Vacuum-field Rabi oscillations in atomically doped carbon nanotubes

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We report a strictly non-exponential spontaneous decay dynamics of an excited two-level atom placed inside or at different distances outside a carbon nanotube. This is the result of strong non-Markovian memory effects arising from the rapid frequency variation of the photonic density of states near the nanotube. The system exhibits vacuum-field Rabi oscillations when the atom is close enough to the nanotube surface and the atomic transition frequency is in the vicinity of the resonance of the photonic density of states.

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It has long been recognized that the spontaneous emission rate of an excited atom is not an immutable property, but that it can be modified by the atomic environment. Generally called the Purcell effect, the phenomenon is qualitatively explained by the fact that the local environment modifies the strength and distribution of the vacuum electromagnetic modes with which the atom can interact, resulting indirectly in the alteration of atomic spontaneous emission properties.

The Purcell effect took on special significance recently in view of rapid progress in physics of nanostructures. Here, the control of spontaneous emission has been predicted to have a lot of useful applications, ranging from the improvement of existing devices (lasers, light emitting diodes) to such nontrivial functions as the emission of nonclassical states of light. In particular, the enhancement of the spontaneous emission rate can be the first step towards the realization of a thresholdless laser or a single photon source. The possibility to control atomic spontaneous emission was shown theoretically for microcavities and microspheres, optical fibers, photonic crystals, semiconductor quantum dots. Recent technological progress in the fabrication of low-dimensional nanostructures has enabled the experimental investigation of spontaneous emission for microcavities, photonic crystals, quantum dots.

Recent successful experiments on encapsulation of single atoms into single-wall carbon nanotubes (CNs) and their intercalation into single-wall CN bundles stimulate an analysis of atomic spontaneous emission properties of such systems. Typically, there may be two qualitatively different regimes of the interaction of the excited atomic state with a vacuum electromagnetic field in the vicinity of the CN. They are the weak coupling regime and the strong coupling regime. The former is characterized by the monotonous exponential decay dynamics of the upper atomic state with the decay rate altered compared with the free-space value. The latter is, in contrast, strictly non-exponential and is characterized by reversible Rabi oscillations where the energy of the initially excited atom is periodically exchanged between the atom and the field. In this Letter, we develop the quantum theory of the spontaneous decay of a two-level atom near a CN, derive the evolution equation of the upper state of the system and, by solving it numerically, demonstrate the strictly non-exponential spontaneous decay dynamics in the case where the atom is close enough to the CN surface. In certain cases, the system exhibits vacuum-field Rabi oscillations – a result already detected for quasi-2D excitonic and intersubband electronic transitions in semiconductor quantum microcavities and never reported for atomically doped CNs so far.

The quantum theory of the spontaneous decay of excited atomic states in the presence of a CN involves an electromagnetic field quantization procedure. Such a procedure faces difficulties similar to those in quantum optics of 3D Kramers-Kronig dielectric media where the canonical quantization scheme commonly used does not work since, because of absorption, operator Maxwell equations become non-Hermitian. As a result, their solutions cannot be expanded in power orthogonal modes and the concept of modes itself becomes more subtle. We, therefore, use an alternative quantization scheme developed in Ref. 4, where Fourier-images of electric and magnetic fields are considered as quantum mechanical observables of corresponding electric and magnetic field operators. The latter ones satisfy the Fourier-domain operator Maxwell equations modified by the presence of a so-called operator noise current written in terms of a vector bosonic field operator and an imaginary dielectric permittivity. This operator is responsible for correct commutation relations of the electric and magnetic field operators in the presence of medium-induced absorption. The electric and magnetic field operators are then expressed in terms of a continuum set of the bosonic fields by means of the convolution of the operator noise current with a classical electromagnetic field Green tensor of the system. The bosonic field operators create and annihilate single-quantum electromagnetic medium excitations. They are defined by their commutation relations and play the role of the fundamental dynamical variables in terms of which the Hamiltonian of the composed sys-

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system "electromagnetic field + dissipative medium" is written in a standard secondly quantized form.

We consider a two-level atom positioned at the point $r_A$ near an infinitely long single-wall CN. The atom interacts with a quantum electromagnetic field via an electric dipole transition of frequency $\omega_A$. The atomic dipole moment, $d = d_z e_z$, is assumed to be directed along the CN axis assigned by the unit vector $e_z$ of the orthonormal cylindrical basis \{$e_r, e_\varphi, e_z$\}. The contribution of the transverse dipole moment orientation is suppressed because of the strong depolarization of the transverse field in an isolated CN [13]. The Hamiltonian of the system is given by

$$\hat{H} = \int d\mathbf{R} \int_0^\infty \frac{d\omega}{2} \hbar \omega \hat{f}^\dagger(\mathbf{R}, \omega) \hat{f}(\mathbf{R}, \omega) + \frac{1}{2} \hbar \omega_A \hat{\sigma}_z$$

$$- \left[ \hat{\sigma}_+ \hat{E}_z^{(+)}(r_A) d_z + \text{h.c.} \right]$$

with the three terms representing the electromagnetic field (modified by the presence of the CN), the two-level atom and their interaction (within the framework of electric dipole and rotating wave approximations [3]), respectively. The operators $\hat{f}^\dagger(\mathbf{R}, \omega)$ and $\hat{f}(\mathbf{R}, \omega)$ are the scalar bosonic field operators defined on the CN surface ($\mathbf{R} = \{R_{cn}, \phi, z\}$ is the radius-vector of an arbitrary point of the CN surface). They play the role of the fundamental dynamical variables of the field subsystem and satisfy the standard bosonic commutation relations. The Pauli operators $\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z$ describe the atomic subsystem. The operators $\hat{E}_{\pm}(r_A)$ represent the electric field the atom interacts with. For an arbitrary $r = \{r, \varphi, z\}$, they are defined as $\hat{E}(r) = \hat{E}^{(+)}(r) + \hat{E}^{(-)}(r)$ with

$$\hat{E}^{(+)}(r) = \int_0^\infty \hat{E}(r, \omega) d\omega, \quad \hat{E}^{(-)}(r) = [\hat{E}^{(+)}(r)]^\dagger.$$  

Here, $\hat{E}$ satisfies the Fourier-domain Maxwell equations

$$\nabla \times \hat{E}(r, \omega) = -ik \hat{H}(r, \omega),$$

$$\nabla \times \hat{H}(r, \omega) = -ik \hat{E}(r, \omega) + \frac{4\pi}{c} \hat{I}(r, \omega),$$

where $\hat{H}$ stands for the magnetic field operator [defined by analogy with Eq. (2)], $k = \omega/c$, and

$$\hat{I}(r, \omega) = \int d\mathbf{R} \delta(r - \mathbf{R}) \hat{J}(r, \omega) = 2\hat{J}(R_{cn}, \varphi, z, \omega) \delta(r - R_{cn})$$

$$\hat{J}(r, \omega) = \sqrt{\frac{\hbar \omega}{2\pi}} \sigma_{zz}(r, \omega)/\pi \hat{f}(r, \omega) e_z$$

being the operator noise current density associated with CN-induced absorption ($\sigma_{zz}$ is the CN surface axial conductivity per unit length; in describing CN electronic properties we use the model of the axial conductivity [24]).

From Eq. (3) it follows that

$$\hat{E}(r, \omega) = i\frac{4\pi}{c} k \int d\mathbf{R} \hat{G}(r, \mathbf{R}, \omega) \cdot \hat{J}(r, \omega)$$

[and $\hat{H} = (ik)^{-1} \nabla \times \hat{E}$ accordingly], where $\hat{G}(r, \mathbf{R}, \omega)$ is the Green tensor of the classical electromagnetic field in the vicinity of the CN. Its components satisfy the equation

$$\sum_{\alpha=\mathbf{r}, \varphi, z} \left( \nabla \times \nabla \times - \kappa^2 \right)_{\alpha z} G_{\alpha z}(r, \mathbf{R}, \omega) = \delta(r - \mathbf{R}),$$

with the radiation conditions at infinity and the boundary conditions on the CN surface. The latter ones are obtained from the classical electromagnetic field boundary conditions derived in Ref. [20] by means of simple relations valid for $r \neq r_A$: $\hat{E}_\alpha(r, \omega) = i k G_{\alpha z}(r, r_A, \omega)$ and $H_\alpha = (ik)^{-1} \sum_{\beta=\mathbf{r}, \varphi, z} \epsilon_{\alpha\beta\gamma} \nabla_\beta \hat{E}_\gamma$ (\(\epsilon_{\alpha\beta\gamma}\) is the totally anti-symmetric unit tensor). The Hamiltonian [17] along with Eqs. (2) and (8) is the modification of the Jaynes–Cummings model [17] for an atom in the vicinity of a solitary CN.

When the atom is initially in the upper state and the field subsystem is in vacuum, the time-dependent wave function of the whole system can be written as

$$|\psi(t)\rangle = C_u(t) e^{-i(\omega_A/2)t} |u\rangle |\{0\}\rangle$$

$$+ \int d\mathbf{r} \int_0^\infty d\omega C_I(\mathbf{r}, \omega, t) e^{-i(\omega - \omega_A/2)t} |l\rangle |\{1(\mathbf{r}, \omega)\}\rangle,$$

where $|u\rangle$ and $|l\rangle$ are the upper and lower atomic states, respectively, $|\{0\}\rangle$ is the vacuum state of the field subsystem, $|\{1(\mathbf{r}, \omega)\}\rangle$ is its excited state where the field is in a single-quantum Fock state. In view of Eqs. (4) and (8), the Schrödinger equation with Hamiltonian [11] and wave function [17] yields the following evolution law for the occupation probability amplitude $C_u$ of the upper state

$$C_u(\tau) = 1 + \int_0^\tau K(\tau' - \tau') C_u(\tau') d\tau'$$

$$K(\tau - \tau') = \frac{\hbar \Gamma_0(x_A)}{4\pi x_A^3} \int_0^\infty dx x^3 \xi(x) e^{-i(x - x_A)(\tau - \tau') - 1} \left( \frac{\hbar c}{i x - x_A} \right).$$

Here, $x = \hbar \omega/2\gamma_0$ and $\tau = 2\gamma_0/\hbar$ are the dimensionless frequency and time, respectively, with $\gamma_0 = 2.7$ eV being the carbon nearest neighbor hopping integral appearing in the CN surface axial conductivity in the noise current in Eq. (4), $\xi(x) = \Gamma(x)/\Gamma_0(x)$ is the relative density (with respect to vacuum) of photonic states near the CN with $\Gamma(x) = (8\pi d_z^2/\hbar c^2)(2\gamma_0 x/h)^2 \text{Im} \ G_{zz}(r_A, r_A, x)$ being the rate of the exponential spontaneous decay of the atom near the CN and $\Gamma_0(x)$, its vacuum counterpart, given by the same formula with $\text{Im} \ G_{zz}(r_A, r_A, x)$ being a well-known Volterra integral equation of the second kind. In our case, it describes the spontaneous decay dynamics of the excited two-level atom in the vicinity of the CN. All the CN parameters that are relevant for the spontaneous decay are contained in the classical electromagnetic field Green tensor in Eq. (8).

In the case where the Markovian approximation is applicable, or, in other words, when the atom-field coupling
with increasing the CN radius, representing the decrease of the (5,5), (10,10) and (23,0) CNs. It is seen to decrease as \( \Gamma \) computed in the same manner as it was done in Ref. [18].

The factor inside and at different distances outside achiral CNs structure constant [21].

To take non-Markovian effects into account, we have solved Eqs. (8) and (9) numerically. The solution of the occupation probability \( |C_u(\tau)|^2 \) of the upper state was obtained for the atom placed in the center inside and at different distances outside achiral CNs of different radii. The factor \( \xi(x) \) in Eq. (9) was computed in the same manner as it was done in Ref. [15]. The free-space spontaneous decay rate was approximated as \( \Gamma_0(x_A) \approx \alpha^2 2\gamma_0 x_A / h \) with \( \alpha = 1/137 \) being the fine-structure constant [21].

Figure 1(a) presents \( \xi(x) \) for the atom in the center of the (5,5), (10,10) and (23,0) CNs. It is seen to decrease with increasing the CN radius, representing the decrease of the atom-field coupling strength as the atom moves away from the CN surface [15]. To calculate \( |C_u(\tau)|^2 \) in this particular case, we have fixed \( x_A = 0.45 \) (indicated by the vertical dashed line), firstly, because this transition is located within the visible light range 0.305 < \( x < 0.574 \), secondly, because this is the approximate peak position of \( \xi(x) \) for all the three CNs. The functions \( |C_u(\tau)|^2 \) calculated are shown in Figure 1(b) in comparison with those obtained in the Markovian approximation yielding the exponential decay. The actual spontaneous decay dynam-

FIG. 1: (Color online) Relative density of photonic states (a) and upper-level spontaneous decay dynamics (b) for the atom in the center of different CNs.

FIG. 2: (Color online) Relative density of photonic states (a) and upper-level spontaneous decay dynamics (b, c) for the atom at different distances outside (9,0) CN.
ics is clearly seen to be non-exponential. For the small radius (5,5) CN, Rabi oscillations are observed, indicating a strong atom-field coupling regime related to strong non-Markovian memory effects. With increasing the CN radius, the decay dynamics approaches the exponential one with the decay rate enhanced by several orders of magnitude compared with that in free space.

Figure 2(a) shows $\xi(x)$ for the atom outside the (9,0) CN at the different distances from its surface. The vertical dashed lines indicate the atomic transitions for which the functions $|C_a(\tau)|^2$ in Figures 2(b) and 2(c) were calculated. Both transitions belong to the visible light range, shortest atom-surface distance) while $x_A = 0.52$ is the position of a dip of $\xi(x)$. Very clear underdamped Rabi oscillations are seen for the shortest atom-surface distance at $x_A = 0.33$ [Figure 2(b)], indicating strong atom-field coupling with strong non-Markovity. For $x_A = 0.52$ [Figure 2(c)], though $\xi(0.52)$ is comparatively small, the spontaneous decay dynamics is still non-exponential, approaching the exponential one only when the atom is far enough from the CN surface. Similar to what takes place in photonic crystals [5], this is because of the fact that, due to the rapid variation of $\xi(x)$ in the neighborhood of this frequency, the correlation time of the electromagnetic vacuum is not negligible on the time scale of the evolution of the atomic system, so that atomic motion memory effects are important and the Markovian approximation in Eq. 9 is inapplicable.

The effects predicted will yield an additional structure in optical absorbance/reflectance spectra (see, e.g., 10) of atomically doped CNs in the vicinity of the energy of an atomic transition. Weak non-Markovity of the spontaneous decay (non-exponential decay with no Rabi oscillations) will cause an asymmetry of an optical spectral line-shape (similar to exciton optical absorption line-shape in quantum dots 22). Strong non-Markovity of the spontaneous decay (non-exponential decay with fast Rabi oscillations) originates from strong atom-vacuum-field coupling with the upper state of the system splitted into two “dressed” states. This will yield a two-component structure of optical spectra similar to that observed for excitonic and intersubband electronic transitions in semiconductor quantum microcavities 15, 16.

To conclude, we predict the existence of frequency ranges where, similar to semiconductor microcavities 16 and photonic band-gap materials 8, CNs qualitatively change the character of atom-electromagnetic-field interaction, yielding strong atom-field coupling — an important phenomenon necessary, e.g., for quantum information processing 23. The present Letter dealt with the simplest manifestation of this general phenomenon — vacuum-field Rabi oscillations in the atomic spontaneous decay dynamics near a single-wall carbon nanotube. Such a strictly non-exponential decay dynamics gives place to the exponential one if the atom moves away from the CN surface. Thus, the atom-field coupling strength and the character of the spontaneous decay dynamics, respectively, may be controlled by changing the distance between the atom and the CN surface by means of a proper preparation of atomically doped CNs. This opens routes for new challenging nanophotonics applications of atomically doped CN systems as various sources of coherent light emitted by dopant atoms. We emphasize that similar manifestations of strong atom-field coupling may occur in many other atom-electromagnetic-field interaction processes in the presence of CNs, such as, e.g., dipole-dipole interaction between atoms by means of a vacuum photon exchange 24, or cascade spontaneous transitions in three-level atomic systems 25.

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