

## Multiphoton wavefunction after the Kerr interaction

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The multiphoton wavefunction after the Kerr interaction is obtained analytically for an arbitrary photon number. The wavefunction is composed of two fundamental functions: the input mode function and the linear response function. The nonlinear effects appearing in this wavefunction are evaluated quantitatively, revealing the limitations of nonlinear quantum optics theories based on single-mode approximations.

### I. INTRODUCTION

With the goal of achieving all-optical quantum information processing, single-photon engineering has become one of the hottest research topics in physics. Rapid progress has been made in generating and detecting single photons, and also in information processing based on linear optics. Furthermore, the discovery of optical nonlinearity that is sensitive to individual photons has raised the possibility of using one photon to control another photon, increasing the need to develop a quantitative theory of nonlinear quantum optics. The simplest method to analyze the nonlinear dynamics of photons is to introduce an effective nonlinear-interaction Hamiltonian based on the single-mode approximation. For example, the following time evolution operator has conventionally been used for the self-Kerr interaction:

$$\hat{U} = \exp(-it\chi c^\dagger c^\dagger cc), \quad (1)$$

where  $c^\dagger$  is a single-mode photon creation operator,  $t$  is the interaction time, and  $\chi$  is the coupling coefficient, which is proportional to the nonlinear susceptibility. This method offers intuitive pictures of nonlinear dynamics, and has led to many proposals in photon engineering based on nonlinear optics. However, such theories are unsuitable for more quantitative analyses due to the phenomenological introduction of  $t$  and  $\chi$ . Furthermore, single-mode treatment generally becomes invalid after photons mutually interact.

Since nonlinear optical processes are sensitive to the spatiotemporal distribution of the photon field, in quantitative analyses it is essential to incorporate the multimode nature of the field. In this direction, a successful approach has been the noise-operator formalism, in which photons are treated as active mechanical degrees of freedom, while optical

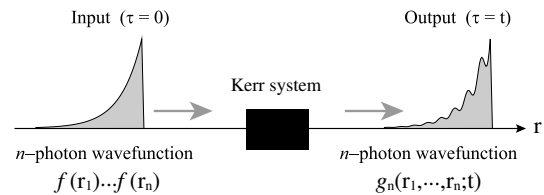


FIG. 1: The physical situation considered in this study. Initially ( $\tau = 0$ ), the  $n$  input photons are uncorrelated and have identical mode functions  $f(r)$ . After the nonlinear interaction ( $\tau = t$ ) the photons become correlated. The output wavefunction is denoted by  $g_n(r_1, \dots, r_n; t)$ .

materials are treated implicitly through non-instantaneous response functions and noise operators [1–3]. A more rigorous approach is the full-quantum formalism, in which both photons and materials are treated as active mechanical degrees of freedom [4]. Since the exchange of quantum coherence between photons and materials can be handled rigorously, this approach is the most convincing in revealing the true nature of nonlinear dynamics of photons.

In this study, the Kerr interaction of photons is analyzed using the full-quantum formalism, by explicitly accounting for the intrinsic wave-packet nature of photons (see Fig. 1). By using a two-level system as a model Kerr system, the output  $n$ -photon wavefunction is derived in an analytic form for an arbitrary photon number  $n$ , and the nonlinear effects appearing in this wavefunction are evaluated quantitatively. As a result, a microscopic basis is provided for effective theories such as that represented by Eq. (1), and the limitations of such theories are simultaneously exposed. The current results demonstrate both the necessity and the potential of multimode full-quantum analysis in nonlinear quantum optics theory.

## II. THEORETICAL MODEL

### A. Hamiltonian

The physical situation considered in this study is illustrated in Fig. 1. The overall system consists of a one-dimensional photon field and a Kerr system. A  $n$ -photon pulse is input from the left-hand side ( $r < 0$ ), interacts with the Kerr system located at the center ( $r \sim 0$ ), and is output into the right-hand side ( $r > 0$ ). The Kerr system is assumed to be transparent and to conserve the photon number. As the simplest system showing the third-order optical nonlinearity, we employ a single two-level system (referred to hereafter as an ‘‘atom’’) as a model Kerr system. In a rotating frame with respect to the atomic resonance, the Hamiltonian of the whole system is given by (setting  $\hbar = c = 1$ )

$$\mathcal{H} = \int dk \left[ kc_k^\dagger c_k + i\sqrt{\Gamma/2\pi}(\sigma^\dagger c_k - c_k^\dagger \sigma) \right], \quad (2)$$

where  $\sigma^\dagger$  is the Pauli raising operator for the atomic excitation, and  $c_k^\dagger$  is the photon creation operator in the wave number representation, and  $\Gamma$  represents the natural linewidth of the atom. The commutators for  $\sigma$  and  $c_k$  are given by  $[\sigma, \sigma^\dagger] = 1 - 2\sigma^\dagger \sigma$  and  $[c_k, c_{k'}^\dagger] = \delta(k - k')$ , respectively. The real-space photon operator  $\tilde{c}_r$  is connected to  $c_k$  by

$$\tilde{c}_r = (2\pi)^{-1/2} \int dk e^{ikr} c_k. \quad (3)$$

The ground state of the whole system (product of the atomic ground state and the photonic vacuum state) is denoted by  $|0\rangle$ .

### B. Input and output photons

The input and output photons are characterized as follows. Throughout this study, the time variable is denoted by  $\tau$ , and the initial and final times are set to  $\tau = 0$  and  $t$ , respectively. At the initial moment ( $\tau = 0$ ), the input photons are in the  $n$ -photon Fock state,  $|n\rangle = (n!)^{-1/2}(c^\dagger)^n|0\rangle$ , where  $c^\dagger$  is a single-mode photon creation operator. In the multimode notation,  $c^\dagger$  is given by  $c^\dagger = \int dr f(r)\tilde{c}_r^\dagger$ , where  $f(r)$  denotes the input mode function, normalized as  $\int dr |f(r)|^2 = 1$  and localized in the  $r < 0$  region. Thus, in the multimode notation the

input state vector is given by

$$|n_{\text{in}}\rangle = (n!)^{-1/2} \int d^n r f(r_1) \cdots f(r_n) \tilde{c}_{r_1}^\dagger \cdots \tilde{c}_{r_n}^\dagger |0\rangle. \quad (4)$$

Namely, the  $n$  input photons are identical and uncorrelated. In contrast, at the final moment ( $\tau = t$ ), the  $n$  photons become correlated as a result of the nonlinear interaction. We therefore employ a general form for the output state vector:

$$|n_{\text{out}}\rangle = (n!)^{-1/2} \int d^n r g_n(r_1, \cdots, r_n; t) \tilde{c}_{r_1}^\dagger \cdots \tilde{c}_{r_n}^\dagger |0\rangle, \quad (5)$$

where  $g_n$  is a symmetric function of the space coordinates, normalized as  $\int d^n r |g_n(r_1, \cdots, r_n; t)|^2 = 1$  and localized in the  $r > 0$  region.

## III. $n$ -PHOTON OUTPUT WAVEFUNCTION

The  $n$ -photon output wavefunction  $g_n(r_1, \cdots, r_n; t)$  can be obtained by solving the Schrödinger equation,

$$|n_{\text{out}}\rangle = e^{-i\mathcal{H}t}|n_{\text{in}}\rangle. \quad (6)$$

However, instead of solving this equation directly in the  $n$ -quanta Hilbert space, it is more convenient to assume a classical input and to solve the resulting equations perturbatively [5]. The  $n$ -photon output wavefunction  $g_n(r_1, \cdots, r_n; t)$  is obtained by the following rules: (i)  $g_n$  is expressed in terms of the input mode function  $f$  and the atomic correlation functions  $s_1, \cdots, s_n$ , as

$$g_n(r_1, \cdots, r_n; t) = f(-t_1) \cdots f(-t_n) \times \left[ 1 - \sum_i \frac{s_1(t_i)}{f(-t_i)} + \sum_{i < j} \frac{s_2(t_i, t_j)}{f(-t_i)f(-t_j)} - \sum_{i < j < k} \frac{s_3(t_i, t_j, t_k)}{f(-t_i)f(-t_j)f(-t_k)} + \cdots + (-1)^n \frac{s_n(t_1, \cdots, t_n)}{f(-t_1) \cdots f(-t_n)} \right], \quad (7)$$

where  $t_j \equiv t - r_j$  and  $\sum_{i < j}$  runs over  $i$  and  $j$  satisfying  $1 \leq i < j \leq n$ . (ii) The  $n$ -point atomic correlation function  $s_n$  can be expressed in terms of the one-point atomic correlation function  $s$  as

$$s_n(t_1, \cdots, t_n) = s(t_n) \prod_{j=1}^{n-1} [s(t_j) - e^{(t_{j+1}-t_j)/2} s(t_{j+1})]. \quad (8)$$

(iii) The one-point atomic correlation function  $s$  is the Laplace transform of the input mode function  $f$ , as given by

$$s(t) = \int_0^\infty d\xi f(-t + \xi) e^{-\xi/2}. \quad (9)$$

Thus, the output wavefunction  $g_n(r_1, \dots, r_n; t)$  can be expressed in terms of two fundamental one-variable functions [the input mode function  $f(r)$ , and the linear response function  $s(t)$  given by Eq. (9)] for an arbitrary photon number  $n$ .  $g_n$  is a symmetric function of the space coordinates, and is given, for  $r_1 \leq \dots \leq r_n$ , by Eqs. (7) and (8), where  $t_j = t - r_j$ . For example,  $g_1$  and  $g_2$  are given by

$$g_1(r; t) = f(r - t) - s(t - r), \quad (10)$$

$$g_2(r_1, r_2; t) = g_1(r_1; t)g_1(r_2; t) - e^{(r_1 - r_2)/2} s^2(t - r_2). \quad (11)$$

#### IV. CHARACTERIZATION OF NONLINEAR EFFECTS

##### A. Input mode function

Now that we have obtained the output wavefunctions, we proceed to characterize the nonlinear effects appearing in the output photons. Hereafter, we employ the following form for the input mode function:

$$f(r) = \begin{cases} \sqrt{2/d} e^{r/d + ikr} & (r \leq 0) \\ 0 & (r > 0) \end{cases}, \quad (12)$$

where  $d$  and  $k$  represent the coherence length and the frequency (measured from the atomic resonance) of the input photons, respectively. The nonlinear effects are maximized when the input photons are in resonance with the material ( $k \sim 0$ ). However, since off-resonant photons are actually used to avoid absorption by the material, we discuss off-resonant photons ( $|k| \gg \Gamma$ ) in the following.

##### B. Nonlinear phase shift

Firstly, we evaluate the nonlinear phase shift appearing in the output wavefunction  $g_n$ . For this purpose, we define a *linear*  $n$ -photon output wavefunction by

$$g_n^L(r_1, \dots, r_n; t) = \prod_{j=1}^n g_1(r_j; t). \quad (13)$$

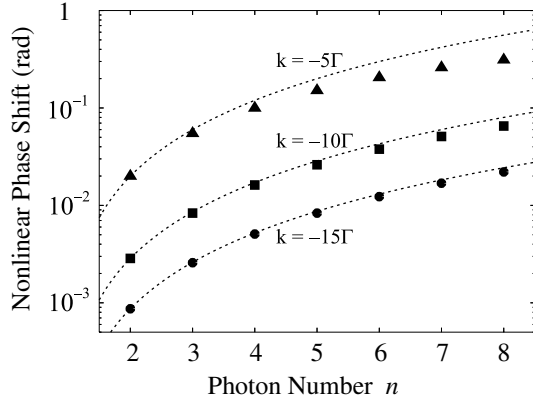


FIG. 2: The nonlinear phase shift as a function of the photon number. The input pulse parameters are  $d = 2\Gamma^{-1}$  and  $k = -5\Gamma$  (triangles),  $-10\Gamma$  (squares) and  $-15\Gamma$  (circles). The dotted lines show the predictions of the effective Hamiltonian of Eq. (1),  $\theta_n = \theta_2 \times n(n-1)/2$ .

This linear output is expected in the absence of the nonlinear interaction. The nonlinear effects are evaluated through the overlap  $\alpha_n$  between the linear and nonlinear output, as given by

$$\alpha_n = \int d^n r [g_n^L(r_1, \dots, r_n; t)]^* g_n(r_1, \dots, r_n; t), \quad (14)$$

which becomes independent of  $t$  sufficiently after the interaction. The nonlinear phase shift  $\theta_n$  is expressed by the phase of  $\alpha_n$ , namely,  $\theta_n \equiv -\text{Im}(\log \alpha_n)$ . The effective theory predicts that  $\theta_n$  is proportional to  $n(n-1)$ , since  $\alpha_n = e^{-it\chi n(n-1)}$  due to Eq. (1). In Fig. 2, the nonlinear phase shift is plotted as a function of the photon number. As expected, the nonlinear phase shift increases with the photon number  $n$  and decreases with the detuning  $|k|$ . The prediction of the effective theory,  $\theta_n = \theta_2 \times n(n-1)/2$ , is also plotted with dotted lines for reference. It is observed that the effective theory agrees well with the rigorous results, provided the nonlinear phase shift is small ( $k = -15\Gamma$  in Fig. 2). However, the effective theory becomes invalid for evaluating larger nonlinear phase shifts. The actual phase shifts are considerably smaller than those predicted by the effective theory ( $k = -5\Gamma$  in Fig. 2). For example, if the allowable error is set at 5%, the effective theory of Eq. (1) can be justified only in the small phase-shift region satisfying  $\theta \lesssim 10^{-2}$ .

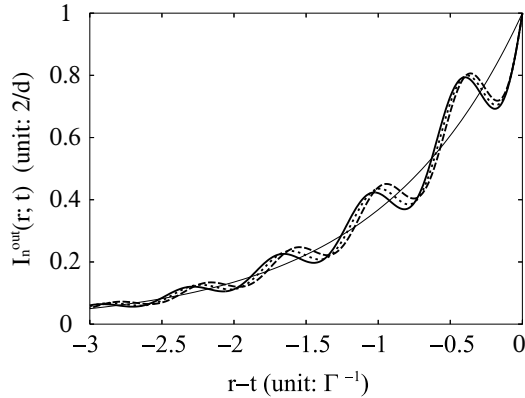


FIG. 3: The intensity profile  $I_n^{\text{out}}(r; t)$  of the output photons, for the photon numbers  $n = 1$  (solid line), 4 (dotted line) and 8 (dashed line). The input pulse parameters are  $d = 2\Gamma^{-1}$  and  $k = -10\Gamma$ . The input photon profile  $I^{\text{in}}(r) = (2/d)e^{2r/d}$  is also plotted using a thin solid line for reference.

### C. Shape of output pulse

Next, we observe the shape of the output photon pulse. In the input state of Eq. (4), all photons have an identical single-mode function  $f(r)$ . However, such a single-mode description cannot be used for the output photons, since the photons become correlated after the nonlinear interaction, as indicated by Eq. (11). Instead, we characterize the profile of the output photons using a normalized intensity distribution  $I_n^{\text{out}}(r; t)$ , defined by

$$I_n^{\text{out}}(r; t) = \frac{\langle n_{\text{out}} | \tilde{c}_r^\dagger \tilde{c}_r | n_{\text{out}} \rangle}{n}. \quad (15)$$

Note that  $I_n^{\text{out}}(r; t)$  is a real and positive function normalized as  $\int dr I_n^{\text{out}}(r; t) = 1$ . In Fig. 3,  $I_n^{\text{out}}(r; t)$  is plotted for the photon numbers  $n = 1, 4$  and  $8$ . The input photon profile [ $I^{\text{in}}(r) \equiv \langle n_{\text{in}} | c_r^\dagger c_r | n_{\text{in}} \rangle / n = (2/d)e^{2r/d}$ , regardless of  $n$ ] is also plotted for reference. The weak oscillation observed in the output photon profile is due to the interference between the transmission and emis-

sion components [i.e., the first and second terms in Eq. (10)]. It is observed that the output photons are delayed relative to the input, due to the absorption and re-emission by the material. The nonlinear effect appears as a slight advancing of the output pulse. This is because the efficiency per photon of the delay mechanism decreases the more photons are involved. This  $n$ -dependent deformation of the pulse profile is completely neglected in the effective theory based on the single-mode approximation. However, since the interferability of photon pulses is sensitive to the pulse profile, such deformation must be taken into account in the construction of single-photon devices.

## V. SUMMARY

In summary, the Kerr interaction of  $n$  photons occurring at a two-level system has been investigated using a multimode full-quantum formalism. The  $n$ -photon output wavefunction has been obtained analytically for an arbitrary photon number  $n$ , and the nonlinear effects appearing in the output have been quantitatively evaluated. The following two features, which are essential for the construction of single-photon devices, have been clarified: (i) the actual nonlinear phase shift is smaller than the phase shift predicted by the effective theory (Fig. 2), and (ii) the output pulse profile varies considerably with the photon number (Fig. 3). These results demonstrate both the necessity and the potential of multimode analysis in nonlinear quantum optics theory.

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