Full-quantum analysis of energy transparency on an antenna-molecule coupled system

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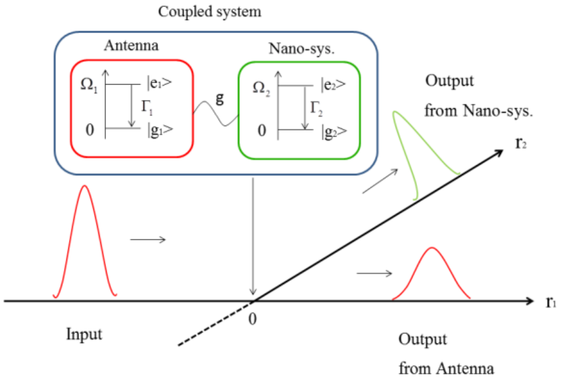

Fig. 1 Physical situation investigated in this study.

The interactions between light and nanomaterials have been intensively studied in various research fields. Nanoscale objects such as single molecules and quantum dots are difficult to react directly with light because of their cross sections which are considerably small compared with the light wavelength. Optical antennas which enhance the light-matter interactions allow us to efficiently transfer the photon energy into nanomaterials. In our previous research, we have demonstrated that controlling the coupling between the molecule and antenna systems and their interference yields the peculiar energy transparency effect in the weak excitation process [1], where although only the antenna system is illuminated, it is not excited; only the molecule system is excited. The energy transparency effect, therefore, significantly reduces energy dissipation in the antenna system. In the previous discussion, however, the incident light is treated classically, where a monochromatic coherent light is assumed. If the energy transparency effect can occur for a weak incoherent light such as sun light, the possibilities of various applications can be expected. To this end, in this contribution, we investigate theoretically the possibility of energy transparency at the single-photon level on the basis of the full-quantum analysis, where lights are treated quantum-mechanically as multimode photon fields.

As illustrated in Fig.1, the system considered in this study is composed of two kinds of one-dimensional photon fields and an antenna-molecule coupled system. We assume that the antenna and molecule systems which are modeled as a two-level system interact with different photon fields. The antenna system is coupled strongly with a one photon field, while the molecule system is coupled weakly with the other photon field. A single-photon pulse is input from the input port (the $r_1 < 0$ region) onto the antenna system located at $r_1 = 0$. The input photon interacts directly only with the antenna system. After interacting with the coupled system, a transmitted photon is output from the molecule system with a certain probability. A high transmission probability of a photon indicates the energy transparency. The level-structure of the antenna and molecule systems is shown in Fig.1. The two levels of the antenna (molecule) system are denoted by $|g_1>$ and $|e_1>$ ($|g_2>$ and $|e_2>$), and the energy of $|e_{1,2}>$ measured from $|g_{1,2}>$ is $\Omega_{1,2}$. $\Gamma_{1,2}$ are the radiative decay rates of the antenna and molecule systems, and the coupling strength between the two systems is denoted by g.

Employing the natural units, the Hamiltonian for the overall system is given by

$$\mathcal{H} = \Omega_1 \sigma_1^\dagger \sigma_1 + \Omega_2 \sigma_2^\dagger \sigma_2 + g(\sigma_1^\dagger \sigma_2 + \sigma_2^\dagger \sigma_1) + \int dk\, k(a_k^\dagger a_k + b_k^\dagger b_k) + \frac{1}{\sqrt{2\pi}} \int dk\, (i\sqrt{\Gamma_1}\sigma_1^\dagger a_k + i\sqrt{\Gamma_2}\sigma_2^\dagger b_k - i\sqrt{\Gamma_1} a_k^\dagger \sigma_1 - i\sqrt{\Gamma_2} b_k^\dagger \sigma_2). \tag{1}$$

where $\sigma_{1,2} = |g_{1,2}><e_{1,2}|$ are the Pauli lowering operators and a_k (b_k) denotes the annihilation operators of the photon field coupled with the antenna (molecule) system. The state vectors of the input and output single-photons can be written as follows:

$$|\psi_{\rm in}\rangle = \int dr' f(r') \tilde{a}^\dagger_{r'} |0\rangle\,, \tag{2}$$

$$\begin{aligned} |\psi_{\rm out}\rangle &= {\rm e}^{-i\mathcal{H}t}|\psi_{\rm in}\rangle\,, &(3)\\ &= \int dr g_1(r;t)\tilde{a}^\dagger_r|0\rangle + \int dr g_2(r;t)\tilde{b}^\dagger_r|0\rangle\,, &(4)\end{aligned}$$

where $f(r)$ is the wavefunction of the input photon pulse, $g_1(r;t)$ and $g_2(r;t)$ are the output photon wavefunctions from the antenna and molecule systems, respectively. The final moment t is a sufficiently large time when the coupled system is completely de-excited. In these expressions, the real-space representation of the photon annihilation operator a_r (b_r) is defined as the Fourier transformation of a_k (b_k). Using the method developed in Ref.2, we can derive the analytical expressions of $g_1(r;t)$ and $g_2(r;t)$ as follows:

$$\begin{aligned} g_1(r;t) = f(r-t) - \frac{\Gamma_1}{2\Lambda}\int_{r-t}^{0} dr'\, f(r')\Bigg[&\left(\frac{\Gamma_1-\Gamma_2}{4}+\Lambda-i\frac{\Omega_2-\Omega_1}{2}\right){\rm e}^{-\left\{i\frac{\Omega_1+\Omega_2}{2}+\frac{\Gamma_1+\Gamma_2}{4}+\Lambda\right\}(t-r+r')}\\ &-\left(\frac{\Gamma_1-\Gamma_2}{4}-\Lambda-i\frac{\Omega_2-\Omega_1}{2}\right){\rm e}^{-\left\{i\frac{\Omega_1+\Omega_2}{2}+\frac{\Gamma_1+\Gamma_2}{4}-\Lambda\right\}(t-r+r')}\Bigg]\,, \end{aligned} \tag{5}$$

$$g_2(r;t) = \frac{ig\sqrt{\Gamma_1\Gamma_2}}{2\Lambda}\int_{r-t}^{0} dr'\, f(r')\left[{\rm e}^{-\left\{i\frac{\Omega_1+\Omega_2}{2}+\frac{\Gamma_1+\Gamma_2}{4}-\Lambda\right\}(t-r+r')} - {\rm e}^{-\left\{i\frac{\Omega_1+\Omega_2}{2}+\frac{\Gamma_1+\Gamma_2}{4}+\Lambda\right\}(t-r+r')}\right]\,, \tag{6}$$

Figure 2 shows the profiles of the output photons, where for simplicity we assume resonance between the antenna and molecule, i.e., $\Omega_1 = \Omega_2$. It is observed that the transmission of the photon from the molecule can be drastically enhanced under a certain condition. We explain the details of this condition and the mechanism on the day of the forum.

In this study, we have investigated the possibility of the energy transparency for a single-photon pulse. As a result, it is revealed that the energy transparency effect can occur for a single-photon pulse under a certain condition, which indicates a positive result to the energy transparency for a weak incoherent light such as sun light, and opens up the possibility of efficient photoexcitation of nanomaterials by sun light.

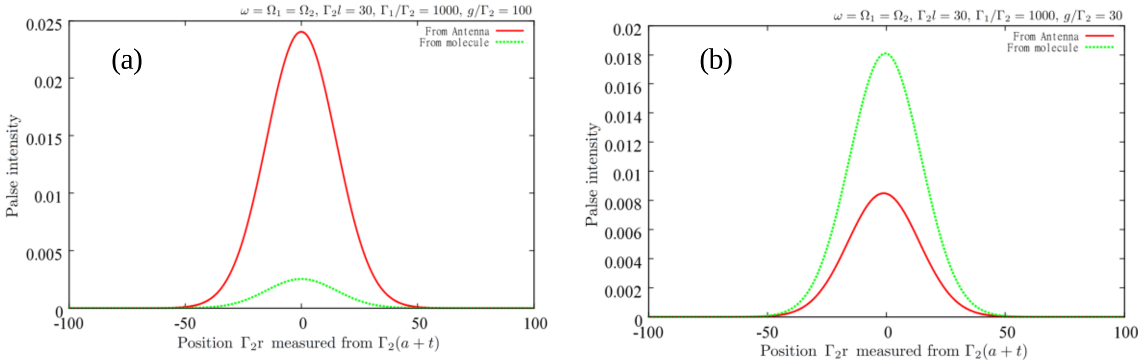

Fig. 2 Pulse profile of output photon. The central energy of the incident pulse is set to be Ω_1(=Ω_2), where l is the input pulse length. The solid and dotted lines indicate the output photons from the antenna and molecule systems, respectively. (a) The photon from the molecule is quite small. (b) The photon from the molecule is strongly enhanced under a certain condition.

References

1. H. Ishihara, A. Nobuhiro, M. Nakatani and Y. Mizumoto: J. Photochem. Photobiol. A (2011) in press.
2. K. Koshino and M. Nakatani: Phys. Rev. A **79**, 055803 (2009).