-order coherence function given by[17]

$$g^{(2)}(T,\tau) = \frac{\langle\sigma^+(T)\sigma^+(T+\tau)\sigma^-(T+\tau)\sigma^-(T)\rangle}{\langle\sigma^+(T)\sigma^-(T)\rangle\langle\sigma^+(T+\tau)\sigma^-(T+\tau)\rangle}, \qquad (3)$$

where $\tau = t_1 - t_2$ represents the time difference between the photons' arrival at the detector and $T = t_1 + t_2/2$. This coherence function can be expressed in a very simple form, valid for any initial condition of the QD, as follows:
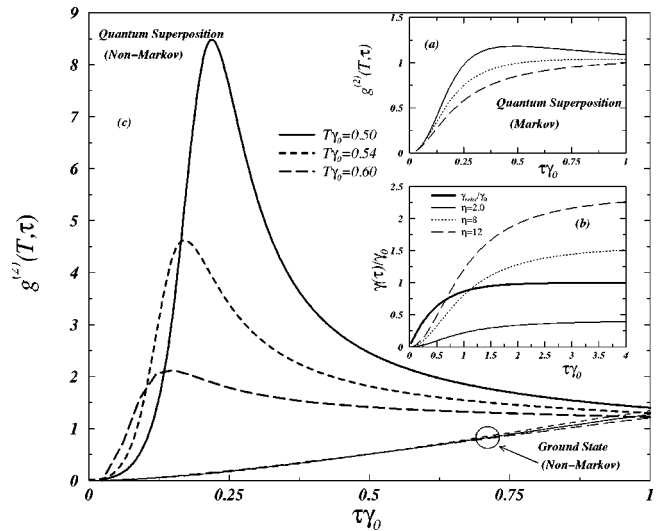

FIG. 1. Second-order coherence function in the Markov approximation for $\eta = 2.85$, $\lambda = 2.0\gamma_0$, and $\Omega = 2.25\gamma_0$. (a) $g^{(2)}$ for three different detecting times in a quantum superposition (QS) initial state using the Markov approximation. (b) Time evolution of the decay rate $\gamma(t)$ and pure dephasing rate $\gamma_{dph}(T_e,t)$ for different temperatures. (c) $g^{(2)}$ including non-Markovian effects for initial QS and ground state (GS).

$$g^{(2)}(T,\tau) = \frac{\langle X|\mathcal{T}e^{\int_0^\tau L(t')dt'}(|0\rangle\langle 0|)|X\rangle}{\rho_{X,X}(T+\tau)}, \qquad (4)$$

where $\mathcal{T}$ denotes the time-ordering operator and ρ is the QD reduced density matrix. By changing the initial preparation state of the QD, the value of the nonstationary second-order coherence function should change through its dependence on $\rho_{X,X}$. A closed expression for $g^{(2)}$ and the antibunching effect, characterized by the growth of $g^{(2)}$ from zero for $\tau = 0$, has been well documented *in the steady-state* situation, with nonzero relaxation decay γ_0 and with $\gamma_{dph} = 0$.[17] However an expression for Eq. (4) in closed form for the ultrafast regime, valid for any T and τ, is not available. A numerical solution of the Bloch equations for the different elements of the QD reduced density matrix must therefore be performed. Our present work represents a far more general study of the variations of $g^{(2)}$, and includes the effect of different initial QD states. In particular, it is interesting to quantify the variations of $g^{(2)}$ for the following initial conditions: (i) a pure state comprising a quantum superposition (QS) of type $|\Psi\rangle = 1/\sqrt{2}(|0\rangle + i|X\rangle)$ where $\rho(0) = |\Psi\rangle\langle\Psi|$, (ii) the usual experimental case in which the QD is prepared in its ground state (GS) given by $\rho(0) = |0\rangle\langle 0|$, and (iii) a statistical mixture (SM) of states $\rho(0) = 1/\sqrt{2}(|0\rangle\langle 0| + |X\rangle\langle X|)$. The initial photon state is the vacuum.

We start by examining the system's qualitative behavior within the Markov approximation. Results for $g^{(2)}$ are shown in Fig. 1(a) for a single QD containing up to one electron-hole pair. The experimentally obtained decay rate[7,18] $\hbar\gamma_{relax} = \hbar\gamma_0 = 20\mu$ eV and $\gamma_{dph} = 0.5\gamma_0$ are used in the calculations. The Rabi frequency is $2.25\gamma_0$ ($\mu \approx 15$ Debyes). A clear sub-Poissonian character is observed at very short times. The enhancement property of $g^{(2)}$ can be readily un-

derstood from Eq. (4), in particular from a vanishing value of the element $\rho_{X,X}$ of the QD density matrix. The second-order time correlation function can be written as $g^{(2)}(T,\tau) = (\rho_{X,X}(\tau)|_{\rho(\tau=0)=|X\rangle\langle X|})/\rho_{X,X}(T,\tau)$, where the numerator represents the density matrix element given that the QD is in its ground state. By solving the Bloch equations for ρ at very short times, it can be seen that this enhancement appears for $\tau \approx (\Omega - \gamma_0)^{-1}$, i.e., when $\rho_{X,X}(T,\tau) \to 0$ in agreement with Fig. 1(a). We stress that this condition *cannot* be obtained if the system is initially prepared in its GS or in a SM of states (not shown). Previous experiments[14] with their limited resolution times could not have detected this feature because the correlations always vanish for long detection times.

Since this antibunching behavior occurs at very short times, we will now consider the quantitative effects of non-Markovian behavior characterized by time-dependent damping rates. In Ref. 16, an explicit expression for $\gamma_{relax}(t)$ is given as

$$\gamma_{relax}(t) = \frac{2\gamma_0 \sinh(dt/2)}{(d/\lambda)\cosh(dt/2) + \sinh(dt/2)} \quad (5)$$

where $d = \sqrt{\lambda^2 - 2\gamma_0\lambda}$, γ_0 is the constant Markov decay rate (time-independent), and γ_0/λ is the ratio between the photon reservoir correlation time and a typical time scale on which the QD exciton changes. For $\gamma_0/\lambda \ll 1$, this yields $\gamma_{relax}(t)/\gamma_0 = 1 - e^{-\lambda t}$. This explicit expression for $\gamma_{relax}(t)$ is appropriate for a Lorentzian photon reservoir.[19] A memoryless photon environment corresponds to λ going to infinity in which case the Markov situation is recovered. It is worth noting that by slightly changing this form of $\gamma_{relax}(t)$, non-Markovian effects in a structured photon environment such as a microcavity could also be modeled.[16]

For the pure dephasing rate, the standard form of the independent boson model[20] is taken as

$$\gamma_{dph}(T_e,t) = \sum_q |f_q|^2 \coth\left(\frac{\Omega_q}{2T_e}\right)\frac{1-\cos(\Omega_q t)}{\Omega_q^2}. \quad (6)$$

In the continuum limit for phonon $\vec{q}$ vectors, all information about the bath which is essential to the dynamics of the QD is contained in the compact form of the spectral density function $J(\omega) = \pi/2\Sigma_q|f_q|^2\delta(\omega - \Omega_q)$. An appropriate choice for the spectral function $J(\omega)$, and its associated cutoff frequency, can be made according to the QD environment. In terms of the Debye model, the natural cutoff is the Debye frequency (ω_D) yielding a spectral function of the form $J(\omega) \sim \omega^3 e^{-\omega/\omega_D}$. An explicit form for γ_{dph}, with the latter choice for $J(\omega)$, has been derived by Palma *et al.*[21] For different temperatures ($\eta = T_e/\omega_D$), $\gamma_{relax}(t)$ and γ_{dph} are shown in Fig. 1(b). On very short time scales the effective decay rates for both processes, radiative and pure dephasing, are very low indicating that Markov approximations (which are valid on long time scales) overestimate the decay effects at short times. As demonstrated, this explicit form of $\gamma_{relax}(\tau)$ and $\gamma_{dph}(T_e,\tau)$ leads to an inefficient damping at times $\tau < \gamma_0^{-1}$ hence the Markov approximation overestimates the damping effects in the ultrafast regime. Therefore,
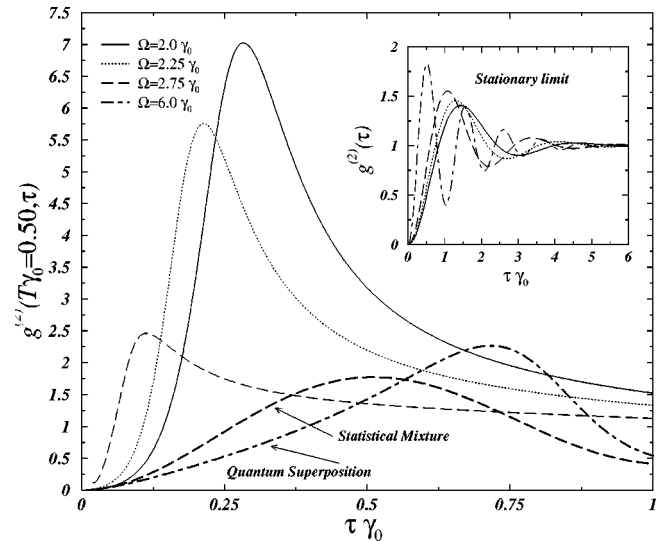

FIG. 2. Second-order coherence function, including non-Markovian effects, for a quantum superposition (QS) and a statistical mixture (SM) of states. Results shown for various Rabi frequencies $\Omega = 2\gamma_0$ (continuous line), $\Omega = 2.25\gamma_0$(dotted-line), $\Omega = 2.75\gamma_0$(dashed-line), $\Omega = 6\gamma_0$(thick dot-dashed line). Here $\eta = 2.85$, $\lambda = 2.0\gamma_0$, and $T\gamma_0 = 0.5$. Inset: the stationary limit for the system prepared in a QS initial state.

any antibunching enhancement that occurs on a very short time scale should be reinforced by non-Markovian phenomena. A demonstration of this statement is shown in Fig. 1(c) with $\eta = 2.85$ and $\Omega = 2.25\gamma_0$, for different resolution detecting times. Clearly, the QS shows a strong enhancement compared with a GS. The second-order time correlation function at very short times therefore provides information about the initial state preparation. The main result of the present work is that for times smaller than the time scale in which dissipation of energy takes place, a strong antibunching effect is predicted. Within this time scale the correlations between the dot and the environment are not very important and clearly nonclassical light emission is enhanced. As a consequence, antibunching enhancement should be most prominent in QDs fabricated from II-VI semiconductors, III-V nitrides, organic materials, or hybrid heterostructures due to the fact that in these systems exciton-photon coupling is not necessarily weak and non-Markovian effects can be important.

Figure 2 shows how the peak in $g^{(2)}(T\gamma_0 = 0.5, \tau)$ develops a symmetric shape, growing quite large at low Rabi frequencies. For large Rabi frequencies the antibunching for QS initial state is observed at very short times and the light emission begins to evolve into a mere statistical mixture of states. Crucial information concerning the state in which the system was initially prepared can hence be obtained via a measurement of $g^{(2)}$. Our results therefore show that QS states can be far more robust to decoherence than other initial states at very short times. We conjecture that this robustness may also hold for multiple qubits (see also Ref. 22). We now turn our attention to the long time behavior ($\tau \gg \gamma_0^{-1}$) for $g^{(2)}(\tau)$. In the inset of Fig. 2, the antibunching effect is

clearly observed, however the peak intensities are not as large as those predicted for the QS state. To avoid confusion concerning this sub-Poissonian characteristic, we stress that this peak intensity comes from the natural Rabi oscillations of the coupled QD-light subsystem. The antibunching signal reported in this work is observed when the vacuum fluctuations dominate, if and only if the system is prepared in a QS. Interestingly the QS initial state is rather insensitive to the decoherence mechanism, for typical experimental values and at very short times. Experimentally, the effects presented here could be observed using ultrafast spectroscopy with a resolution of the order of the dephasing times, e.g., 50 ps for a semiconductor QD. We also note that different initial QS states can be probed by tailoring the polarization of the driving field.

In summary, we have shown that ultrafast fluorescence intensity-correlation measurements in single QDs provide a sensitive probe not only of the photonic and phononic environment, but also of memory effects such as those determined by specific initial state preparation. At the heart of our results is an enhancement of the lack of photon coincidence on short time scales as a result of the exciton superposition state. As a side product of our findings, the transformation from sub-Poissonian ($g^{(2)} < 1$) to super-Poissonian ($g^{(2)} > 1$) behavior on short time scales may be of practical interest in the design of devices which act as triggered single-photon sources. This paper has concentrated on the basic building block comprising single nanostructures and superpositions created by the application of one-qubit gates. Non-classical light features in the fluorescence of coupled QD systems will be dealt with elsewhere.

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