Entangled-photon generation via biexcitons in nano-structures

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We have constructed quantum electrodynamics theory for excitons whose center-of-mass motion is confined in arbitrary-structured dielectrics with considering their nonlocal optical susceptibility. We systematically investigate entangled-photon generation via biexcitons in nano-to-macro scale materials by this theory. It is revealed that nano-structured materials provide scattering spectra explicitly reflecting quantum states of confined exciton-polaritons and are promising sources of entangled photons from the viewpoints of both efficiency and signal-to-noise ratio.

1 Introduction Entangled-photon pairs, which play an important role in quantum information technology, are usually generated by parametric-down conversion (PDC). On the other hand, it is reported that hyper-parametric scattering (HPS) in CuCl also generates ultraviolet entangled photons [1]. In the HPS process, resonantly created biexcitons with no angular momentum collapse with the generation of polarization-correlated photon pairs conserving the angular momentum. This scheme shows much higher efficiency than the usual method by PDC because of the giant two-photon absorption in CuCl. Although the bulk crystal was used in Ref. [1], it is worth investigating the HPS process in nano-structures such as quantum dots [2, 3], because manipulating the excitonic confinement has a potential to control characteristics of the entangled-photon generation. The purpose of our study is the investigation of HPS in nano-to-macro scale materials, where center-of-mass motions of excitons and biexcitons are confined.

2 Theory The entangled-photon generation by HPS was predicted by Savasta et al. [4, 5]. They calculated the scattering intensity and two-photon coincidence obtained from CuCl bulk film, and also showed their thickness dependence. Their calculation was based on the quantum electrodynamics (QED) theory in dispersive and absorptive dielectrics [6]. However, their calculation did not include the nonlocal optical susceptibility caused by the center-of-mass motion of excitons, which must be considered to discuss the optical processes in nano-structures where excitons are weakly confined. Such a nonlocality has been studied in the semiclassical framework [7]. We have extended the microscopic nonlocal theory to be able to consider quantum characteristics of the electromagnetic field by using the QED theory applicable to arbitrary-structured dielectrics [8], and we also consider absorption relaxation process of excitons. As far as the linear optical process is concerned, the nonlocal equations of motion reduce to the linear simultaneous equations as in the semiclassical nonlocal theory [7]. The QED is described by the commutation relation of electric field $\hat{\boldsymbol{E}}_0^{\pm}(\boldsymbol{r},\omega)$ before the interaction with excitons:

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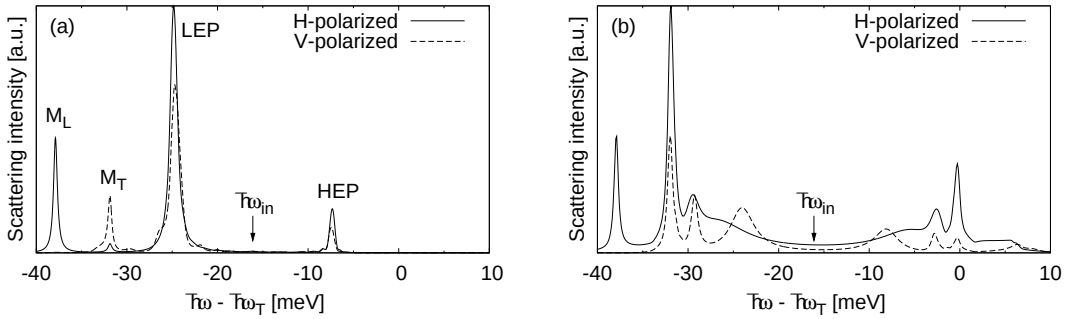

Fig. 1 Forward HPS spectra of a CuCl film with thickness (a) $d = 7\ \mu$m and (b) $d = 200$ nm. Incident frequency is $\hbar\omega_{\rm in} \sim \hbar\omega_{\rm T} - 16.1$ meV, and scattering angle is $\theta = 60^\circ$

$[\hat{\boldsymbol{E}}_0^+(\boldsymbol{r},\omega), \hat{\boldsymbol{E}}_0^-(\boldsymbol{r}',\omega')] = \delta(\omega-\omega')(\mu_0\hbar\omega^2/\pi)\mathrm{Im}[\mathsf{G}(\boldsymbol{r},\boldsymbol{r}',\omega)]$, where G is a Green's tensor satisfying $\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathsf{G}(\boldsymbol{r},\boldsymbol{r}',\omega) - (\omega^2/c^2)\varepsilon_{\rm bg}(\boldsymbol{r},\omega)\mathsf{G}(\boldsymbol{r},\boldsymbol{r}',\omega) = \mathsf{I}\delta(\boldsymbol{r}-\boldsymbol{r}')$ and background dielectric structure is described by the dielectric function $\varepsilon_{\rm bg}(\boldsymbol{r},\omega)$. This scheme has a good correspondence with the original QED theory [8].

In a previous study of HPS by Savasta et al., the biexciton states were described by correlation functions between excitons that were calculated from first principles in another study [9]. Instead of this kind of numerical treatment, we use the simplified model of biexcitons including fitting parameters for the purpose of the quantitative comparison with the experimental results. Using this biexciton model, we describe the HPS process with the exciton-exciton interaction as a perturbation. The essential progress made in our HPS calculation method from the previous one is as follows: 1) We consider arbitrary-shaped nano-to-macro scale materials, where excitons and biexcitons are weakly confined, and 2) calculate the polarization correlation of generated pairs expected from the angular momentum of biexcitons, and 3) discuss the ratio of number of correlated pairs (signal) to uncorrelated ones (noise), which come from coincidentally collapsed different biexcitons.

3 HPS in nano-structures First, we investigate the HPS intensity spectra in vacuum obtained from a CuCl film with thickness d. We assume that the transverse exciton energy at $k = 0$ is $\hbar\omega_{\rm T} = 3.2022$ eV, binding energy of biexcitons is $\Delta_{\rm bx} = 32.2$ meV, and absorption width of excitons is $\gamma = 0.5$ meV. The wave functions of center-of-mass motion of excitons and biexcitons in the confined direction are in the form of $g_m(z) = (2/d)^{1/2}\sin(q_m z)$, $q_m = m\pi/d$, $m = 1,\ 2,\ \cdots$, translational mass of excitons is $m_{\rm ex} = 2.3m_0$, and that of biexcitons is $m_{\rm bx} = 2.3m_{\rm ex}$, where m_0 is the free electron mass. Incident light is perpendicular to the surface, and scattering field goes forward at angle $\theta = 60^\circ$, where θ gives the in-plane wavenumber $k_\parallel = (\omega_{\rm T}/c)\sin\theta$ and is understood as the approximate scattering angle in vacuum. We define the horizontal (H) and vertical (V) direction of the polarization with respect to the scattering plane.

Fig. 1 shows the scattering spectra for each linearly polarized field at thickness (a) $d = 7\ \mu$m and (b) $d = 200$ nm. The incident frequency is $\hbar\omega_{\rm in} \sim \hbar\omega_{\rm T} - 16.1$ meV for the degenerate two-photon excitation of biexcitons. In Fig. 1(a), four peaks called $\mathrm{M_L}$, $\mathrm{M_T}$, LEP and HEP (lower and higher energy polariton) appear as the experimental HPS spectrum [1]. On the other hand, the LEP and HEP peaks disappear in Fig. 1(b), because the phase matching condition is relaxed with decreasing thickness. Instead, new multiple peaks arise in the LEP-HEP frequency region due to the discretization of exciton energy levels.

In order to analyze these new peaks, we calculate the quantum states of the exciton-polaritons confined in the nano-film. The result for V-polarized polaritons is shown in Fig. 2(b). Dotted lines represent the dispersion relations of longitudinal excitons, upper and lower branch polaritons (UBP and LBP) in bulk CuCl. The discretized eigen-energies of confined-polaritons are plotted with vertical bars, which indicate the sum

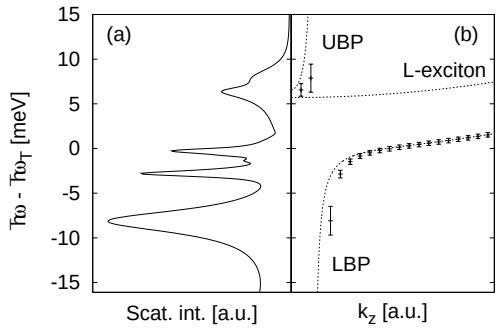

Fig. 2 (a) Scattering spectrum and (b) dispersion relation for V-polarized field: $d = 200$ nm, $\theta = 60^\circ$. Vertical bars in (b) indicate the sum of radiative and absorption widths of confined-polariton states.

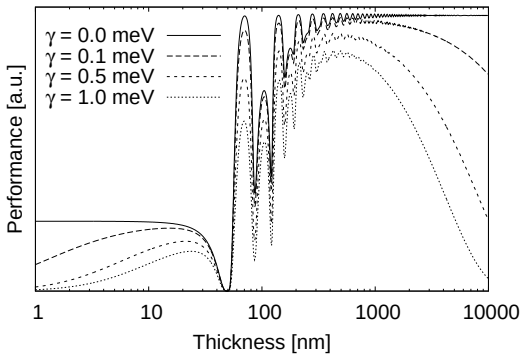

Fig. 3 Thickness dependence of the performance for each absorption width γ with neglecting the surface effect: $\theta = 0^\circ$, $\omega = \omega_{\rm in}$.

of radiative and absorption widths of the polariton states. This confined-polariton mode structure has been discussed in the semiclassical framework [7, 10], and also it has been observed experimentally [11]. Fig. 2(a) is the enlarged view of higher energy side of the spectrum for V-polarized field in Fig. 1(b). Comparing these two figures, we find that the discretized energy levels and widths of the confined-polaritons agree with the positions and widths of the new peaks in the scattering spectrum. This can be understood as follows: The biexcitons relax to the confined-polariton states by emitting photons with frequency of the lower energy side peaks satisfying the energy conservation, and remaining polaritons also go out to the vacuum with their radiative width. In this way, the HPS spectra of nano-structures reflect the quantum states of the confined exciton-polaritons.

4 Entangled-photon generation by nano-structures Next, we calculate the coincidence intensity both for correlated pairs (signal S) and uncorrelated ones (noise N). Although the thickness dependence of S has already been calculated in Ref. [5], we must evaluate the S/N ratio to estimate the performance of the entangled-photon generation by HPS. When we write the incident power as I, S is proportional to I^2, but N is to I^4, because an uncorrelated pair is involved with two biexcitons, so the signal quality becomes worse with increasing I. Therefore, we evaluate the performance of the entangled-photon generation as $S(I_0)$, where I_0 satisfies $S(I_0) = N(I_0)$.

Fig. 3 shows the thickness dependence of the performance for each absorption width γ. In order to discuss the essence of the influence of the thickness and γ, the surface effect is neglected, scattering angle is set as $\theta = 0^\circ$, and scattering frequency as $\omega = \omega_{\rm in}$. The multi-peak structure shown in nano-scale region is due to the excitonic confinement, i.e., good performance is obtained only at particular thicknesses, where biexcitons are effectively created. On the other hand, we can find that the performance is not enhanced with increasing thickness, but rather becomes worse due to the absorption relaxation. And it turns out that the maximum performance is obtained at thickness of a few hundred nanometers, where the absorption influence is not so critical. However, since signal intensity by a nano-film is very weak compared to that by bulk materials, we must enhance the generation efficiency by some techniques as proposed as follows.

Instead of the degenerate two-photon excitation of biexcitons, induced excitation by two incident beams with different frequencies can greatly enhance the signal intensity even for nano-structures. This is because of the real excitation of excitons in the latter regime, in contrast to the virtual excitation in the former. Below we show one of the good conditions for entangled-photon generation by HPS.

Fig. 4 shows the signal and performance spectra for both excitation regimes. The parameters are the same as those of Fig. 1 except the incident frequencies in the induced excitation regime taken at $d = 412$ nm. One beam resonantly creates exciton-polaritons of center-of-mass motion $m = 1$, and

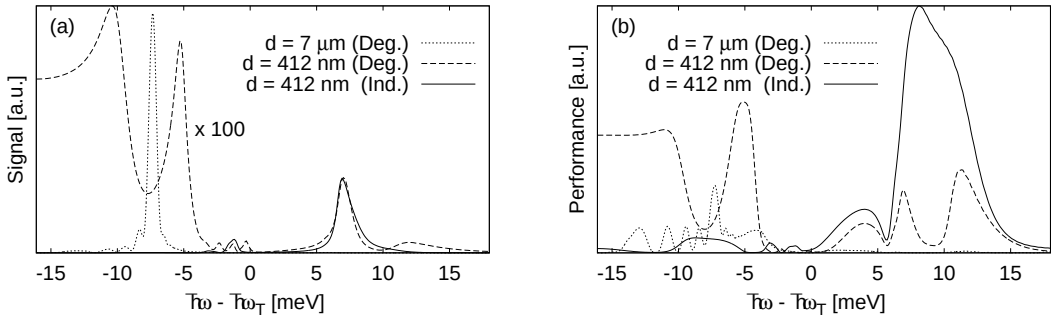

Fig. 4 (a) Signal and (b) performance spectra for degenerate excitation at $d = 7\ \mu\mathrm{m}$ (dotted line) and $d = 412\ \mathrm{nm}$ (dashed), and for induced excitation at $d = 412\ \mathrm{nm}$ (solid).

the frequency of the other beam agrees with the transition energy from its polariton state to $m = 6$ biexciton state. In the case of the degenerate excitation regime, we can find that the nano-film provides higher performance but much weaker signal than the bulk film. On the other hand, as expected, the induced excitation at the nano-film enhances the signal intensity at $\hbar\omega \sim \hbar\omega_{\mathrm{T}} + 7\ \mathrm{meV}$ to the same order as that for bulk. Although the performance spectrum is modified by changing the incident light condition, higher performance is obtained at $\hbar\omega \sim \hbar\omega_{\mathrm{T}} + 8\ \mathrm{meV}$. In this way, it turns out that high performance and efficiency can be realized simultaneously by the induced excitation at a nano-film with a particular thickness.

The reason of this high performance is as follows. Compared to the spectral shape of signal and noise at $\hbar\omega \sim \hbar\omega_{\mathrm{T}} + 7\ \mathrm{meV}$ peak, we find that the signal has much broader width than the noise at the higher energy side, in contrast to almost the same widths of HEP and LEP peaks for the degenerate excitation at a bulk film. Therefore, very high performance is obtained in that frequency region. And we consider that the difference of the peak widths is due to the interference of the scattering fields induced by the multiple discretized exciton-polariton modes, which exist on the UBP dispersion curve as shown in Fig. 2(b).

5 Summary We have constructed a QED theory for excitons with their microscopic nonlocality, and developed a HPS calculation method applicable to nano-to-macro scale materials. It is revealed that quantum states of confined exciton-polaritons are reflected in HPS spectra by nano-structures, and high performance and high efficiency can be simultaneously realized by the induced excitation of biexcitons confined in a nano-film with a particular thickness. This result indicates that nano-structured materials are promising sources of entangled-photon pairs.

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