

Supplementary information: *Generating spatially separated correlated multiphoton states in nonlinear waveguide quantum electrodynamics*

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I. DERIVATIONS IN MOMENTUM SPACE

In this section, we derive the scattering state in momentum space, which is formally equivalent to the real-space treatment. Applying a Fourier transform to convert the photon position coordinate x into the wave vector k , the interaction Hamiltonian is

$$H_{\text{int}} = \sum_K \sum_k g M^*(K, k, 0) D_K^\dagger \sigma_- a_k + \text{H.c.}, \quad (1)$$

$$M(K, k, 0) = \frac{\sqrt{2}}{N} \sum_m e^{-ikm} e^{iKm/2} u_0 e^{-|m|/L_u(K)}, \quad (2)$$

and the scattering-state solution has the form

$$|\Psi(t)\rangle = \left[\sum_k c_{e,k}(t) \sigma_+ a_k^\dagger + \sum_K c_K(t) D_K^\dagger \right] |\text{vac}\rangle, \quad (3)$$

where $c_{e,k}(t)$ denotes both the emitter and the single-photon state k being excited. For notational simplicity, we suppress the emitter label, so that $c_{e,k} \rightarrow c_k$.

From the secular equation, we obtain the following system of equations:

$$(\omega_k + \Delta_e) c_k + g \sum_K M(K, k, n_1) c_K = (\omega_{k_0} + \Delta_e) c_k, \quad (4)$$

$$E_K c_K + g \sum_k M^*(K, k, n_1) c_k = (\omega_{k_0} + \Delta_e) c_K. \quad (5)$$

Note that these equations describe *the coupling between two momentum modes k and K* . They can be rearranged into the form

$$c_k = \delta_{k,k_0} + g \sum_K \frac{M(K, k, n_1) c_K}{\omega_{k_0} - \omega_k + i0^+}, \quad (6)$$

$$c_K = \frac{g \sum_k M^*(K, k, n_1) c_k}{E_{K_r} - E_K + i0^+}, \quad (7)$$

where δ_{k,k_0} indicates that the initial state is a right-going single photon.

By defining

$$C_0(m) = \sum_k e^{ikm} c_k, \quad (8)$$

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we obtain a complex convolution equation

$$C_0(m_0) = e^{ik_0 m_0} - \frac{1}{v_{k_0} v_{K_r}} \times \sum_{m_1, m_2} g_{m_1} g_{m_2} e^{iK_r |m_1 - m_2|/2} e^{ik_0 |m_0 - m_1|} C_0(m_2), \quad (9)$$

where

$$g_m = \sqrt{2} u_0 g e^{-|m|/L_u(K_r)} \quad (10)$$

denotes the effective coupling strength per pseudo-coupling point of the PGA. The first term in Eq. (9), $\exp(ik_0 m_0)$, corresponds to the free propagation of the incident wave, while the second term captures the scattering disturbance due to the nonlocal potential. The convolution arises naturally from this nonlocality: the output $C_0(m_0)$ depends not only on the input at position m_0 , but also on the responses $C_0(m_2)$ at neighboring sites. This formulation describes multiple scattering events and reflects interference among different scattering channels.

Following the approach in main text, we find that the final state takes the form

$$|f_i\rangle = |\Psi(t \rightarrow \infty)\rangle \simeq \left[\sum_{\pm k} c_{e,\pm k} (+\infty) \sigma_+ a_{\pm k}^\dagger + \sum_{\pm K} u_{\pm K} (+\infty) D_{\pm K}^\dagger \right] |\text{vac}\rangle, \quad (11)$$

where the momentum components are explicitly separated into positive and negative branches to distinguish the right- and left-going single-photon waves and the generation of doublon waves. After obtaining the scattering state for a single-photon plane-wave input, we now return to L-S equation to derive the output state by projecting the final state onto the subspaces spanned by $|e, k\rangle$ and $|K\rangle$, respectively.

A. Doublon component

We substitute the scattering state from Eq. (3) and the interaction Hamiltonian from Eq. (1) into L-S equation, and project onto the right-going doublon state $|+K\rangle$, yielding

$$\begin{aligned} \langle +K|f\rangle &= \langle +K|\Psi_{\text{sc}}\rangle - \langle +K| \frac{1}{E_{K_r} - H_0 - i0^+} H_{\text{int}} |\Psi_{\text{sc}}\rangle \\ &= \sum_{K'} \langle +K| \frac{g \sum_k M^*(K', k, n_1) c_k}{E_{K_r} - E_{K'} + i0^+} |K'\rangle - \sum_{K'} \langle +K| \frac{g \sum_k M^*(K', k, n_1) c_k}{E_{K_r} - E_{K'} - i0^+} |K'\rangle. \end{aligned} \quad (12)$$

Using the expression for M in Eq. (2), the definition of $C_0(m)$ in Eq. (8), and evaluating the integrals via the retarded and advanced Green's functions, we simplify Eq. (12) to

$$\begin{aligned} \langle +K|f\rangle &= \langle +K_r|f\rangle \\ &= -i \frac{1}{v_{K_r}} \sum_m g_m e^{-iK_r m/2} [\Theta(-m) + \Theta(+m)] C_0(m) = -i \frac{1}{v_{K_r}} \sum_m g_m e^{-iK_r m/2} C_0(m). \end{aligned} \quad (13)$$

Here, the retarded and advanced Green's functions, corresponding to the first and second terms in Eq. (12), contribute $\Theta(-m)$ and $\Theta(+m)$, respectively. This temporal and spatial structure reflects the full scattering history: the initial state evolves into the scattering state under the retarded Green's function from $t = -\infty$ to $t = 0$, covering the negative spatial part of the nonlocal potential. The final state evolves back under the advanced Green's function from $t = +\infty$ to $t = 0$, accounting for the positive spatial part. Together, these contributions encode the complete scattering process across both time ($t = -\infty \rightarrow t = 0 \rightarrow t = +\infty$) and space ($x = -\infty \rightarrow x = 0 \rightarrow x = +\infty$), as captured by the combination $\Theta(-x) + \Theta(+x)$. Thus, the doublon components of the final state are

$$u_{+K_r} = \langle +K_r|f\rangle = -i \frac{1}{v_{K_r}} \sum_m g_m e^{-iK_r m/2} C_0(m), \quad (14)$$

$$u_{-K_r} = \langle -K_r|f\rangle = -i \frac{1}{v_{K_r}} \sum_m g_m e^{+iK_r m/2} C_0(m). \quad (15)$$

B. Single-photon component

We now project the final state onto the right-going single-photon state $|+k, e\rangle$ (suppressing the emitter label, we denote this state as $|k\rangle$):

$$\begin{aligned}\langle +k|f\rangle &= \langle +k|\Psi_{\text{sc}}\rangle - \langle +k|\frac{1}{E - H_0 - i0^+}H_{\text{int}}|\Psi_{\text{sc}}\rangle \\ &= \langle +k|+k_0\rangle + \sum_k \langle +k|\frac{g\sum_K M(K, k, n_1)c_K}{\omega_{k_0} - \omega_k + i0^+}|k\rangle - \sum_k \langle +k|\frac{g\sum_K M(K, k, n_1)c_K}{\omega_{k_0} - \omega_k - i0^+}|k\rangle.\end{aligned}\quad (16)$$

Here, the first term corresponds to the initial state [i.e., the δ_{k,k_0} in Eq. (6)].

The Green's function integrals over k are

$$\frac{1}{N}\sum_k \frac{e^{-ikm/2}}{\omega_{k_0} - \omega_k + i0^+}|k\rangle = -\frac{i}{v_k}\left[e^{-ik_0m/2}\Theta(-m)|+k_0\rangle + e^{ik_0m/2}\Theta(+m)|-k_0\rangle\right],\quad (17)$$

$$\frac{1}{N}\sum_k \frac{e^{-ikm/2}}{\omega_{k_0} - \omega_k - i0^+}|k\rangle = +i\frac{1}{v_k}\left[e^{-ik_0m/2}\Theta(+m)|+k_0\rangle + e^{ik_0m/2}\Theta(-m)|-k_0\rangle\right].\quad (18)$$

Thus, Eq. (16) is

$$\langle +k_0|f\rangle = 1 + \sum_{m_1} \frac{-i}{v_k} g_{m_1} e^{-ik_0m_1} \sum_K e^{iK m_1/2} c_K.\quad (19)$$

The summation over K can be evaluated using Eq. (7):

$$\sum_K e^{iK m_1/2} c_K = \sum_{m_2} \frac{-i}{v_{K_r}} e^{iK_r|m_1-m_2|/2} g_{m_2} C_0(m_2).\quad (20)$$

The final state amplitudes for the single-photon components are then

$$t = \langle +k_0, e|f\rangle = 1 - \frac{1}{v_{k_0}v_{K_r}} \sum_{m_1, m_2} g_{m_1} g_{m_2} e^{-ik_0m_1} e^{iK_r|m_1-m_2|/2} C_0(m_2),\quad (21)$$

$$r = \langle -k_0, e|f\rangle = -\frac{1}{v_{k_0}v_{K_r}} \sum_{m_1, m_2} g_{m_1} g_{m_2} e^{ik_0m_1} e^{iK_r|m_1-m_2|/2} C_0(m_2).\quad (22)$$

Ultimately, the final outgoing state, given an initial incident single-photon plane wave $\exp(ik_0x)$, is

$$|i\rangle = |e, +k_0\rangle,\quad (23)$$

$$|f_i\rangle = t|e, +k_0\rangle + r|e, -k_0\rangle + u_{\pm K_r}|\pm K_r\rangle,\quad (24)$$

where t and r denote the transmission and reflection amplitudes, respectively, of the single photon, and $u_{\pm K_r}$ is the probability amplitudes for the generated right- and left-going doublon modes. Note that, although the incident state only consists of a single-photon plane wave, the scattered state contains not only the reflection r and transmission t single-photon components, but also the right- and left-going doublon waves $|\pm K_r\rangle$. The final results can be obtained numerically by solving the complex convolution equation [Eq. (9)] for $C_0(m)$, and then substituting into Eqs. (14), (15), (21), and (22).

II. DERIVATION OF EQS. (37) AND (38)

In this section, we provide the intermediate equations from Eq. (28) to Eq. (29). First, the incident energy is

$$H|\Psi_i\rangle = (v_k k + \Delta_e)|\Psi_i\rangle.\quad (25)$$

The free terms are

$$H_0|\Psi_{\text{sc}}\rangle = \int dx \left[\Delta_e \varphi_R(x) - i v_k \frac{\partial}{\partial x} \varphi_R(x) \right] |e, x_R\rangle + \int dx \left[\Delta_e \varphi_L(x) + i v_k \frac{\partial}{\partial x} \varphi_L(x) \right] |e, x_L\rangle + \sum_K E_K u_K |g, K\rangle.\quad (26)$$

The interaction terms are

$$\begin{aligned}
H_{\text{int}}|\Psi_{sc}\rangle &= \sum_{K,n} gM^*(K,n,0) \int dx \{ \delta(x-n) [\varphi_R(x) + \varphi_L(x)] \} |g,K\rangle \\
&+ \sum_{K,n} gM(K,n,0) u_K \int dx \{ \delta(x-n) (|e,x_R\rangle + |e,x_L\rangle) \}.
\end{aligned} \tag{27}$$

Substituting into the secular equation $H|\Psi_{sc}\rangle = E|\Psi_{sc}\rangle$, we obtain the following set of coupled equations

$$Eu_K|g,K\rangle = \left[E_K u_K + \sum_n gM^*(K,n,0) \int dx \{ \delta(x-n) [\varphi_R(x) + \varphi_L(x)] \} \right] |g,K\rangle, \tag{28a}$$

$$E\varphi_R(x)|e,x_R\rangle = \left[\Delta_e \varphi_R(x) - iv_k \frac{\partial}{\partial x} \varphi_R(x) + \sum_{K,n} gM(K,n,0) u_K \delta(x-n) \right] |e,x_R\rangle, \tag{28b}$$

$$E\varphi_L(x)|e,x_L\rangle = \left[\Delta_e \varphi_L(x) + iv_k \frac{\partial}{\partial x} \varphi_L(x) + \sum_{K,n} gM(K,n,0) u_K \delta(x-n) \right] |e,x_L\rangle. \tag{28c}$$

To simplify these equations, we use the relations

$$\int dx \delta(x-n) \varphi_R(x) = \varphi_R(n) = e^{ikn} (R_n + R_{n+1})/2, \tag{29}$$

$$\int dx \delta(x-n) \varphi_L(x) = \varphi_L(n) = e^{-ikn} (L_n + L_{n+1})/2, \tag{30}$$

$$\frac{\partial}{\partial x} \varphi_R(x) = ik\varphi_R(x) + \sum_{n=-N+1}^{N-1} e^{ikn} (R_{n+1} - R_n) \delta(x-n), \tag{31}$$

$$\frac{\partial}{\partial x} [-\varphi_L(x)] = ik\varphi_L(x) + \sum_{n=-N+1}^{N-1} e^{-ikn} (L_n - L_{n+1}) \delta(x-n). \tag{32}$$

Inserting these into Eq. (28) yields

$$(E_{K_r} - E_K) u_K = \sum_n gM^*(K,n,0) [e^{ikn} (R_n + R_{n+1}) + e^{-ikn} (L_n + L_{n+1})]/2, \tag{33}$$

$$iv_k e^{ikn} (R_{n+1} - R_n) = \sum_K gM(K,n,0) u_K, \quad iv_k e^{-ikn} (L_n - L_{n+1}) = \sum_K gM(K,n,0) u_K. \tag{34}$$

III. WAVE-PACKET SHAPE

Different from stimulated emission and resonant scattering, the far-detuned TLE does not resonantly exchange photons (i.e., no Wigner delay), leading to two distinct results.

First, the scattering process does not distort the wavepacket profile: the single-photon component retains its original shape after scattering, as shown in Fig. 1. In the numerical simulation, the finite wavepacket bandwidth induces an interaction duration given by $t_{\text{int}} = W_S/v_k$. Correspondingly, the generated doublon wavepacket has a bandwidth of $W_D = v_K t_{\text{int}} = W_S v_K/v_k$. Figure 1 displays the photonic field at $Jt = 600$, showing a clear spatial separation between the single-photon and two-photon components. After scattering, the single-photon and doublon output states *inherit the spatial profile of the incident wave packet*, centered at $n_S = v_{k_0}(t - t_0)$ and $n_D = v_{K_r}(t - t_0)$, with amplitudes given by $|t|^2$, $|r|^2$, and $|u_{\pm}|^2 u_{K_r}(r)$, respectively. One can more clearly observe that the two-photon field exhibits strong spatial correlations, featuring a sharp peak at $r_c = 0$ and rapid decay.

Second, the scattering dynamics becomes insensitive to the bandwidth and shape of the incident wavepacket. We can perform numerical simulations using a Gaussian wavepacket with a broader bandwidth of $L_G = 0.02$ (in momentum space), and a Lorentzian wavepacket. The initial state of the Lorentzian wavepacket in momentum and real space is

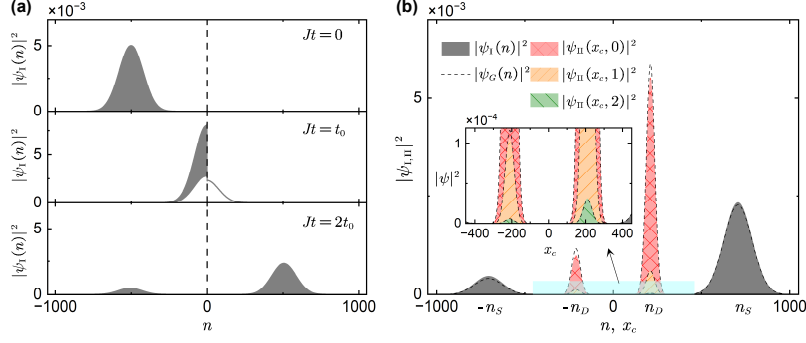


FIG. 1. Photon distribution fields. (a) Single-photon field $|\psi_I(n)|^2$ at different times: $Jt = 0$ (incident single-photon wavepacket), $Jt = t_0$ (t_0 is given in Fig. 4 in the main text), and $Jt = 2t_0$ where the reflected single-photon wave packet returns to its initial position. The dashed line indicates the emitter position $n_1 = 0$. (b) Spatial distribution of the single-photon field $|\psi_I(n)|^2$ and the two-photon field $|\psi_{II}(x_c, r_c)|^2$ at $Jt = 600$ from Fig. 4 in the main text. Black solid curves represent analytical Gaussian wave packets, with spectral width L_G for the single photons and $L_G v_{K_r}/v_{k_0}$ for the doublons. n_S and n_D mark the positions of the single-photon and doublon wavepackets, respectively. Amplitudes are $|t|^2$, $|r|^2$ and $|u_{\pm}|^2 \times u(r_c)$ for the different components of the photon field.

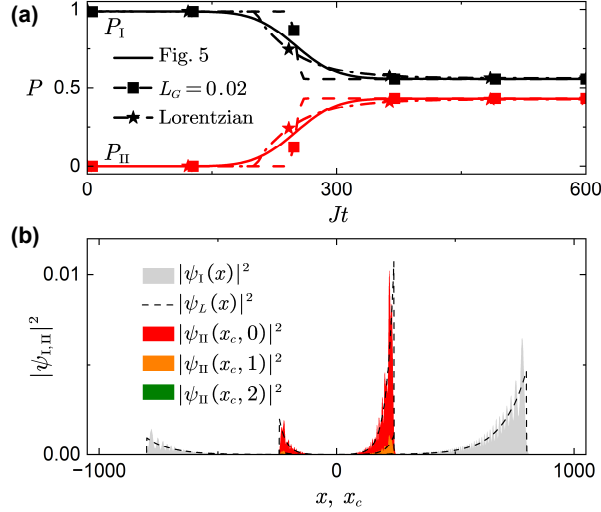


FIG. 2. Scattering results for a Gaussian wavepacket with a broader bandwidth of $L_G = 0.02$, and a Lorentzian wavepacket with $L_L = 0.005$. (b) The scattering field at $Jt = 600$. The other parameters are the same as in Fig. (5) in the main text.

then given by

$$\begin{aligned}\psi_L(k) &= \sqrt{\frac{L_L}{\pi}} \frac{1}{L_L - i(k - k_0)}, \\ \psi_L(n) &= \sqrt{2L_L} e^{ik_0 n} e^{nL_L} \Theta(-n_0).\end{aligned}\tag{35}$$

Here, L_L denotes the spatial spread of the Lorentzian wave packet. Using the same simulation parameters as for the Gaussian wave packet discussed in the main text, we present the results in Fig. 2. After scattering, the scattering results remains consistent with those shown in Fig. 1 in the main text, and the generated doublon wavepacket retains the profile of the incident photon wavepacket, albeit with a modified bandwidth $W_D = (v_K/v_k)W_S$.

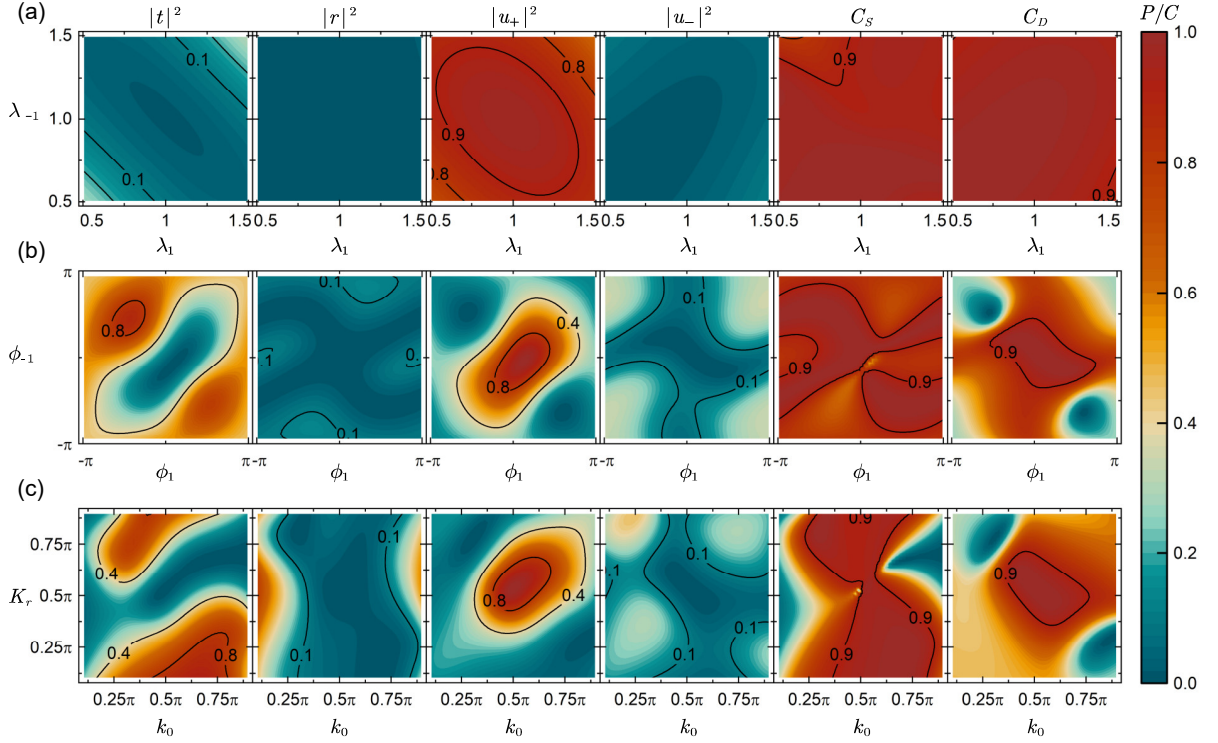


FIG. 3. Influence of the coupling parameters of the giant atom on the system dynamics. The single-photon transmission and reflection, the right- and left-going doublon populations, and the single-photon and doublon directional factors are shown as functions of: (a) the coupling strength ratio $\lambda_{\pm 1} = g^{(\pm 1)}/g^{(0)}$; (b) the coupling phases $\phi_{\pm 1}$ of the two outer coupling points; and (c) the resonant modes k_r, K . Different contours are used for clarity. The parameters of the central coupling point are fixed as a reference, with $g^{(0)} = 0.31J$ and $\phi_0 = 0$, corresponding to the optimal point in Fig. 8 of the main text. The other parameters are: (a) $\lambda_{\pm 1} = 1$, $k_0 = K_r = \pi/2$; (b) $\phi_{\pm 1} = \pm 0.05\pi$, $k_r = K_0 = \pi/2$; (c) $\lambda_{\pm 1} = 1$, $\phi_{\pm 1} = \pm 0.05\pi$.

IV. GIANT-ATOM PARAMETERS

We first generalize the interaction Hamiltonian to account for an emitter coupled to the waveguide at N sites, each with a coupling strength $g^{(\tau)}$ and phase ϕ_τ

$$H_{\text{int}} = \sigma_- \sum_{\tau} g^{(\tau)} e^{i\phi_\tau} a_{n_\tau}^\dagger + \text{H.c.}, \quad (36)$$

where τ labels the coupling leg, and $g^{(\tau)}$, ϕ_τ , and n_τ denote the coupling strength, phase, and position of the τ -th leg, respectively. By substituting this giant-atom interaction into the Lippmann-Schwinger equation, we can derive the giant-atom versions of expressions for the single-photon and doublon components of the final state. The transmission t^{Giant} , reflection r^{Giant} , and generated doublon amplitudes $u_{\pm K_r}^{\text{Giant}}$ are given by

$$t^{\text{Giant}} = 1 - \frac{1}{v_{k_0} v_{K_r}} \sum_{\tau_1, m_1} g_{|m_1 - \tau_1|}^{(\tau_1)} e^{i\phi_{\tau_1}} e^{-ik_0 m_1} C_0^K(\tau_1, m_1), \quad (37)$$

$$r^{\text{Giant}} = -\frac{1}{v_{k_0} v_{K_r}} \sum_{\tau_1, m_1} g_{|m_1 - \tau_1|}^{(\tau_1)} e^{i\phi_{\tau_1}} e^{+ik_0 m_1} C_0^K(\tau_1, m_1), \quad (38)$$

$$u_{\pm K_r}^{\text{Giant}} = -\frac{i}{v_{K_r}} \sum_{\tau, m} g_{\tau, |m - \tau|}^{(\tau)} e^{i\phi_\tau} e^{\mp i K_r (m + \tau)/2} C_0^{\text{Giant}}(m). \quad (39)$$

The central quantities C_0 obey the following set of coupled convolution equations:

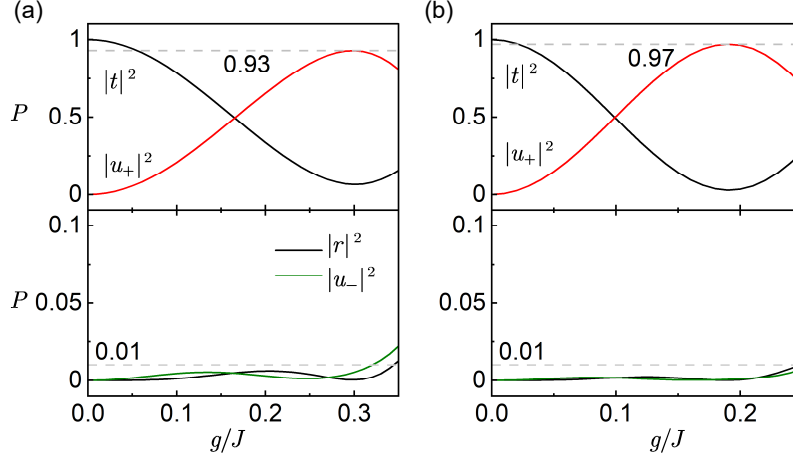


FIG. 4. Scattering results as functions of the coupling strength g , for three coupling points $\{n_{-1}, n_0, n_1\}$, and five coupling points $\{n_{-2}, n_{-1}, n_0, n_1, n_2\}$. The coupling strength is uniform across all legs, and the coupling phase is set to zero. All other parameters are the same as in Fig. (8) in the main text.

$$C_0^K(\tau_1, m_1) = -\frac{i}{v_{K_r}} \sum_{\tau_2, m_2} g_{|m_2 - \tau_2|}^{(\tau_2)} e^{i\phi_{\tau_2}} e^{iK_r|m_1 + \tau_1 - m_2 - \tau_2|/2} C_0(m_2), \quad (40)$$

$$C_0^{\text{Giant}}(m_0) = e^{ik_0 m_0} - \frac{1}{v_{k_0} v_{K_r}} \sum_{\tau_1, m_1} \sum_{\tau_2, m_2} g_{|m_1 - \tau_1|}^{(\tau_1)} e^{i\phi_{\tau_1}} g_{|m_2 - \tau_2|}^{(\tau_2)} e^{i\phi_{\tau_2}} e^{ik_0|m_0 - m_1|} e^{iK_r|m_1 + \tau_1 - m_2 - \tau_2|/2} C_0^{\text{Giant}}(m_2). \quad (41)$$

Here, $g_{|m - \tau|}^{(\tau)} = \sqrt{2}g^{(\tau)}u_0 e^{-\frac{|m - n_\tau|}{L_{K_r}}}$.

Compared to the small-atom version in Eq. (9), this formalism introduces more parameters, including the number of coupling points, their position n_τ , strength g^τ , and phases ϕ_τ . Due to the exponential decay of the scattering potential, the range of the PGA can be truncated to $m = 0, \pm 1, \pm 2, \pm 3$. By numerically solving these equations for a given parameter set $\{n_\tau, g_\tau, \phi_\tau\}$, one can obtain the $C_0^{\text{Giant}}(m)$, and thus the final state, t^{Giant} , r^{Giant} , and $u_{\pm K_r}^{\text{Giant}}$.

The presence of this multi-variable convolution structure makes analytical solutions difficult. Therefore, the design problem naturally transforms into a numerical optimization problem. One can employ optimization algorithms to search the parameters space $\{n_\tau, g_\tau, \phi_\tau\}$, which realizes higher efficiency. Using this method, we can obtain other parameter sets, but not limited to, the patterns shown in the main text.

A. Influence of parameter deviations from the optimal point

Starting from the optimal working point in Fig. (8) of the main text, we analyze how deviations of the system parameters affect the scattering dynamics. In our model, the RGA couples to the waveguide at three points $-1, 0, 1$ with frequency detuning Δ_e . The coupling at each point is denoted by a strength $g^{(-1,0,1)}$ and an encoded phase $\phi_{-1,0,1}$. We set the parameters of the central coupling point fixed as a reference: $g^{(0)} = 0.31J$ and $\phi_0 = 0$. In Fig. 3, we present the scattering results ($|t|^2, |r|^2, |u_+|^2, |u_-|^2$) and directional factors C_S, C_D as functions of the varied parameters.

First, we examine the effect of varying the coupling strengths at the outer coupling points, parametrized by the ratio $\lambda_{\pm 1} = g^{(\pm 1)}/g^{(0)}$, as shown in Fig. 3(a). The results indicate that the system maintains over 90% conversion efficiency and preserves good directionality across the displayed range of $\lambda_{\pm 1}$.

Next, in Fig. 3(b), we investigate the dependence on the encoded coupling phases $\phi_{\pm 1}$. High conversion efficiency and directionality are retained only within a central region around the optimal phase values.

Finally, in Fig. 3(c), we consider the effect of the frequency detuning Δ_e . Under the resonant condition $\Delta_e + E_{k_0} = E_{K_r}$, deviations in Δ_e can be interpreted as shifts in the resonant modes k_0 (incident photon) and K_r (excited doublon). The resulting behavior is similar to that observed for the coupling phases.

These results demonstrate that the scattering process is insensitive to variations in coupling strength, maintaining high efficiency and directionality despite deviations. In contrast, due to the interference of the giant atom config-

uration, the system is sensitive to the phase-related parameters, including the encoded coupling phases ϕ and the propagation phases associated with resonant modes. However, this sensitivity manifests as a gradual trend rather than a sharp change, suggesting that the proposed scheme remains feasible under realistic experimental conditions. When parameters deviate from the optimal point, appropriate adjustment can be employed to compensate for these changes and restore performance.

B. Giant atom coupling without coupling phases

In Fig. 4, we show that the RGA enables a theoretical maximum efficiency of 93% for three coupling points and 97% for five coupling points. Note that, without coupling phases, when the coupling strength is small compared to the optimal value, the system supports a well-defined unidirectional scattering process with negligible single-photon backscattering, and the left-going doublon population remains below 1%.
