Enhanced up-conversion of entangled photons and quantum interference under localized field in nanostructures

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We theoretically investigate up-conversion process of entangled two photons on a dimer molecule, which is coupled by a cavity or nanoscale metallic structure. Within one-dimensional input-output theory, the propagators of the photons are derived analytically and the up-conversion probability is calculated numerically. It is shown that the coupling with the nanostructure clearly enhances the process. We also find that the enhancement becomes further pronounced for some balanced system parameters such as the quantum correlation between photons, radiation decay rates and coupling between the nanostructure and molecule. The non-monotonic dependencies are reasonably explained in view of quantum interference between the coupled modes of the whole system. This result can provide a guideline for nonlinear optical reactions by weak light of a few photons.

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Nonlinear optical responses have appeared ubiquitously in modern technologies with photophysics, photophysics, classical and quantum communication [1]. As the application range is expanded, in addition to high-power optics using lasers, nonlinearity induced by a few photons is attracting increasingly much attention. Such a few-photon optics can lead to various new technologies, e.g., up-conversion for efficient solar cells [2], visible-to-telecom frequency conversion of single photon [3] and two-photon gateway for quantum communication [4]. On the other hand, photons with quantum correlation, i.e., entangled photons have been key issues especially in quantum information technology [5]. For such photons, the generation efficiency is growing large recently [6–9], and then they are expected to become a new type of luminous source. Therefore the study of nonlinear response by the correlated photons is not only interesting in itself, but also can contribute to further development in opto-science and technology.

In order to enhance the nonlinearity of a few photons, it is useful to utilize absorption saturation of discrete levels, e.g., in a molecule and quantum dot [10]. Besides, it is well-known that one can enhance interactions between photons and nanoscale materials, by introducing cavity in which a localized field is generated [11, 12]. Furthermore, for example, the usage of resonator can make spatial configuration of the order of the nano- or micrometers in electric field intensity [13, 14]. Another approach to strong localized field is to introduce localized surface plasmon resonance (LSPR) in metallic nanostructures such as nanochips and nanorods [15, 16]. Nearby the structures, localized fields of extraordinary high intensity are generated, and they have steep gradient of nanometer scale in intensity. Such localized fields can break long-wavelength approximation, and are expected to open a new type of optics including dipole-forbidden excitation. Actually, the existence of such forbidden excitations is suggested theoretically [17, 18]. Then, by embedding discrete levels in cavity or nearby nano-scale metallic structure, one can prepare a challenging stage for the studies of nonlinear few-photon optics.

In this work, we focus on two-photon up-conversion process on a dimer molecule (or double quantum dot) coupled to localized field in nanostructures. Here we assume that the two photons have correlated in space, and are described by bi-variable gaussian pulse. Additionally, the localized field in the nanostructure is assumed to have spatial inhomogeneity, and then both dipole-allowed and -forbidden states through the field can be accessed. We use a fairly intuitive model for the system in order to extract the essential effect of quantum mechanical correlations and coherence. We analytically derive the propagator for the process, and numerically calculate the wave function of the converted output photon. As a result, it is shown that the coupling to the cavity strongly enhances the up-conversion process. We also find that the spatial correlation militates for the process. There exists the regime where the photon-pair rarely induces the up-conversion without entanglement. Furthermore, there is some suitable set of system parameters. The origin of the non-monotonic dependencies can be interpreted in view of quantum interference between the coupled modes of the open cavity-molecule system. It holds the line against the rapid intuition that nonlinearities of photons favor the stronger cavity-molecule couplings than the cavity decay rates. In this sense, our result can provide a guideline for nonlinear optical reactions, and contributes to future applications of weak light physics.

The system under consideration is shown in Fig. 1(a). The molecule has four levels; the states \(|a\rangle\) and \(|e\rangle\) are dipole-allowed from the ground state \(|g\rangle\), and the state \(|f\rangle\) is dipole-forbidden owing to the parity of the wavefunction. The Hamiltonian of the whole system is given by \(H = H_c + H_m + H_p + H_{cp} + H_{mp} + H_{cm}\). Here we model the cavity mode (or polarization mode by LSPR) by simple boson, i.e., \(H_c = \hbar \omega_c p^\dagger p\) with \(p\) being the anni-
hilation operator. As for the molecule state, the Hamiltonian is $H_m = \hbar \omega_a \sigma_{aa} + \hbar \omega_f \sigma_{ff} + \hbar \omega_c \sigma_{ee}$, in which the operator $\sigma_{mn} = \langle m | n \rangle$ with $\{m,n\} = \{g,a,f,e\}$ and the resonant energies are measured from the ground state $|g\rangle$. The resonant energies of $|a\rangle$ and $|f\rangle$ differ from each other by $2\Delta$. In this work, we employ the one-dimensional mode and then the photon field can be described as $H_p = \int dk \hbar \sigma(\tilde{a}_k^\dagger \tilde{a}_k + \tilde{b}_k^\dagger \tilde{b}_k)$. Here the operator $\sigma(\tilde{a}_k)$ annihilates a photon at position $r$, and $\tilde{a}_k = \sqrt{1/2\pi} \int dr \tilde{a}_r e^{-ikr}$. Although the model seems oversimplified, most of the systems can be expressed by superposing the one-dimensional cases, and then this simplification can reasonably extract an aspect of the problem.

Input two photons interact with the coupled system at the origin ($r = 0$). The interactions are characterized by the radiation decay rates as shown in Fig. 1(b). Then, the cavity-photon coupling is written within rotating-wave approximation as

$$H_{cp} = i\hbar \sqrt{\Gamma_0} (p^l a_{r=0} - a^l_{r=0} p),$$

in which $\Gamma_0$ is the decay rate of the cavity mode. In a similar fashion, the radiations of the photons from the molecule are

$$H_{mp} = i\hbar \sqrt{\gamma_{\bar{f}}a_0^\dagger \sigma_{ag} a_{r=0} + i\hbar \sqrt{\gamma_{ag} \sigma_{ef} a_{r=0} + i\hbar \sqrt{\gamma_{ag} \sigma_{ef} a_{r=0} + i\hbar \sqrt{\gamma_{gf} \sigma_{cg} a_{r=0} + \text{h.c.}},}}$$

with the constants $\{\gamma_{\bar{a}a}, \gamma_{ag}, \gamma_{ag}, \gamma_{gf}, \Gamma_3\}$. The photon with the operator $b_{r=0}$ corresponds to the up-converted one. In general, the decay rates of the forbidden transitions are much smaller than the ones of the allowed transitions ($\gamma_{\bar{a}a} \gg \gamma_{ag}$). The last part of the Hamiltonian is for the cavity-molecule couplings. In the system under consideration, the localized field with spatial gradient can produce similar intensities of absorptions in both the dipole-allowed and forbidden transitions [10]. Then, in the Hamiltonian

$$H_{cm} = \hbar g_{1a} \sigma_{ag} p + \hbar g_{2f} \sigma_{ef} p + \hbar g_{1f} \sigma_{f} p + \hbar g_{2a} \sigma_{a} p + \hbar g_{2f} \sigma_{ef} p + \text{h.c.},$$

the decay rates $\{g_{1a}, g_{1f}, g_{2a}, g_{2f}\}$ is considered to have the magnitudes of the same order.

We analyze the whole process by using one-dimensional input-output theory. As an initial state, we prepare the input state vector which can be written as

$$|\psi_{in}\rangle = \int dr_1 dr_2 \frac{f(r_1, r_2) a_{r_1}^\dagger a_{r_2}^\dagger |V\rangle}{\sqrt{2}}.$$
In calculating the propagators as a practical matter, we employ the method developed in Ref. [20], in which a coherent state $|\phi\rangle$ is introduced. For that state, one can write down the relations $a_\epsilon |\phi\rangle = \sum_{j=1,2} \mu_j \delta(r - r_j') |\phi\rangle$ and $b_\epsilon |\phi\rangle = 0$, where $\mu_j$ are perturbation coefficients. From the Heisenberg equations for the operators of the photons, cavity, and molecule, we obtain the simultaneous equations of motion for the expectation values of them. Within the first order with respect to $\mu_1$ and $\mu_1 \mu_2$, it is found that, e.g.,

$$G_1(r, r_1, r_2; \tau) \equiv \frac{1}{\sqrt{2}} \langle V | b_\epsilon(\tau) a^\dagger_{r_1} a_{r_2} | V \rangle \propto \langle \sigma_{ge} (\tau - r_j/\epsilon) \rangle^{\mu_1 \mu_2}, \quad (8)$$

in which $\langle \sigma_{ge} \rangle^{\mu_1 \mu_2}$ means the perturbation component proportional to $\mu_1 \mu_2$ in $\langle \sigma_{ge} \rangle$. Then we can analytically solve the equation and obtain the propagators. The up-conversion probability here we focus on is given by $P = \int \{ r | h(r) |^2 \}$. Hereafter we assume that the frequency of the cavity mode is set to be $\omega_c = (\omega_a + \omega_f) / 2 = \omega_c / 2$. Then, the up-converted photon has twice the frequency of the that of the input photon $\omega_0$. Because the decay rate of the cavity mode is large compared to the other rates, we use $\Gamma_3/\Gamma_0 = 0.2$, $\gamma_{1a, 2a}/\Gamma_0 = 0.01$, $\gamma_{f, 2f}/\Gamma_0 = 0.001$. In addition, for the sake of simplicity, we assume that all the cavity-molecule coupling constants are equal, i.e., $g_{1a} = g_{1f} = g_{2a} = g_{2f} = g$. When $\Gamma_0 \gg \Delta$, the cavity modes cannot make out one of the excited states $|a\rangle$ and $|f\rangle$ energetically.

Figure 2 shows the correlation parameter dependence of the up-conversion probability for $g/\Gamma_0 = 0.2$. Here we set the pulse length to be $d \Gamma_0 / c = 7$, which corresponds to $d = 138 \mu$m for $\Gamma_0 = 20$ meV. The different lines correspond to the ones for different detunings $2 \Delta$ between the states $|a\rangle$ and $|f\rangle$. Because the probability for $g = 0$ is at most $P \lesssim 0.01$ in the same condition, it is apparent that the couplings to the cavity enhance the up-conversion. We calculate $\rho$-dependence of the up-conversion probability, and find that it exceeds $P = 0.8$ when the input two-photon is correlated state ($\rho = 0.9$). On the other hand, as the detuning increases, the probability decreases. When the detuning $2 \Delta$ becomes larger than the broadening due to the cavity-radiation ($\sim \Gamma / 2$), the probability is negligibly small for non-correlated photon-pair ($\rho = 0$). However, as the correlation becomes strong, it exceeds $P = 0.4$ even when $\Delta / \Gamma_0 = 0.3$. Just near $\rho = 1$, the up-conversion probability turns to decrease. This is because, if the two photons interact with the molecule at the same moment, the sequential process of the up-conversion is inhibited [21].

Subsequently, we investigate the dependence on the cavity-molecule coupling. In Fig. 3(a), we plot the up-conversion probability as the function of the correlation parameter $\rho$ and coupling $g$ for $\Delta = 0$. It should be noted that the optimum parameter regime exists, where $g / \Gamma_0 = 0.15 \sim 0.2$. This can be explained in view of quantum mechanical interference between the coupled modes in the open system. When we focus on the first excitation by one of the photons, the system can be seen as V-type three level coupled to the cavity. Then, three eigenmodes can be considered; one dark mode and two bright modes. Neglecting the decay rates in the molecule, the effective bright modes in the open system can be simplified as

$$E_{\pm} = \frac{\hbar}{2} (\omega_a + \omega_c - i \Gamma_0 / 2 \pm \sqrt{(\omega_a - \omega_c + i \Gamma_0 / 2)^2 + 4g^2} ) \quad (9)$$

for $\Delta = 0$. When $\Gamma_0 = 0$, the level splitting between the two bright modes becomes $2 \sqrt{2} g$ at the anti-crossing point. Then the splitting of three eigenmodes is disappeared in the presence of the broadening by the cavity $\Gamma_0 / 2 \geq 2 \sqrt{2} g$, i.e., they oscillate effectively in the same frequency. This is the same physics as the damped Rabi oscillation in open quantum mechanics [22]. Thus the two bright modes, both the cavity mode and molecule level are included, can interfere constructively and destructively. When the destructive interference occurs in the cavity, it is possible to make only the molecule oscillate, i.e., the ground state of the cavity becomes transparent for the incident photon. In addition, the imaginary part of the eigenmodes, which corresponds to effective decay rates, take maximum for $\Gamma_0 / 2 \leq 2 \sqrt{2} g$. Almost the same circumstance exists for the second excitation to the state $|e\rangle$; the lead difference comes from the radiation of the up-converted photon by $\Gamma_3$. Therefore, the up-conversion is considered to be enhanced when $\Gamma_0 / 2 \sim 2 \sqrt{2} g$, which is consistent with the result in Fig. 3(a).
In conclusion, we have analyzed up-conversion process of entangled two photons on a dimer molecule, which is coupled by a cavity or nanoscale metallic structure. As a result, we found that the coupling to the cavity enhances nonlinear up-conversion process, and the usage of spatially entangled pair makes the up-conversion further facile. We also elucidated suitable conditions and their origins for the energy of input photons to be transferred efficiently. This phenomenon is caused by quantum interference between two eigenmodes, which include cavity mode and molecule level. Then, the results make a clear departure from the traditional idea that the stronger cavity-molecule couplings are favorable for multi-photon processes.

Having discussed in fairly simple model so far, it is quite important for practical applications to take into account individual circumstances of particular systems, e.g., dephasing in the whole system and vibration levels in the molecule and so on. Actually few-photon responses have recently appeared not only in quantum physics but also in photochemistry, e.g., two-photon photopolymerization in SU-8 molecules embedded on Au nanostructure [23]. Thus presenting an intuitive guideline for efficient nonlinear responses can contribute also to further promoting more elaborate studies in various fields, and development in future photo-science.

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FIG. 3: (a) Up-conversion probability is plotted against the coupling constant $g$ and the correlation parameter $\rho$. Here we set the detuning and pulse length to $\Delta = 0$ and $d\Gamma_0/c = 7$. It is apparent that the large correlation ($\rho \sim 1$) is preferred for the up-conversion. (b) The plot against $g$ and $d$ for $\Delta = 0$ and $\rho = 0.9$, which shows the long pulse has advantage for the conversion. In both the figures, one can see that the optimal regime for $g$ exists between 0.15 and 0.2.
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