Purcell factor engineering in plasmonic nanostructures for the enhanced generation of energy-time entangled states

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Abstract

We summarize our studies of the plasmonic enhancements of the spontaneous two-photon emission. In particular, we draw attention to the rigorous investigation of quantum framework for the multi-photon spontaneous decay rates in an environment with arbitrary structure of electromagnetic modes. We demonstrate that nanostructured environment can be used for tailoring the basic characteristics of the two-photon emission such as spectrum, lifetime, and entanglement. Moreover, careful engineering of the two-photon Purcell enhancement may provide any desired spectral response and may serve as an ultimate route for designing light sources with novel properties.

1. Introduction

Entanglement is one of the most fascinating phenomena in modern physics, which supplies the vast of challenges for fundamental as well as for applied research, promising numerous novel applications along with outstanding solutions for existing tasks. In terms of quantum communication applications the photonic entanglement is one of the most reliable approaches, demanding, however, efficient intergable sources of photon pairs. Multi-photon and in particular two-photon states are playing key roles in quantum cryptography and computing. Specific form of entanglement is dictated by system requirements, in particular, large distance communication channels, robust to environmental noises, may be realized by the energy-time entanglement at telecom wavelength.

Spontaneous two-photon emission (STPE) is the second-order quantum process where an excited electron decays to its ground state by simultaneous emission of a photon pair. The phenomenon allows any combination of photon energies with the only restriction on total energy conservation, resulting in very broad emission spectrum and, as the result, energy-time entanglement of the emitted pair.

STPE has recently been observed in optically pumped and electrically driven semiconductor structures [1]. The distinctive feature of semiconductors is a high concentration of charged carriers together with continuum of electronic states within conduction and valence bands. As a result, STPE from semiconductors becomes competitive with other nonlinear processes, such as spontaneous parametric down-conversion (SPDC), and it may become a new route for quantum devices [2].

2. Energy-time entanglement and multi-photon Purcell enhancement – General concept

Feynman's diagrams for the STPE process are presented in Fig. 1a. The intermediate state ($|n>$), assisting the transition, is virtual and, hence, has a very short lifetime, resulting in simultaneous emission of a photon pair. The emission spectrum is always symmetrical around half of the resonant transition frequency ($\Omega=2\omega_0$). The resulting entangled state is given by $\psi=\int d\omega|\omega_0-\omega>_A|\omega_0+\omega>_B$, where indexes A and B stand for the communication channels - Alice and Bob. The realisation of this sort of entanglement requires Franson interferometer [3] which introduces time bins for photon time arrivals.

Since the pioneering work of Purcell [4] it is well known that the strength of light-matter interaction can be significantly influenced by an engineering proper structure of electromagnetic modes, like in a cavity. A number of different systems, such as photonic crystals, metal nanostructures [5], and metamaterials [6] are used for different applications at the optical frequencies. Here we study *multi-photon spontaneous decay rates* in an environment with arbitrary structure of electromagnetic modes. The Purcell factor (P_n) for high-order processes with participation of n photons is of great importance, since being initially weak, these processes may be enhanced as roughly n^{th} power of general one-photon (P_1) enhancement. STPE from bulk semiconductors with deposited metallic nano-cavities on the top was recently demonstrated to have 10^3-fold overall enhancement, relatively to the sample without special patterning [7]. The proper theoretical treatment is very essential for the processes where photonic states are claimed to have some levels of non-classicality.

The difference between single- and two-photon Purcell effect is summarized in Fig. 1b. While the one-photon emission is enhanced only being resonant with a structure, the two-photon emitter, being naturally wideband, is enhanced twice – at the resonant frequency of a structure and at the complementary one, emphasizing the 'paired' nature of energy-time entangled emission process.

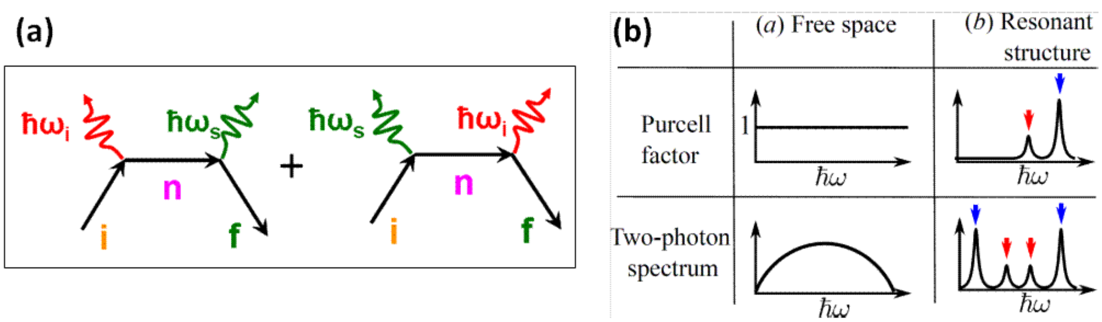

Fig. 1: (a) Feynman diagrams for STPE. (b) Difference between 1-photon and 2-photon Purcell factors. 1-photon emission is enhanced at the structure resonances. 2-photon emission is enhanced twice – at the resonant frequency of a structure and at the complementary. Different scenarios are marked by a set of red and blue arrows.

3. Multi-photon Purcell Enhancement by Nano-plasmonic Particle

The quantization of electromagnetic field in the presence of material bodies requires a significant care. Material dispersion and absorption rise difficulties for canonical Hamiltonian quantization, since additional 'material' degrees of freedom have to be included [8]. The approach for the field quantization, based on the field expansion over the classical electromagnetic modes of a structure is not applicable to lossy systems. Rigorously, the field operators are strictly related to the classical electromagnetic Green's functions and the dispersion and losses are included without violating canonical commutation rules. We consider a light emitting system such as an atom or a quantum dot, small compared with light wavelength and placed in the photonic environment characterized by the (dispersive and lossy) dielectric function (e.g. Fig. 2a). The resulting STPE rate is given by:

$$R_{STPE}=\int_0^{\omega_{if}} d\omega \mathrm{Tr}\left[\vec{P}_1(\omega)\vec{P}_1(\omega_{if}-\omega)\right]U(\omega), \quad (1)$$

where P_1 is tensor of one-photon Purcell factor and $U(\omega)$ spectral parameter, defining the infraction strength. Eq. 2 is the general formulae that may be straightforwardly applied to quite general physical system and is proportional to the convolution of one-photon Purcell factor, having the transparent physical meaning. To examine the effect of local field enhancement we have analyzed the case, when the emitter is placed in SiO_2 matrix in the proximity of subwavelength silver sphere. Fig. 2b presents the correspondence between the single-photon and two-photon emission for a source located at the different gaps (*d*) from plasmonic particle. Panel (a) shows the calculated energy dependence of the one-photon Purcell factor along the line between emitter and nanoparticle centre. The peak locations correspond to presents of plasmonic resonances, marked by $j=1,..$ (dipole and higher). Panel (b) presents normalized spectral dependence of STPE rate. These graphs possess mirror symmetry with respect to the central energy, which is a characteristic feature of STPE. Blue dashed curve corresponds to STPE spectrum for the infinite distance between the atom and the nanoparticle, i.e. the scenario of empty silica matrix. In this case STPE spectrum is rather smooth and depends only on the 'free space like' photon density of states. The highest emission probability corresponds to the equal energies of the emitted photons, so that the product of their densities of states is at maximum. The overall enhancement of STPE at d=10 nm is about 140-fold.

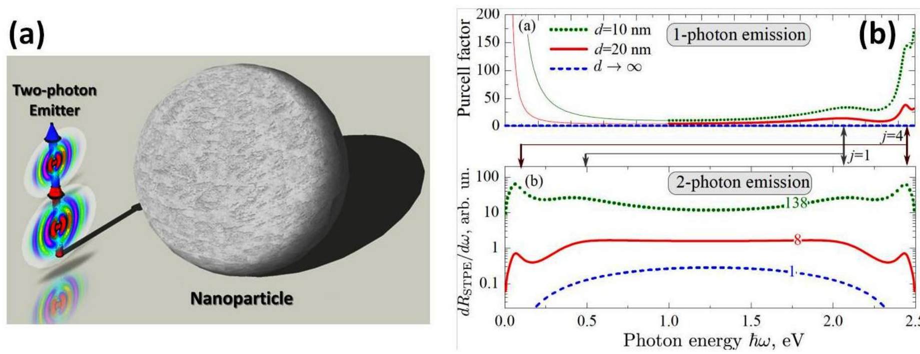

Fig. 2: (a) Geometry of the structure– two-photon dipole emitter in the proximity of nanosphere. (b) Difference between 1-photon and 2-photon Purcell factors. 1-photon emission from equally distributed in frequency sources is enhanced at the structure resonances, marked by red and blue arrows. 2-photon is wideband and enhanced twice – at the resonant frequency of a structure and at the complementary.

In summary, STPE is a very promising physical phenomenon that in near future may become the main route for realization of quantum information devices. With the help of nanoplasmonics and metamaterials the process was shown to be dramatically improved in terms of emission rates and quantum state manipulations, making this symbiosis to be extremely interesting for investigations.

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