Multiphoton wave function after Kerr interaction

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

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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 [1–11]. 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 [12–15]. 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:

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

where c^{\dagger} is a single-mode photon creation operator, t is the interaction time, and χ is the coupling coefficient, which is proportional to the nonlinear susceptibility. (For other types of nonlinear processes, refer to Ref. [16].) This method offers intuitive pictures of nonlinear dynamics and has led to many proposals in photon engineering based on nonlinear optics [17–22]. However, such theories are unsuitable for more quantitative analyses due to the phenomenological introduction of t and χ . Furthermore, the 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 materials are treated implicitly through noninstantaneous response functions and noise operators [23–25]. A more rigorous approach is the full-quantum formalism, in which both photons and materials are treated as active mechanical degrees of freedom [26]. 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 wave function is derived in an analytic form for an arbitrary photon number n, and the nonlinear effects appearing in this wave function 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. SYSTEM

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 $(r\sim0)$, 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 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)

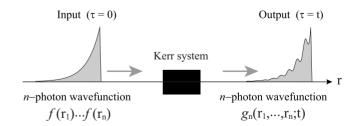


FIG. 1. The physical situation considered in this study. Initially $(\tau=0)$, the n input photons are uncorrelated and have identical mode functions f(r). After nonlinear interaction $(\tau=t)$ the photons become correlated. The output wave function is denoted by $g_n(r_1,\ldots,r_n;t)$.

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

where σ^{\dagger} is the Pauli raising operator for the atomic excitation, c_k^{\dagger} is the photon creation operator in the wave-number representation, and Γ represents the natural linewidth of the atom. The commutators for σ 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

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

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 τ , and the initial and final times are set to τ =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 $c^{\dagger} = \int dr \, f(r) \widetilde{c}_r^{\dagger}$, 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

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

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

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

III. ANALYSIS

A. Strategy

We now start to determine the output wave function g_n . A straightforward method is to solve the Schrödinger equation $|n_{\text{out}}\rangle = e^{-i\mathcal{H}t}|n_{\text{in}}\rangle$ in the n-quantum Hilbert space. However, instead of working in this Hilbert space, it is more convenient to consider a *classical* input pulse and to collect the relevant terms afterwards [27,28]. Thus, we consider the following classical state as the input:

$$|\phi_{\rm in}\rangle = \mathcal{N} \exp\left[\mu \int dr f(r) \tilde{c}_r^{\dagger}\right] |0\rangle,$$
 (6)

where μ is a complex amplitude and $\mathcal{N}(=e^{-|\mu|^2/2})$ is a normalization constant. This state represents a classical photon

pulse having an amplitude of $\mu f(r)$. This state consists of different number states, as expressed by

$$|\phi_{\rm in}\rangle = \mathcal{N} \sum_{n=0}^{\infty} \frac{\mu^n}{\sqrt{n!}} |n_{\rm in}\rangle.$$
 (7)

From the linearity of the Schrödinger equation, the output state for this classical input is given by

$$|\phi_{\text{out}}\rangle = e^{-i\mathcal{H}t}|\phi_{\text{in}}\rangle = \mathcal{N}\sum_{n=0}^{\infty} \frac{\mu^n}{\sqrt{n!}}|n_{\text{out}}\rangle.$$
 (8)

The *n*-point correlation function for this output, defined by $\mathcal{G}_n(r_1,\ldots,r_n;t) = \langle \phi_{\text{out}} | \widetilde{c}_{r_1} \cdots \widetilde{c}_{r_n} | \phi_{\text{out}} \rangle$, is given by

$$G_n(r_1, \dots, r_n; t) = \mu^n g_n(r_1, \dots, r_n; t) + O(|\mu|^{n+2}),$$
 (9)

which is derived with the help of Eq. (5). Thus, the *n*-photon output wave function g_n can be obtained by assuming a classical input pulse of Eq. (6), calculating the *n*-point correlation function \mathcal{G}_n , and collecting the lowest-order components proportional to μ^n .

B. Field correlation function

The correlation functions are calculated in the Heisenberg representation. Throughout this study, the initial and final field operators $\tilde{c}_r(0)$ and $\tilde{c}_r(t)$ are referred to as the input and output operators, respectively. As observed in Fig. 1, the negative-r region (-t < r < 0) is relevant for input operators, whereas the positive-r region (0 < r < t) is relevant for output operators. Setting $\Gamma = 1$, the Heisenberg equation for the atom is given by

$$\frac{d}{d\tau}\sigma = -\frac{\sigma}{2} + (1 - 2\sigma^{\dagger}\sigma)\tilde{c}_{-\tau}(0), \tag{10}$$

where $\tilde{c}_{-\tau}(0)$ is the input operator at $r=-\tau(<0)$. The output operator is given by

$$\widetilde{c}_r(t) = \widetilde{c}_{r-t}(0) - \sigma(t-r), \tag{11}$$

where 0 < r < t. Note that both Eqs. (10) and (11) are derivable from Eq. (2) (see Appendix A). Employing the notation $\langle \cdots \rangle \equiv \langle \phi_{\rm in} | \cdots | \phi_{\rm in} \rangle$ hereafter, the correlation function \mathcal{G}_n can be written as

$$G_n(r_1, \dots, r_n; t) = \langle \widetilde{c}_{r_1}(t) \cdots \widetilde{c}_{r_n}(t) \rangle.$$
 (12)

Since the simultaneous field operators are commutable, we can set $r_1 \le \cdots \le r_n$ in Eq. (12) without loss of generality. Furthermore, Eqs. (11) and (12) show that \mathcal{G}_n is a function of $t-r_j$ $(j=1,\ldots,n)$. Therefore, we introduce a new set of variables $t_j \equiv t-r_j$ $(j=1,\ldots,n)$, satisfying $0 < t_n \le \cdots \le t_1 < t$.

The following properties are useful for evaluating the correlation function. (I) $|\phi_{\text{in}}\rangle$ is an eigenstate of the input operator $\tilde{c}_r(0)$ —namely, $\tilde{c}_r(0)|\phi_{\text{in}}\rangle=\mu f(r)|\phi_{\text{in}}\rangle$. Note that $\tilde{c}_r(0)$ is an operator, whereas $\mu f(r)$ is a c number. (II) The input operator $\tilde{c}_r(0)$ commutes with $\sigma(\tau)$ if $r < -\tau$ (see Appendix B).

Using these properties and Eqs. (11) and (12), we obtain, for n=1,2, for example,

$$G_1(r_1;t) = \mu f(-t_1) - S_1(t_1),$$
 (13)

$$G_2(r_1, r_2; t) = \mu^2 f(-t_1) f(-t_2) - \mu f(-t_1) S_1(t_2) - \mu f(-t_2) S_1(t_1) + S_2(t_1, t_2),$$
(14)

where $S_n(t_1,\ldots,t_n)$ is the *n*-point atomic correlation function, defined by $S_n = \langle \sigma(t_1) \cdots \sigma(t_n) \rangle$. As will be observed later, S_n is composed of terms proportional to $\mu^n |\mu|^{2j}$ $(j=0,1,\ldots)$. However, by comparing Eqs. (13) and (14) with Eq. (9), it is noticed that only the lowest-order components of S_n (proportional to μ^n) are relevant for g_n . Denoting the lowest-order components of S_n by S_n , the output wave function S_n is given by

$$g_{n}(r_{1}, \dots, r_{n}; t)$$

$$= f(-t_{1}) \cdots f(-t_{n}) \left[1 - \sum_{i} \frac{s_{1}(t_{i})}{f(-t_{i})} + \sum_{i < j} \frac{s_{2}(t_{i}, t_{j})}{f(-t_{i})f(-t_{j})} - \sum_{i < j < k} \frac{s_{3}(t_{i}, t_{j}, t_{k})}{f(-t_{i})f(-t_{j})f(-t_{k})} + \cdots + (-1)^{n} \frac{s_{n}(t_{1}, \dots, t_{n})}{f(-t_{1}) \cdots f(-t_{n})} \right],$$

$$(15)$$

where, for example, $\Sigma_{i < j}$ runs over i and j satisfying $1 \le i < j \le n$. Thus, g_n is expressed in terms of the input mode function f and the atomic correlation functions s_1, \ldots, s_n .

C. Atomic correlation function

Our final task is to evaluate the atomic correlation function. From Eqs. (6) and (10), we obtain

$$\frac{d}{dt_1}S_1(t_1) = -\frac{S_1(t_1)}{2} + \mu f(-t_1) - 2\mu f(-t_1)\langle \sigma^{\dagger} \sigma \rangle. \quad (16)$$

Since $\langle \sigma^{\dagger} \sigma \rangle$ is at least a second-order quantity in $|\mu|$, the last term is irrelevant for the first-order quantity s_1 . Thus, the equation of motion for s_1 is given by

$$\frac{d}{dt_1}s_1(t_1) = -\frac{s_1(t_1)}{2} + f(-t_1),\tag{17}$$

which describes the linear response of the atom. This equation is formally integrated to give

$$s_1(t_1) = \int_0^\infty d\xi f(-t_1 + \xi)e^{-\xi/2}.$$
 (18)

The equation of motion for $S_2(t_1,t_2)$ is given, from Eqs. (6) and (10), by

$$\frac{d}{dt_1}S_2(t_1, t_2) = -\frac{S_2(t_1, t_2)}{2} + \mu f(-t_1)S_1(t_2)
-2\mu f(-t_1)\langle \sigma^{\dagger}(t_1)\sigma(t_1)\sigma(t_2)\rangle.$$
(19)

Neglecting again the last term yielding the higher-order contributions, the equation of motion for the second-order component s_2 is given by

$$\frac{d}{dt_1}s_2(t_1, t_2) = -\frac{s_2(t_1, t_2)}{2} + s_1(t_2)f(-t_1), \tag{20}$$

with the initial condition of $s_2(t_2, t_2) = 0$ since $\sigma^2 = 0$. s_2 is given by

$$s_2(t_1, t_2) = [s_1(t_1) - e^{(t_2 - t_1)/2} s_1(t_2)] s_1(t_2), \tag{21}$$

where the second term in the brackets is introduced to satisfy the initial condition. Using the same logic for arbitrary n, s_n can be expressed in terms of s_1 as

$$s_n(t_1, \dots, t_n) = [s_1(t_1) - e^{(t_2 - t_1)/2} s_1(t_2)] s_{n-1}(t_2, \dots, t_n)$$

$$= s_1(t_n) \prod_{i=1}^{n-1} [s_1(t_i) - e^{(t_{j+1} - t_j)/2} s_1(t_{j+1})]. \quad (22)$$

D. n-photon output wave function

Thus, the output wave function $g_n(r_1, \ldots, r_n; t)$ can be expressed in terms of two fundamental one-variable functions [the input mode function f(r) and the linear response function $s_1(\tau)$ given by Eq. (18)] for an arbitrary photon number n. g_n is a symmetric function of the space coordinates and is given, for $r_1 \le \cdots \le r_n$, by Eqs. (15) and (22), where $t_i = t - r_i$. For example, g_1 and g_2 are given by

$$g_1(r;t) = f(r-t) - s_1(t-r),$$
 (23)

$$g_2(r_1, r_2; t) = g_1(r_1; t)g_1(r_2; t) - e^{(r_1 - r_2)/2}s_1^2(t - r_2).$$
 (24)

IV. CHARACTERIZATION OF NONLINEAR EFFECTS

A. Input mode function

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

$$f(r) = \begin{cases} \sqrt{2/d}e^{r/d + ikr} & (r \le 0), \\ 0 & (r > 0), \end{cases}$$
 (25)

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

First, we evaluate the nonlinear phase shift appearing in the output wave function g_n . For this purpose, we define a *linear n*-photon output wave function by

$$g_n^{L}(r_1, \dots, r_n; t) = \prod_{i=1}^n g_1(r_i; t).$$
 (26)

This linear output is expected in the absence of the nonlinear interaction. The nonlinear effects are evaluated through the

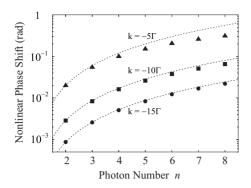


FIG. 2. The nonlinear phase shift as a function of the photon number. The input pulse parameters are $d=2\Gamma^{-1}$ and $k=-5\Gamma$ (triangles), -10Γ (squares), and -15Γ (circles). The dotted lines show the predictions of the effective Hamiltonian of Eq. (1), $\theta_n = \theta_2 n(n-1)/2$.

overlap α_n between the linear and nonlinear output, as given by

$$\alpha_n = \int d^n r [g_n^{L}(r_1, \dots, r_n; t)]^* g_n(r_1, \dots, r_n; t), \qquad (27)$$

which becomes independent of t sufficiently after the interaction. The nonlinear phase shift θ_n is expressed by the phase of α_n —namely, $\theta_n \equiv -\text{Im}(\ln \alpha_n)$. The effective theory predicts that θ_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 $(k=-5\Gamma \text{ in Fig. 2})$. 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 $\theta \lesssim 10^{-2}$.

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. (24). Instead, we characterize the profile of the output photons using a normalized intensity distribution $I_n^{\text{out}}(r;t)$, defined by

$$I_n^{\text{out}}(r;t) = \frac{\langle n_{\text{out}} | \tilde{c}_r^{\dagger} \tilde{c}_r | n_{\text{out}} \rangle}{n}.$$
 (28)

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

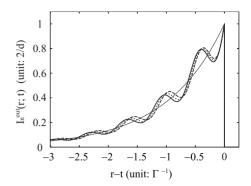


FIG. 3. The intensity profile $I_n^{\text{out}}(r;t)$ of the output photons for the photon numbers n=1 (solid line), 4 (dotted line), and 8 (dashed line). The input pulse parameters are $d=2\Gamma^{-1}$ and $k=-10\Gamma$. The input photon profile $I^{\text{in}}(r)=(2/d)e^{2r/d}$ is also plotted using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

 $[I^{\rm in}(r) \equiv \langle n_{\rm in}|c_r^{\dagger}c_r|n_{\rm in}\rangle/n = (2/d)e^{2r/d}$, 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 emission components [i.e., the first and second terms in Eq. (23)]. It is observed that the output photons are delayed relative to the input, due to the absorption and reemission 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 a 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 wave function 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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APPENDIX A: DERIVATION OF EQS. (10) and (11)

The Heisenberg equations for σ and c_k are given from Eq. (2) by

$$\frac{d}{d\tau}\sigma = \sqrt{\Gamma}(1 - 2\sigma^{\dagger}\sigma)\tilde{c}_0,\tag{A1}$$

$$\frac{d}{d\tau}c_k = -ikc_k - \sqrt{\frac{\Gamma}{2\pi}}\sigma. \tag{A2}$$

From Eq. (A2), the operator c_k at time τ (0 < τ < t) is represented in two ways:

$$c_k(\tau) = c_k(0)e^{-ik\tau} - \sqrt{\frac{\Gamma}{2\pi}} \int_0^{\tau} d\tau' \ \sigma(\tau')e^{-ik(\tau - \tau')} \quad (A3)$$

$$=c_k(t)e^{-ik(\tau-t)}+\sqrt{\frac{\Gamma}{2\pi}}\int_{\tau}^t d\tau' \ \sigma(\tau')e^{-ik(\tau-\tau')}. \tag{A4}$$

Using the above two forms of $c_k(\tau)$, $\tilde{c}_0(\tau)$ is recast into the following two forms:

$$\widetilde{c}_0(\tau) = \widetilde{c}_{-\tau}(0) - \frac{\sqrt{\Gamma}}{2}\sigma(\tau) \tag{A5}$$

$$=\widetilde{c}_{t-\tau}(t)+\frac{\sqrt{\Gamma}}{2}\sigma(\tau). \tag{A6}$$

Note that \tilde{c}_r is in the real-space representation, as defined by Eq. (3). Equating the right-hand sides and introducing a new label $r(=t-\tau)$, we obtain the input-output relation of Eq.

(11), in which the output field operator is expressed in terms of the input field operator and the atomic operator at $\tau = t - r$. Substituting Eq. (A5) into Eq. (A1), the atomic Heisenberg equation (10) is obtained.

APPENDIX B: PROOF OF COMMUTATIVITY

Here we prove that the input operator $\tilde{c}_r(0)$ commutes with the atomic operator $\sigma(\tau)$, if $r < -\tau$ is satisfied. For this purpose, we formally solve Eq. (10) as the following form:

$$\sigma(\tau) = \sigma(0)e^{-\tau/2} + \int_0^{\tau} d\tau' \ e^{(\tau' - \tau)/2} [1 - 2\sigma^{\dagger}(\tau')\sigma(\tau')] \widetilde{c}_{-\tau'}(0).$$
(B1)

Thus, $\sigma(\tau)$ is composed of $\sigma(0)$, $\tilde{c}_{r'}(0)$, and $\sigma(\tau')$ and its conjugate, where r' and τ' satisfy $-\tau < r' < 0$ and $0 < \tau' < \tau$, respectively. In order to express $\sigma(\tau)$ in terms only of the initial operators, we use an iteration method. Then, $\sigma(\tau')$ in Eq. (B1) is composed of $\sigma(0)$, $\tilde{c}_{r''}(0)$, and $\sigma(\tau'')$ and its conjugate, where r'' and τ'' satisfy $-\tau' < r'' < 0$ and $0 < \tau'' < \tau'$. Note that r'' necessarily satisfies $-\tau < r'' < 0$ since $\tau' < \tau$.

After infinite iterations, we can confirm that $\sigma(\tau)$ becomes a function of $\sigma(0)$ and $\tilde{c}_{r'}(0)$ and their conjugates, where the space coordinate r' lies in the $-\tau < r' < 0$ region. Therefore, $\sigma(\tau)$ commutes with the input operator $\tilde{c}_r(0)$ satisfying $r < -\tau$.

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