Evaluation of Two-Photon Nonlinearity by a Semiclassical Method

Kazuki Koshino* and Hajime Ishihara

CREST, Japan Science and Technology Agency, 4-1-8 Honcho, Kawaguchi, Saitama 332-0012, Japan
Department of Materials Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan

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In order to discuss the two-photon nonlinearity theoretically, both photons and nonlinear materials should be treated quantum mechanically, which usually is a heavy theoretical task. Contrarily, nonlinear optics for classical light has been developed well and a detailed analysis is possible for realistic complex nonlinear systems. Here we show that the two-photon nonlinearity can be evaluated from the linear and third-order nonlinear output fields against a classical input pulse, which contains $2^{-1/2}$ photons on average.

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When two photons are input simultaneously into a nonlinear optical system, the two photons may interact with each other and may show nonlinear behavior in principle. Such two-photon nonlinearity was supposed to be negligibly small, because the photon field per one photon is too weak to induce significant nonlinear effects. However, it was experimentally shown that the effective nonlinearity of a system may be enhanced drastically by using optical cavities, which opened the possibility of obtaining a large nonlinear response even by a weak input field, such as two photons [1,2]. Interest in two-photon nonlinearity is growing rapidly, due to its applicability to quantum logic gates [3] and also to advances in the photon manipulation technique [4,5].

The photon number states belong to nonclassical states of photons. In order to theoretically discuss the two-photon nonlinearity, it is therefore indispensable to treat photons, as well as nonlinear materials, in a quantum-mechanical fashion. However, it is a heavy theoretical task to treat both photons and nonlinear materials quantum mechanically; such analyses are carried out up to date only in cases where the nonlinear systems are simple ones, such as a bare two-level atom [6] and a two-level atom placed in a cavity [7]. In the field of conventional nonlinear optics, in contrast, the nonlinear optical responses have been theoretically investigated in various complex systems, but the analyses are based on the semiclassical formalism, where light fields are always treated classically [8]. Therefore, it seems that vast wisdom accumulated in the conventional nonlinear optics cannot be applied in consideration of the two-photon nonlinearity.

The objective of the present study is to clarify the link between the two-photon nonlinearity and the conventional nonlinear optics. More concretely, we are showing that a parameter representing the degree of two-photon nonlinearity can be evaluated from the results of semiclassical calculations, which are usually much simpler tasks than fully quantum-mechanical ones. We propose a formula which connects the semiclassical results to the two-photon nonlinearity. The validity of the formula is demonstrated taking an atom-cavity system as an example of nonlinear optical systems.

The situation considered in this study is illustrated in Fig. 1. Two input photons propagate in one-dimensional space into the positive direction and interact with the nonlinear system located around the origin. We assume that photons are not lost by the interaction and, therefore, two photons always appear in the output port. Focusing on the case where the two photons have identical wave function $f(r)$ in the input state, the input state vector is given by

$$|\Psi_{\mathrm{in}}\rangle = \int dr_1 dr_2 f(r_1) f(r_2) a_{r_1}^\dagger a_{r_2}^\dagger |0\rangle, \tag{1}$$

where $a_r^\dagger$ is the photon creation operator at r. $f(r)$ is localized in the negative region and is normalized as $\int dr |f(r)|^2 = 2^{-1/2}$. Similarly, we denote the output state vector by

$$|\Psi_{\mathrm{out}}\rangle = \int dr_1 dr_2 \tilde{g}(r_1, r_2) a_{r_1}^\dagger a_{r_2}^\dagger |0\rangle, \tag{2}$$

where $\tilde{g}(r_1, r_2)$ is localized in the positive region and satisfies $\tilde{g}(r_1, r_2) = \tilde{g}(r_2, r_1)$. It should be noted that the output wave function $\tilde{g}(r_1, r_2)$ is, in general, no more factorizable; i.e., the correlation between two photons

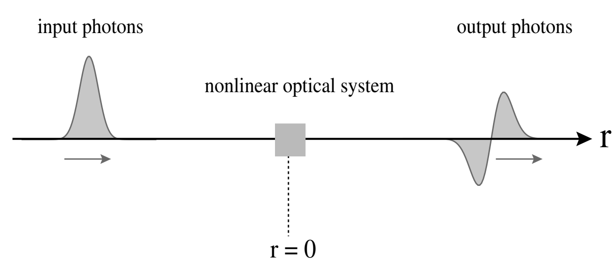

FIG. 1. Illustration of the input and output photons. The input photons interact with a nonlinear optical system placed at $r = 0$, and, after the interaction, propagate into the positive direction.

may be generated after the interaction at the nonlinear system.

We here define the measure of nonlinearity appearing in the output two-photon wave function. As is well known, the field amplitude (the expectation value of the field operator) is zero for the photon number states, so the conventional characterizations of optical nonlinearity cannot be applied in the present case. The nonlinearity appears, not in the field amplitude, but in the wave function of photons. Thus, we quantify the nonlinearity by comparing the output wave function with the *linear* output wave function [7]. Using the one-photon output wave function $\bar{f}(r)$, which is a resultant of a one-photon input $\int dr f(r) a_r^\dagger |0\rangle$, the linear output wave function is given by

$$|\Psi_{\rm out}^{\rm L}\rangle = \int dr_1 dr_2 \bar{f}(r_1)\bar{f}(r_2) a_{r_1}^\dagger a_{r_2}^\dagger |0\rangle. \qquad (3)$$

Such a linear output is expected when the nonlinearity of the system is completely removed. We employ the overlap β between $|\Psi_{\rm out}\rangle$ and $|\Psi_{\rm out}^{\rm L}\rangle$ as the measure of the nonlinearity;

$$\beta = \langle \Psi_{\rm out}^{\rm L}|\Psi_{\rm out}\rangle = 2\int dr_1 dr_2 \bar{f}^*(r_1)\bar{f}^*(r_2)\tilde{g}(r_1, r_2). \quad (4)$$

The nonlinearity parameter β always lies in the unit circle ($|\beta| \le 1$) and becomes unity when the response of the system is completely linear. Thus, deviation of β from unity represents the nonlinearity in the output wave function. The π nonlinear phase shift, which is required for a conditional-NOT gate, is attained when β reaches -1. In the following part of this Letter, we are showing that β can be evaluated from the results of semiclassical theory, bypassing fully quantum-mechanical calculations, which are usually heavy theoretical tasks.

In the semiclassical framework, the light field is treated classically; i.e., the field is characterized by its c-number amplitude. We now consider a situation where a classical field with amplitude $f(r)$ is prepared as the input state. The corresponding input wave function is given by

$$|\Phi_{\rm in}\rangle = \exp\left[-\int dr |f(r)|^2/2\right]\exp\left[\int dr f(r) a_r^\dagger\right]|0\rangle, \quad (5)$$

which is composed by superposition of different number states, as is characteristic to the classical (coherent) state. The zero-, one-, and two-photon components are transformed, after the interaction with the nonlinear system, repsectively, as

$$|0\rangle \to |0\rangle, \qquad (6)$$

$$\int dr f(r) a_r^\dagger |0\rangle \to \int dr \bar{f}(r) a_r^\dagger |0\rangle, \qquad (7)$$

$$\int dr_1 dr_2 f(r_1) f(r_2) a_{r_1}^\dagger a_{r_2}^\dagger |0\rangle \to \int dr_1 dr_2 \tilde{g}(r_1, r_2) a_{r_1}^\dagger a_{r_2}^\dagger |0\rangle. \qquad (8)$$

Thus, up to two-photon states, the output wave function is given by

$$|\Phi_{\rm out}\rangle = \exp\left[-\int dr |f(r)|^2/2\right]\left[1 + \int dr \bar{f}(r) a_r^\dagger + 2^{-1}\int dr_1 dr_2 \tilde{g}(r_1, r_2) a_{r_1}^\dagger a_{r_2}^\dagger\right]|0\rangle. \qquad (9)$$

Using this output wave function, the output field amplitude $f_{\rm out}(r) = \langle \Phi_{\rm out}|a_r|\Phi_{\rm out}\rangle$ is given, up to the third-order response, by

$$f_{\rm out}(r) = f_{\rm out}^{(1)}(r) + f_{\rm out}^{(3)}(r) + \mathcal{O}(f_{\rm out}^{(5)}), \qquad (10)$$

$$f_{\rm out}^{(1)}(r) = \bar{f}(r), \qquad (11)$$

$$f_{\rm out}^{(3)}(r) = \int dr' \bar{f}^*(r')\tilde{g}(r, r') - \bar{f}(r)\int dr' |\bar{f}(r')|^2. \quad (12)$$

The linear output $f_{\rm out}^{(1)}(r)$ is identical to the one-photon output wave function $\bar{f}(r)$, while the third-order output $f_{\rm out}^{(3)}(r)$ contains contracted information of the two-photon output wave function $\tilde{g}(r_1, r_2)$. Remembering that $\bar{f}(r)$ is normalized as $\int dr |\bar{f}(r)|^2 = 2^{-1/2}$, it is confirmed that the following quantity,

$$\beta' = 1 + 2\int dr [f_{\rm out}^{(1)}(r)]^* f_{\rm out}^{(3)}(r), \qquad (13)$$

is identical to the nonlinearity parameter β defined by Eq. (4). Thus, we can evaluate the nonlinearity parameter β from $f_{\rm out}^{(1)}(r)$ and $f_{\rm out}^{(3)}(r)$; these quantities can be determined, within the semiclassical theory, as the linear and the third-order output field against a *classical* input field $f(r)$, which contains $2^{-1/2}$ photons on average.

In order to check the validity of the above semiclassical evaluation, we investigate a concrete example. As an example of a nonlinear system, we employ a two-level system (called an "atom") placed inside of a cavity. The Hamiltonian of the system is given, putting $\hbar = c = 1$, by

$$\mathcal{H} = \omega_a \sigma_+ \sigma_- + g(\sigma_+ c + c^\dagger \sigma_-) + \omega_c c^\dagger c + \int dk k b_k^\dagger b_k + \int dk (\sqrt{\kappa/2\pi} c^\dagger b_k + {\rm H.c.}), \qquad (14)$$

where σ_-, c, and b_k are the annihilation operators for the atom, cavity mode, external photon mode, respectively. ω_a and ω_c represent the frequencies of the atom and the cavity mode (hereafter, $\omega_a = \omega_c$ for simplicity), g represents the atom-cavity coupling, and κ denotes the damping rate of the cavity mode into the external field. The weak (strong) coupling regime is specified by $\kappa/g \gtrsim 4$ ($\kappa/g \lesssim 4$). In this model, due to the neglect of the radiative loss into the noncavity modes (usually denoted by γ), the input photons always appear in the output port. The real-space representation of the external field, b_r, is obtained as the Fourier transform of b_k;

$$b_r = (2\pi)^{-1/2}\int dk e^{ikr} b_k. \qquad (15)$$

This indicates that the interaction between the external field and the cavity mode takes place locally at $r = 0$. In fact, the above Hamiltonian describes the one-sided cavity, where the external field extends only in the positive r region. However, by regarding the incoming photons (which actually propagate in the positive r region into the negative direction) to propagate in the negative r region into the positive direction [9], this system may be reduced to the situation illustrated in Fig. 1.

Using this system, we evaluate the nonlinear parameter β by the following two different methods and compare the results. In one method, after calculating the output wave functions $\bar{f}(r)$ and $\tilde{g}(r_1, r_2)$ in a fully quantum-mechanical fashion, β is rigorously evaluated by Eq. (4). An advantage of using this simple system is that the analytic forms of one- and two-photon propagators are known [7]. In the other method, we calculate the linear and nonlinear components of the output field against a classical input light pulse and evaluate β' by Eq. (13).

The linear and nonlinear optical responses to a classical light pulse are calculated as follows. With the help of the input-output formalism [10], the Heisenberg equations for the cavity mode and the atom are given, respectively, by

$$\dot{c} = (-i\omega_c - \kappa/2)c - ig\sigma_- - i\sqrt{\kappa}b_{t_0-t}(t_0), \qquad (16)$$

$$\dot{\sigma}_- = -i\omega_a\sigma_- - igc + 2ig\sigma_+\sigma_-c, \qquad (17)$$

where the initial moment is denoted by t_0. The third term on the right-hand side of Eq. (17) is the origin of the nonlinearity. The output field is given, in terms of the input field and the cavity mode, by

$$b_r(t) = -b_{r-t+t_0}(t_0) + i\sqrt{\kappa}c(t - r). \qquad (18)$$

The above equations are rigorous equations of motion in the operator form. The semiclassical theory is obtained by approximating the field operator $b_r(t_0)$ for the input field by the c-number amplitude $f(r)$. Then, from Eqs. (16) and (17), we derive the equations of motion for expectation values of operators composed by c, σ_- and their conjugates. (For our purpose of determining up to third-order response, it is sufficient to treat the following quantities: $\langle\sigma_-\rangle$, $\langle c\rangle$, $\langle\sigma_+\sigma_-\rangle$, $\langle\sigma_+c\rangle$, $\langle c^\dagger c\rangle$, $\langle\sigma_-c\rangle$, $\langle cc\rangle$, $\langle\sigma_+\sigma_-c\rangle$, $\langle\sigma_+cc\rangle$, $\langle c^\dagger\sigma_-c\rangle$, and $\langle c^\dagger cc\rangle$). The equations of motion for these quantities are solved perturbatively. The amplitude of the output field is given, taking the expectation value in Eq. (18), as $f_{\mathrm{out}}(r, t) = -f(r - t + t_0) + i\sqrt{\kappa}\langle c(t - r)\rangle$. The first- and third-order components of the output field are given, respectively, by

$$f_{\mathrm{out}}^{(1)}(r, t) = -f(r - t + t_0) + i\sqrt{\kappa}\langle c^{(1)}(t - r)\rangle, \qquad (19)$$

$$f_{\mathrm{out}}^{(3)}(r, t) = i\sqrt{\kappa}\langle c^{(3)}(t - r)\rangle, \qquad (20)$$

where $\langle c^{(1)}\rangle$ and $\langle c^{(3)}\rangle$ represent the linear and the third-order nonlinear responses of the cavity mode. The forms of $f_{\mathrm{out}}^{(1)}$ and $f_{\mathrm{out}}^{(3)}$ are plotted in Fig. 2, where a Gaussian wave packet is chosen for input photons:

$$f(r) = \left(\frac{1}{\pi d^2}\right)^{1/4} \exp\left[-\left(\frac{r-a}{d}\right)^2 + i(q + \omega_c)(r - a)\right], \qquad (21)$$

which is characterized by the coherence length d and the central frequency q (measured from ω_c). $a(<0)$ is an irrelevant parameter denoting the initial position of the packet. From $f_{\mathrm{out}}^{(1)}$ and $f_{\mathrm{out}}^{(3)}$, β' is evaluated by Eq. (13).

Comparison between rigorous quantum-mechanical evaluations (solid lines) and semiclassical evaluations (dotted lines) is carried out in Fig. 3. In Fig. 3(a), the atom-cavity system is in the weak coupling regime ($\kappa/g = 5$). In this case, large nonlinearity is obtained by photons with $q = 0$. In Fig. 3(b), the atom-cavity system is in the strong coupling regime ($\kappa/g = 0.5$). Then, photons with $q = 0$ are no more resonant and small nonlinearity results; large nonlinearity is obtained by photons with $q \simeq \pm g$, which are tuned to the Rabi-split frequency. The optimum conditions for obtaining large nonlinearity have been clarified in Ref. [7]. This figure shows the validity of the semiclassical evaluation; the two-photon nonlinearity can be evaluated by the semiclassical method under any choice of parameters. The slight deviations of the semiclassical evaluation from the rigorous results are ascribed to the semiclassical approximation, in which the operator $b_r(t_0)$ for the input field is replaced with the c-number amplitude $f(r)$. Although this approximation is validated when the input field is intense, it becomes less valid when the input field is weak, as is considered here. However, it should be noted

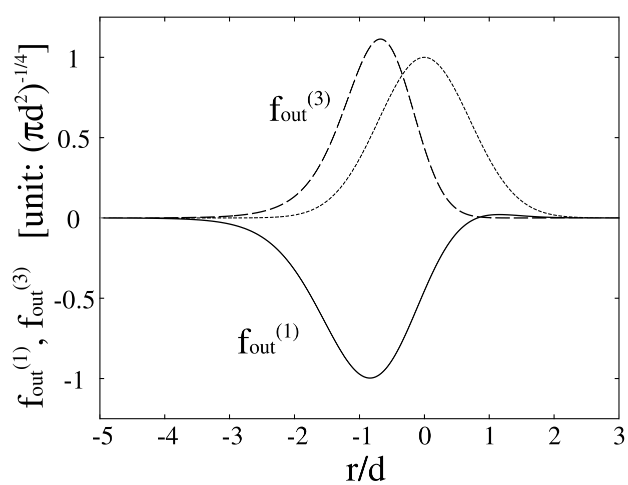

FIG. 2. The forms of the linear (solid curve) and third-order nonlinear (dashed curve) components of the output field. The thin dotted line shows the input field. A moving coordinate system at the light velocity is employed for the horizontal axis. The cavity parameter is $\kappa/g = 5$ (weak coupling regime), and the parameters for the input pulse are chosen as $g^2d/\kappa = 1$ and $q = 0$. The input and output fields become real after removing the natural phase, $e^{i\omega_c r}$.

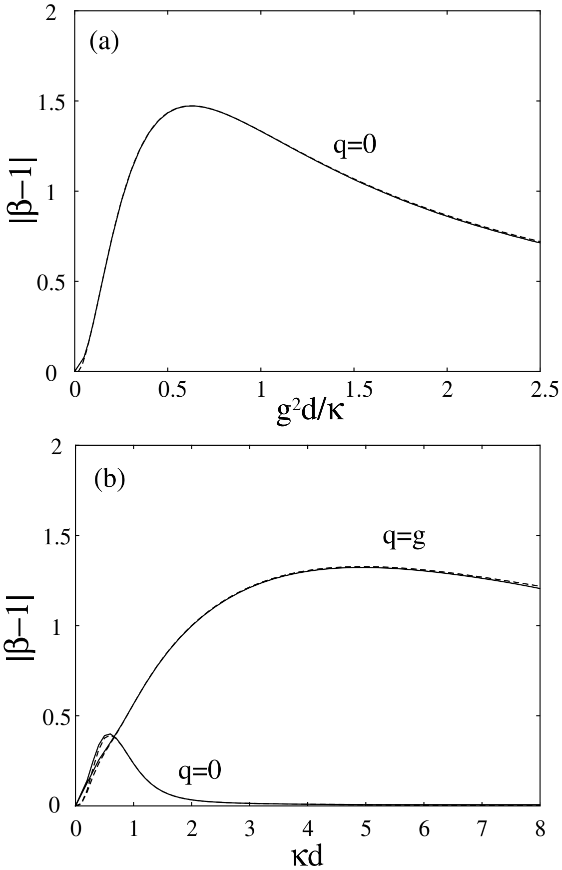

FIG. 3. Dependence of the two-photon nonlinearity β on the pulse length d. The frequencies q of input photons are indicated. The atom-cavity system is in the weak coupling regime ($\kappa/g = 5$) in (a) and in the strong coupling regime ($\kappa/g = 0.5$) in (b). The rigorous evaluations are plotted by the solid lines, and the semiclassical evaluations are plotted by the dotted lines.

that the above replacement is rigorous in the limit of infinitely weak coupling, $\kappa/g \rightarrow \infty$. This explains the fact that the deviations are smaller in the weak coupling regime than in the strong coupling regime.

Finally, we comment on the applicability of the semiclassical evaluation. In deriving Eq. (13), it is assumed in Eqs. (2), (3), (7), and (8) that the input photons appear in the output port without leaving any trace (elementary excitations) in the nonlinear material, and no other system-dependent features are used. It is therefore expected that this evaluation method is widely applicable to nonlinear systems satisfying the above condition. Even for the dissipative nonlinear systems exposed strongly to environmental agents, we can use this method for the estimation of the two-photon nonlinearity in the dissipation-free limit of such systems, which would still be informative in designing quantum-optical devises. This method would be particularly valuable in evaluating the two-photon nonlinearity in complex nonlinear systems with many mechanical degrees of freedom, where full quantum-mechanical analyses are much more difficult than semiclassical ones. It should also be noted that the use of this method is restricted to cases where the input two-photon wave function is separable; this is because the entangled input state has no correspondent classical state.

In summary, we have proposed a method to evaluate the two-photon nonlinearity using a semiclassical method, bypassing full quantum-mechanical calculations. The prescription is as follows: calculate the linear and the third-order nonlinear components of the output field against a classical input pulse containing $2^{-1/2}$ photons on average; then, the two-photon nonlinearity is evaluated by Eq. (13). Taking an atom-cavity system as an example of nonlinear systems, we have compared the semiclassically evaluated values with rigorous values in Fig. 3, which demonstrated the validity of the evaluation method. Thus, the link between the conventional nonlinear optics and the quantum-mechanical optical nonlinearity has been clarified. This would be helpful in utilizing vast knowledge accumulated in the field of nonlinear optics for the investigation of two-photon nonlinearity and other quantum-mechanical optical nonlinearities.

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*Electronic address: ikuzak@aria.mp.es.osaka-u.ac.jp

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