Supplemental Material for "Resonant enhancement of three-body loss between strongly interacting photons"

Marcin Kalinowski,[1,2,*] Yidan Wang,[1,*] Przemyslaw Bienias,[1,3] Michael J. Gullans,[1,3,4] D. P. Ornelas-Huerta,[1] Alexander N. Craddock,[1] Steven L. Rolston,[1] J. V. Porto,[1] Hans Peter Büchler,[5] and Alexey V. Gorshkov[1,3]

[1]*Joint Quantum Institute, NIST/University of Maryland, College Park, Maryland 20742 USA*
[2]*Faculty of Physics, University of Warsaw, Pasteura 5, 02-093 Warsaw, Poland*
[3]*Joint Center for Quantum Information and Computer Science, NIST/University of Maryland, College Park, Maryland 20742 USA*
[4]*Department of Physics, Princeton University, Princeton, New Jersey 08544 USA*
[5]*Institute for Theoretical Physics III and Center for Integrated Quantum Science and Technology, University of Stuttgart, 70550 Stuttgart, Germany*

In this Supplemental Material, we provide detailed calculations used to obtain results mentioned in the main text. This covers the in-depth introduction of the microscopic model and related quantities (Sec. I), the solution to the two-body problem (Sec. II), the solution of three-body problem (Sec. III), and the description of our effective transmission model (Sec. IV).

I. RUNNING-WAVE CAVITY HAMILTONIAN

In this section, we introduce the details of the microscopic model used in this work.

We consider a running-wave optical cavity in one dimension, supporting a single photonic mode. Moreover, we assume a constant density of the atomic cloud, so that the system is translationally invariant. The cavity mode with the profile $u_0(z)$ is created by the operator $a^\dagger$. While there is only one photonic mode present, the atomic medium can support a broad range of excitations, which are captured by introducing additional mode functions $u_{q\neq 0}(z)$. Together, $\{u_0, u_{q\neq 0}\}$ form an orthonormal basis and can be used to express various field operators

$$E^\dagger(z) = u_0^*(z)a^\dagger, \qquad P_q^\dagger = \int dz\, u_q(z)P^\dagger(z), \qquad S_q^\dagger = \int dz\, u_q(z)e^{ik_c z}S^\dagger(z), \tag{S1}$$

where $E^\dagger(z)$ creates the cavity photon at position z, while $P^\dagger(z)$ and $S^\dagger(z)$ create an excitation of the medium at position z to the atomic state $|p\rangle$ ans $|s\rangle$, respectively. Thanks to the translational symmetry, we can identify index q with momentum and write the explicit form of these mode functions: $u_q(z) = \frac{1}{\sqrt{L}}e^{iqz}$. This way, the momentum $q = 0$ corresponds to the cavity photon in the rotating frame. The Hamiltonian for such a system is

$$H_{\text{cav}} = \underbrace{\begin{pmatrix} a \\ P_0 \\ S_0 \end{pmatrix}^\dagger \begin{pmatrix} \delta_r & g & 0 \\ g & \Delta & \Omega \\ 0 & \Omega & -i\gamma_s \end{pmatrix} \begin{pmatrix} a \\ P_0 \\ S_0 \end{pmatrix}}_{\text{Photon mode}} + \underbrace{\sum_{q\neq 0} \begin{pmatrix} P_q \\ S_q \end{pmatrix}^\dagger \begin{pmatrix} \Delta & \Omega \\ \Omega & -i\gamma_s \end{pmatrix} \begin{pmatrix} P_q \\ S_q \end{pmatrix}}_{\text{Spin waves}} + H_{\text{int}}, \tag{S2}$$

where $\Delta = \delta - i\gamma$ is the complex detuning of the classical field with Rabi frequency Ω, $2\gamma_s$ is the decay rate of the Rydberg state, and δ_r is the (two-photon) detuning from the EIT resonance. Note the difference in form from Eq. (1) in the main text: we write Eq. (S2) in a momentum basis, whereas Eq. (1) is in a real-space basis. The real-space representation is convenient for presentation because it naturally generalizes to other cavity geometries. The interaction Hamiltonian H_{int} is

$$H_{\text{int}} = \frac{1}{2}\int dz dz' S^\dagger(z)S^\dagger(z')V(z-z')S(z')S(z') = \frac{1}{2}\sum_{q_1,q_2} S^\dagger_{q_1}S^\dagger_{-q_1}S_{q_2}S_{-q_2}\int \frac{dr}{L}V(r)e^{i(q_1-q_2)r}, \tag{S3}$$

where second equality holds for vanishing total momentum $K = 0$, which is the case we consider here (as we will explain in Sec. III, only the $K = 0$ solution to the two-body problem will be necessary for our solution to the three-body problem). We assume $\Delta = \delta$ to be real throughout all derivations. Then, we analytically continue the result to the complex case $\Delta = \delta - i\gamma$. Throughout all derivations, we also consider the situation where the photon field is on the EIT resonance ($\delta_r = 0$) and the Rydberg state decay rate is negligible ($\gamma_s = 0$).

Spectral decomposition of the spin-wave part of the Hamiltonian (S2) gives energies $\epsilon_\pm$ of the upper/lower spin wave and their respective overlaps $\alpha_\pm$ with the $|s\rangle$ state:

$$\epsilon_\pm = \frac{1}{2}\left(\Delta \pm \sqrt{\Delta^2 + 4\,\Omega^2}\right), \qquad \alpha_\pm = \frac{1}{2}\left(1 \pm \frac{\Delta}{\sqrt{\Delta^2 + 4\,\Omega^2}}\right). \tag{S4}$$

In the coordinate space, the single-particle propagator ($\hat{g}_s$ in the main text) is

$$g_s(\omega; x, x') = \underbrace{\sum_{\mu=\pm} \frac{\alpha_\mu}{\omega - \epsilon_\mu + i0^+}}_{\eta(\omega)} \sum_{q\neq 0} e^{iq(x-x')} + \frac{1}{\omega + i0^+}, \tag{S5}$$

where the first term corresponds to the spin-wave excitation and the second to the dark-state polariton. In the large-coupling limit $g \gg \Omega, |\Delta|$, only the dark branch of polaritons contributes because the energy of bright polaritons is proportional to $g \to \infty$, so their effect can be neglected – see Ref. [S1] for a more detailed discussion. In particular, we remark that we can leave g large, but finite, and take the limit $g \to \infty$ at the end of the calculation. However, keeping a finite g significantly obscures the calculation so we first take the limit of infinite g to simplify the presentation. The energy-dependent factor $\eta(\omega)$ can be evaluated to

$$\eta(\omega) = \frac{\omega - \Delta}{\omega^2 - (\Omega^2 + \Delta\,\omega)}, \tag{S6}$$

and one needs to use the full expression with $i0^+$ included if integration occurs.

II. TWO-BODY PROBLEM

In this section, we introduce notation important for our scattering analysis and solve the two-body problem.

To gain insight into the influence of the additional spin-wave branch, we begin by studying the two-body problem. Description of the two-body processes is contained in the off-shell two-body T-matrix $T_2(\omega)$. Here, we derive Eq. (3) from the main text. An equation for $T_2(\omega)$ can be written explicitly using a supporting definition $T_2(\omega) = \int \frac{dr}{L} T_2(\omega; r)$, where $T_2(\omega; r) = \int \frac{d\vec{x}'}{L^2} \frac{dR}{L} T_2(\omega; \vec{x}, \vec{x}')$, $\vec{x} = (x_1, x_2)$, $\vec{x}' = (x_1', x_2')$, $r = x_1 - x_2$ is the relative coordinate, and $R = (x_1 + x_2)/2$ is the center of mass coordinate. $T_2(\omega; r)$ is given by

$$T_2(\omega; r) = V(r) + \int \frac{dr'}{L} V(r) G_2(\omega; r, r') T_2(\omega; r'), \tag{S7}$$

where the two-body propagator G_2 can be obtained from the Hamiltonian (S2) and is given by

$$G_2(\omega; r, r') = (\omega + i0^+)^{-1} + \chi(\omega) \sum_{q\neq 0} e^{iq(r-r')}, \tag{S8}$$

where the zero-momentum term $(\omega + i0^+)^{-1}$ is the propagator of two dark-state polaritons. It can be related to our formulation using $\hat{g}_s$ via

$$G_2(\omega; r, r') \overset{!}{=} \int \frac{d\omega'}{2\pi i} g_s(\omega'; x_1, x_1') g_s(\omega - \omega'; x_2, x_2'), \tag{S9}$$

where $\overset{!}{=}$ means that it is equal only under the integral in Eq. (S7) and after enforcing the total-momentum conservation. The coefficient $\chi(\omega)$, in the part of G_2 corresponding to the double excitation of spin waves, is

$$\chi(\omega) = \frac{\Delta - \frac{\omega}{2} - \frac{\Omega^2}{\Delta - \omega}}{\omega\left(\Delta - \frac{\omega}{2}\right) + 2\Omega^2}, \tag{S10}$$

which coincides with the two-body propagator in free space in the infinite-momentum limit [S1]. Equation (S7) is represented schematically in Fig. 2(a) of the main text. To solve it, we rewrite the propagator from Eq. (S8) as

$$G_2(r, r') = [(\omega + i0^+)^{-1} - \chi(\omega)] + L\chi(\omega)\,\delta(r - r') \tag{S11}$$

and plug it back into Eq. (S7). After rearrangement and integration of both sides by $\int \frac{dr}{L}$, we obtain the equation for $T_2(\omega)$, which gives

$$T_2(\omega) = U_2(\omega) + U_2(\omega)[(\omega + i0^+)^{-1} - \chi(\omega)] T_2(\omega), \tag{S12}$$

where $U_2(\omega)$ is a well-known [S1] renormalized two-body interaction (for $g \to \infty$) between dark-state polaritons, presented in Eq. (2) of the main text. In the limit of large separation, it reduces to the bare van der Waals potential. Conversely, for small distances, it saturates at a finite value, an effect caused by the so-called Rydberg blockade mechanism.

Solving algebraic equation (S12) for $T_2(\omega)$ reproduces Eq. (3) from the main text. Notice that, to the leading order in r_b/L, equation (S12) describes the scattering of two infinitely heavy particles under the influence of potential $U_2(\omega; r)$. Additional terms encapsulate effects specific to the cavity setup.

III. THREE-BODY PROBLEM

In this section, we investigate the scattering of three dark-state polaritons relying heavily on definitions and results from the previous two sections. Schematic representation of the key equations is presented in Fig. 2(b-c) of the main text.

As in the two-body case, the influence of the interaction on the physics of the three-body problem is captured by the analytical structure of the T-matrix $\boldsymbol{T_3}$. Specifically, the energy shift corresponds to the pole of the integrated T-matrix $T_3(\omega)$.

To calculate this object, one can in principle employ the Schrödinger equation. However, such a treatment can lead to spurious, nonphysical solutions. In order to avoid this issue, another approach to the quantum three-body problem was developed by Faddeev [S2]. In this formulation, all scattering processes are grouped depending on which two particles interact first. This way one introduces a rigorous method for expressing the three-body scattering as a series of two-body processes (a three-body force can also be included). As a consequence, the three-body T-matrix can now be written as a sum of three (sub)T-matrices

$$\hat{T}_3(\omega) = \frac{1}{3}\sum_{i<j}\hat{T}_3^{ij}(\omega, \epsilon_k), \tag{S13}$$

where $\hat{T}_3^{ij}(\omega;\epsilon_k)$ denotes the T-matrix for the group of processes, where particles labeled i and j interacted first and the third particle $k \neq i, j$ has energy ϵ_k. These $\hat{T}_3^{ij}(\omega, \epsilon_k)$ objects are coupled to each other by the set of equations called Faddeev equations. In our case, the situation is further complicated by the multi-component nature of the polariton system. Let us define $\hat{T}_3^{ij}(\omega) \equiv \hat{T}_3^{ij}(\omega; 0)$ and $\hat{T}_\mu^{ij}(\omega) \equiv \hat{T}_3^{ij}(\omega; \epsilon_{\mu=\pm})$.

Every T-matrix we consider has all three outgoing DSPs. In our system, exact Faddeev equations [S2] describing the off-shell scattering of three zero-momentum DSPs are

$$\begin{aligned} T_3^{12}(\omega) &= T_2(\omega)\frac{2}{\omega+i0^+}T_2(\omega) + T_2(\omega)\frac{1}{\omega+i0^+}\left(T_3^{13}(\omega)+T_3^{23}(\omega)\right) \\ &+ \sum_{\mu=\pm}\int\frac{dr_{12}}{L}\frac{dr_3}{L}T_2(\omega;r_{12})\,\alpha_\mu\eta_\mu\left(T_\mu^{13}(\omega;\vec{x})+T_\mu^{23}(\omega;\vec{x})\right) - T_2(\omega)\,\alpha_\mu\eta_\mu\left(T_\mu^{13}(\omega)+T_\mu^{23}(\omega)\right) \end{aligned} \tag{S14}$$

and the two analogous equations for $T_3^{13}(\omega)$ and $T_3^{23}(\omega)$ obtained through permutation of indices. Here $T_\mu^{ij}(\omega;\vec{x}) = \int \frac{d\vec{x}'}{L^3}\frac{dR}{L}T_\mu^{ij}(\omega;\vec{x},\vec{x}')$ is the T-matrix for the process with the third incoming leg being a spin wave belonging to the branch μ. Variable r_{12} denotes the distance between particles labeled 1 and 2, $r_3 = (x_1+x_2)/2 - x_3$ is the standard third Jacobi coordinate, and $\eta_\mu = \eta(\omega - \epsilon_\mu)$.

Eq. (S14) is a direct implementation of the operator equation [Eq. (5)] from the main text. Here, we explicitly separated spin-wave and DSP terms of $\hat{g}_s$ as in Eq. (S5) and then performed the $\tilde{\epsilon}$ integration with the enforcement of momentum invariance. Note that, in contrast to the two-body problem, here we do not obtain an effective few-body propagator as $\chi(\omega)$, since the three-body T-matrix depends on the energy of the third particle and prevents such grouping.

Now we introduce our main approximation, which allows us to write a self-consistent system of equations to the lowest non-trivial order in r_b/L. For this purpose, we keep only those terms where either the sum over a macroscopic number of spin waves is present or the all-dark intermediate state arises. The sum over a macroscopic number of spin waves introduces a factor of $L\,\delta(x)$, while the all-dark-state propagator contributes $(\omega+i0^+)^{-1}$, which is of order L/r_b because the energy shift satisfies $\omega \sim r_b/L$. In comparison, all other terms are negligible in the limit of vanishing r_b/L, as they are of order ~ 1 or smaller. The key consequence of these rules is as follows: $\{T_3\} = 1$ and $\{T_\mu\} = 2$, where $\{X\}$ denotes the leading order of X in r_b/L. Finally, the two-body term $\{T_2\} = 1$, which follows directly from Eq. (S12).

For clarity, we will omit terms that arise from higher-order corrections to the g_s propagator (such as the last term in Eq. (S14)) as they do not contribute to our lowest-order solution. We note that, in general, the T_2 matrix depends on the total momentum of two particles K, but in all our equations we can either truncate it to U_2, or it arises in a situation (outer T_2 contributions) where $K = 0$.

In order to close the system of equations, we need to calculate T_μ^{ij}, which is described by another set of Faddeev equations expanded to leading order in r_b/L

$$\begin{aligned} T_\mu^{12}(\omega,\vec{x}) &= \sum_{b\neq(12)(3)}\left[U_2^\mu(x_1-x_2)\eta_\mu\,T_2(\omega;x_{b_1}-x_{b_2}) + \sum_{\nu=\pm}\alpha_\nu\,U_2^\mu(x_1-x_2)\eta_{\mu\nu}T_\nu^{b_1b_2}(\omega;\vec{x})\right] \\ &+ \sum_{b\neq(12)(3)}U_2^\mu(x_1-x_2)\eta_\mu\,T_2(\omega;x_{b_1}-x_{b_2})\frac{1}{\omega+i0^+}\left[2\,T_2(\omega) + \sum_{c\neq(b_1b_2)(b_3)}T_3^{c_1c_2}(\omega)\right], \end{aligned} \tag{S15}$$

where $U_2^\mu(x) \equiv U_2(\omega-\epsilon_\mu;x)$ and $\eta_{\mu\nu} = \eta(\omega-\epsilon_\mu-\epsilon_\nu)$. The second line involves processes with intermediate 3-DSP resonances (associated with the $1/\omega$ propagator), and so they become enhanced by an additional factor of $O(L/r_b)$; thus, we must include these terms since, in total, they contribute at the same order as the first line.

For technical reasons, it is easier to first calculate $\tilde{T}_\mu^{12}$, where the tilde denotes that it is a T-matrix where the intermediate DSPs come only from nonperturbative corrections to T_2. This is given by

$$\tilde{T}_\mu^{12}(\omega,\vec{x}) = \sum_{b\neq(12)(3)} \left[U_2^\mu(x_1-x_2)\eta_\mu\, T_2(x_{b_1}-x_{b_2}) + \sum_{\nu=\pm} \alpha_\nu\, U_2^\mu(x_1-x_2)\eta_{\mu\nu}\tilde{T}_\nu^{b_1b_2}(\omega;\vec{x}) \right] \tag{S16}$$

and is related to Eq. (S15) by

$$T_\mu^{12}(\omega;\vec{x}) = \tilde{T}_\mu^{12}(\omega;\vec{x}) + \tilde{T}_\mu^{12}(\omega;\vec{x})\frac{1}{\omega+i0^+}\left(T_3^{13}(\omega)+T_3^{23}(\omega)+2\,T_2(\omega)\right). \tag{S17}$$

This solution can be verified by inserting Eqs. (S16) and (S17) into Eq. (S15) and seeing that everything cancels out (we also use $T_3^{ij}(\omega) = T_3^{kl}(\omega)$ for any $i\neq j$, $k\neq l$). The system of algebraic equation in Eq. (S16) can be solved analytically. Inserting this result into Eq. (S14) and summing over all pairs i,j gives the renormalized system of equations for the dark-state T-matrix

$$T_3(\omega) = T_2(\omega)\frac{2}{\omega+i0^+}T_2(\omega) + T_2(\omega)\frac{2}{\omega+i0^+}T_3(\omega) + \Phi^2\,U_3(\omega) + \Phi^2\,U_3(\omega)\frac{2}{\omega+i0^+}\left[T_3(\omega)+T_2(\omega)\right] + \mathcal{O}(r_b^3/L^3), \tag{S18}$$

where $\Phi = T_2(\omega)/U_2(\omega)$ is the non-perturbative correction. The effective three-body potential $U_3(\omega)$ can be concisely written as

$$U_3(\omega) = \int \frac{dx}{L}\frac{dy}{L}\sum_{\mu=\pm} 2\,U_2(\omega;x-y)\,\eta_\mu\alpha_\mu\,\widehat{T}_\mu^{12}(\omega;x,y), \tag{S19}$$

which was possible due to symmetries with respect to the relabeling of particles and coordinates. Variables $x = x_1 - x_2$ and $y = x_3 - x_2$ describe relative distances of pairs of particles and $\widehat{T}_\mu^{12}(\omega;x,y) = \tilde{T}_\mu^{12}(\omega;x,y)/\Phi$ is governed by Eq. (S16) with $T_2 \to U_2$. The function $U_3(\omega)$ can be intuitively understood as an effective three-body potential, with direct analogy to $U_2(\omega)$ in the two-body scenario.

Solving Eq. (S18) for $T_3(\omega)$, we get

$$T_3(\omega) = \frac{2T_2(\omega)^2 + (\omega+2T_2(\omega))\,U_3/(1-U_2[1-\chi(\omega)\,\omega])^2}{\omega - 2\left(T_2(\omega)+U_3(\omega)/(1-U_2(\omega)[1-\chi(\omega)\,\omega])^2\right)}, \tag{S20}$$

Its pole gives the equation for the three-body energy shift:

$$\delta E_3 = 3U_2(\delta E_3) + 3U_3(\delta E_3) - \chi(0)U_2(\delta E_3)\,\delta E_3 + \mathcal{O}(r_b^3/L^3). \tag{S21}$$

To obtain a self-consistent solution, we must expand both sides to the appropriate order in r_b/L. For this purpose, we analyze the order of each constituent:

$$\{\delta E_3\} = 1, \qquad \{U_2\} = 1, \qquad \{U_3\} = 2, \qquad \{\chi(0)\} = 0. \tag{S22}$$

This allows us to write the final solution

$$\delta E_3 = \underbrace{3U_2}_{\mathcal{O}(r_b/L)} + \underbrace{3U_3 + 3U_2\left(3U_2' - \chi\,U_2\right)}_{\mathcal{O}(r_b^2/L^2)} + \mathcal{O}(r_b^3/L^3), \tag{S23}$$

where $U_2' = dU_2/d\omega$ and all functions are evaluated at $\omega = 0$. Notice that the multi-branch character of the problem is contained in U_3 and in the more complicated form of χ compared to the regime $\Omega \ll |\delta|$.

We note that our calculations apply only when the perturbative expansion in Eq. (S23) is valid. As a result, at the three-body resonance, we cannot definitively say whether the three-body loss dominates over two-body loss for finite r_b/L because we have not obtained any bounds on higher order terms in the expansion. However, if the perturbative series has a nonzero radius of convergence and higher-order terms do not have resonances at exactly the same Δ/Ω (we expect both of these to be true), then there exist values of small enough γ and r_b/L for which the three-body loss dominates over 2-body. We also remark, that, even if the above does not hold, we will still get an enhancement of three-body loss upon approaching the resonance.

IV. TRANSMISSION CALCULATIONS

In this section, we present the model describing the transmission of photons in a chiral waveguide coupled to a single-mode cavity described by Eq. (6) in the main text. We provide analytic expressions of the three-body S-matrix when $\kappa = 0$, which allows us to compute $\eta(0,0)$ presented in Fig. 4(b). We also analytically calculate the three-body parameter r_3 in the case when $u_2 = \kappa = 0$, which corresponds to the situation where three-body effects dominate. The results presented in this section are used to obtain Fig. 4(a-b) in the main text.

When $\kappa = 0$, we use an effective low-energy model that describes the coupling between the waveguide and the DSPs in terms of the transmission Hamiltonian

$$H_{\mathrm{tr}} = \int_{-\infty}^{+\infty} dk\, k\, C^\dagger(k)C(k) + \int_{-\infty}^{+\infty} dk\, \sqrt{2\pi} g_w \left(b^\dagger C(k) + C^\dagger(k) b\right) + u_2\, (b^\dagger)^2 b^2 + u_3\, (b^\dagger)^3 b^3, \tag{S24}$$

where the first term describes the Hamiltonian of the photons in the chiral waveguide. $C^\dagger(k)$ and $C(k)$ are creation and annihilation operators of chiral photons at momentum k, respectively. Speed of light is set to unity ($c = 1$). The second term describes the quadratic coupling between photons in the waveguide and the cavity DSPs, where $b^\dagger$ (b) creates (destroys) a cavity DSP. The last two terms describe the two-body and three-body nonlinear interactions of DSPs, respectively. This treatment neglects higher order corrections to $u_{2,3}$ in the bare waveguide-cavity coupling g_w and similarly neglects r_b/L corrections to g_w.

To compute few-photon scattering, it is convenient to partition the Hamiltonian into the quadrtic part H_0 and the nonlinear interactions:

$$\begin{aligned} H_{\mathrm{tr}} &\equiv H_0 + U, \\ H_0 &= \int_{-\infty}^{+\infty} dk\, k\, C^\dagger(k)C(k) + \int_{-\infty}^{+\infty} dk\, \sqrt{2\pi} g_w \left(b^\dagger C(k) + C^\dagger(k) b\right), \\ U &= u_2\, (b^\dagger)^2 b^2 + u_3\, (b^\dagger)^3 (b)^3. \end{aligned} \tag{S25}$$

The quadratic part H_0 of the Hamiltonian H_{tr} can be diagonalized into the scattering eigenstates:

$$\begin{aligned} H_0 &= \int_{-\infty}^{+\infty} dk\, k \psi_k^\dagger \psi_k, \\ \psi_k^\dagger &= e_k b^\dagger + \int_{-\infty}^{+\infty} dk'\, \psi_k(k') C^\dagger(k), \end{aligned} \tag{S26}$$

where $e_k = \frac{1}{\sqrt{2\pi}} \frac{g_w}{k + i\kappa + i\Gamma}$, $\Gamma \equiv |g_w|^2/2$ and $[\psi_k, \psi_{k'}^\dagger] = \delta(k - k')$. $\{\psi_k^\dagger | k \in (-\infty, +\infty)\}$ form a complete basis of the Hilbert space, which we refer to as the dressed-photon basis. Let $\psi_k(z)$ be the Fourier transform of $\psi_k(k')$ in the coordinate space. The asymptotic behavior of $\psi_k(z)$ gives the single-photon transmission coefficient

$$t_k = \frac{\lim_{z\to+\infty} \psi_k(z)}{\lim_{z\to-\infty} \psi_k(z)} = \frac{k - i\Gamma}{k + i\Gamma}. \tag{S27}$$

The nonlinear interaction U can also be expressed in the dressed photon basis:

$$U = \int dk_1 dk_2 dk_1' dk_2'\, U^{(2)}(k_1, k_2, k_1', k_2') \psi_{k_1'}^\dagger \psi_{k_2'}^\dagger \psi_{k_1} \psi_{k_2} + \int d\vec{k} d\vec{k'}\, U^{(3)}(\vec{k}, \vec{k'}) \psi_{k_1'}^\dagger \psi_{k_2'}^\dagger \psi_{k_3'}^\dagger \psi_{k_1} \psi_{k_2} \psi_{k_3}, \tag{S28}$$

$$U^{(2)}(\vec{k}, \vec{k'}) = e_{k_1}^* e_{k_2}^* e_{k_1'} e_{k_2'} u_2, \qquad U^{(3)}(\vec{k}, \vec{k'}) = e_{\vec{k}}^* e_{\vec{k'}} u_3, \tag{S29}$$

where we have used the definitions $\vec{k} = (k_1, k_2, k_3)$ and $e_{\vec{k}} = e_{k_1} e_{k_2} e_{k_3}$. Note that, in the dressed-photon basis, both the two-body and three-body interactions $U^{(2)}(\vec{k}, \vec{k'})$, $U^{(3)}(\vec{k}, \vec{k'})$ are expressed as the product of separable functions of the incoming and outgoing momenta. This feature allows the few-body scattering processes to be computed analytically.

Before discussing three-photon scattering, we compute the two-body T-matrix. The separable form of the interactions allows us to introduce a simple ansatz for the two-body dressed-photon T-matrix, which is a solution to the Lippmann-Schwinger equation:

$$T^{(2)}(\omega, k_1, k_2, k_1', k_2') = U^{(2)}(k_1, k_2, k_1', k_2') + \int_{-\infty}^{+\infty} dk_1'' dk_2'' \frac{U^{(2)}(k_1'', k_2'', k_1', k_2')}{\omega - k_1'' - k_2''} T^{(2)}(\omega, k_1, k_2, k_1'', k_2''). \tag{S30}$$

Proposing an ansatz $T^{(2)}(\omega, k_1, k_2, k_1', k_2') = \bar{T}^{(2)}(\omega) e_{k_1}^* e_{k_2}^* e_{k_1'} e_{k_2'}$ and inserting it into Eq. (S30), we find

$$\bar{T}^{(2)}(\omega) = \frac{1}{\frac{1}{u_2} - \frac{1}{\omega + 2i\Gamma}}, \tag{S31}$$

where we have assumed that Im$[\omega] > 0$.

Hence we obtain the unsymmetrized two-photon S-matrix for the dressed photons:

$$S^{(2)}(k_1, k_2, k_1', k_2') = \delta(k_1 - k_1')\delta(k_2 - k_2') - 2\pi i\delta(k_1 + k_2 - k_1' - k_2')T^{(2)}(k_1 + k_2 + i0, k_1, k_2, k_1', k_2'), \tag{S32}$$

which represents the transmission amplitude of incoming dressed photons with energy k_1, k_2 scattered into outgoing dressed photons with energy k_1', k_2'. Note that the S-matrix $S_0^{(2)}$ for free photons (as opposed to dressed photons) is more relevant for direct experimental measurements and can be obtained from the dressed-photon S-matrix $S^{(2)}$ using single-photon transmission coefficients: $S_0^{(2)}(k_1, k_2, k_1', k_2') = t_{k_1'} t_{k_2'} S^{(2)}(k_1, k_2, k_1', k_2')$, where $S^{(2)}$ is given by Eq. (S32).

We continue with the discussion of the three-photon S-matrix. Similar to the two-photon case, the S-matrix for the free photons $S_0^{(3)}(\vec{k}, \vec{k'})$ is related to the S-matrix for dressed photons via $S_0^{(3)}(\vec{k}, \vec{k'}) = t_{k_1'} t_{k_2'} t_{k_3'} S^{(3)}(\vec{k}, \vec{k'})$, where $S^{(3)}$ is the dressed-photon S-matrix given by

$$S^{(3)}(\vec{k}, \vec{k'}) = \delta(\vec{k} - \vec{k'}) - 2\pi i\delta(K - K')T^{(3)}(K + i0, \vec{k}, \vec{k'}), \tag{S33}$$

which represents the transmission amplitude of incoming dressed photons with energy $\vec{k} = (k_1, k_2, k_3)$ scattered into outgoing dressed photons with energy $\vec{k'} = (k_1', k_2', k_3')$. K and K' are the total momenta/energies of the incoming and outgoing photons: $K = k_1 + k_2 + k_3$, $K' = k_1' + k_2' + k_3'$.

Although $T^{(3)}(K + i0, \vec{k}, \vec{k'})$ has analytic solutions, its derivation for general values of u_2, u_3 is complicated and is beyond the scope of this supplement. Below, we provide the analytic expressions of $T^{(3)}(K + i0, \vec{k}, \vec{k'})$ for general values of u_2, u_3 without derivation, while details of the derivation will be given in our upcoming work [S3]. But before doing that, we want to show that $T^{(3)}(K + i0, \vec{k}, \vec{k'})$ can be solved easily for the case of $u_2 = 0$. In this case, the three-body T-matrix is computed from the Lippmann-Schwinger equation:

$$T^{(3)}(\omega, \vec{k}, \vec{k'}) = U^{(3)}(\vec{k}, \vec{k'}) + \int_{-\infty}^{+\infty} d\vec{k''} \frac{U^{(3)}(\vec{k''}, \vec{k'})}{\omega - k_1'' - k_2'' - k_3''} T^{(3)}(\omega, \vec{k}, \vec{k''}), \tag{S34}$$

which has a similar form compared to Eq. (S34) for the two-body case with nonzero u_2. Proposing an ansatz $T^{(3)}(\omega, \vec{k}, \vec{k'}) = \bar{T}_c^{(3)}(\omega) e_{\vec{k}}^* e_{\vec{k'}}$ and inserting it into Eq. (S34), we can get

$$\bar{T}_c^{(3)}(\omega) = \frac{1}{\frac{1}{u_3} - \frac{1}{\omega + 3i\Gamma}}, \tag{S35}$$

where we have assumed that Im$[\omega] > 0$. In the following, we provide the analytic solution of $T^{(3)}(K + i0)$ for general values of u_2, u_3 without derivation.

A. $T^{(3)}$ for general u_2, u_3

In many experiments where a coherent pulse is sent through the waveguide, the incoming photons have the same frequency. Here, we only provide the expression of $T^{(3)}(\omega, \vec{k}, \vec{k'})$ when the incoming photons have the same momentum/energy k: $\vec{k} = (k, k, k)$. $T^{(3)}(\omega, \vec{k}, \vec{k'})$ for different incoming photon momenta can be found in our upcoming work [S3].

When $u_2 \neq 0$, the photons can interact pairwise; the dressed-photon T-matrix can be decomposed into the two-body T-matrices between any pair of dressed photons, and the connected part $T_c^{(3)}$ that describes the scattering processes involving all three photons:

$$T^{(3)}(K + i0, \vec{k}, \vec{k'}) = \sum_{i=1, i\neq j\neq l}^{3} \delta(k - k_i')T^{(2)}(2k + i0, k, k, k_j', k_l') + T_c^{(3)}(K + i0, \vec{k}, \vec{k'}). \tag{S36}$$

Here, $T^{(2)}(\omega, k, k, k_j', k_l')$ is the two-body T-matrix that describes the scattering of a dressed photon pair (labelled by j, l), isolated from a freely propagating dressed photon (labelled by i). From Eq. (S31), we have, for Im$[\omega] > 0$,

$$T^{(2)}(\omega, k, k, k_j', k_l') = \bar{T}^{(2)}(\omega) e_{k_j'} e_{k_l'} e_k^{*2}, \quad \bar{T}^{(2)}(\omega) = \frac{1}{\frac{1}{u_2} - \frac{1}{\omega + 2i\Gamma}}. \tag{S37}$$

The connected part of the three-photon T-matrix, $T_c^{(3)}(\omega,\vec{k},\vec{k}')$, can be written as

$$T_c^{(3)}(\omega,\vec{k},\vec{k}')=\bar{T}_c^{(3)}(\omega,\vec{k},\vec{k}')e_{\vec{k}'}e^*_{\vec{k}}, \tag{S38}$$

where, when $\omega=K+i0$,

$$\bar{T}_c^{(3)}(K+i0,\vec{k},\vec{k}')=\bar{T}^{(2)}(2k+i0)\left\{2\sum_{i=1}^{3}h(k_i')+\frac{3}{2}\frac{[3B(2A+1)+4A]\sum_{i=1}^{3}f_i(\vec{k}')+3B}{1-A(2+3B)}\right\}+\bar{V}^{(3)}\frac{1-2A+\sum_{i=1}^{3}f_i(\vec{k}')}{1-2A-3AB}. \tag{S39}$$

Here, $\bar{V}^{(3)}$, A, and B are constants that depend on the total energy K but not the individual outgoing momenta:

$$\bar{V}^{(3)}=\frac{1}{\frac{1}{u_3}-\frac{1}{K+3i\Gamma}},\quad A=\frac{\bar{T}^{(2)}(K+i\Gamma)}{K+3i\Gamma},\quad B=\frac{\bar{V}^{(3)}}{K+3i\Gamma}, \tag{S40}$$

where $\bar{V}^{(3)}$ is equal to the value of $\bar{T}_c^{(3)}(K+i0)$ corresponding to $u_2=0$ (see Eq. (S35)). $f(k_i')$ and $h(k_i')$ are functions that depend on the outgoing momenta:

$$f(k_i')=\bar{T}^{(2)}(K-k_i'+i0)\frac{1}{K-k_i'+2i\Gamma}, \tag{S41}$$

$$h(k_i')=\bar{T}^{(2)}(K-k_i'+i0)\frac{1}{2k-k_i'+i\Gamma}. \tag{S42}$$

When $u_2=0$, $\bar{T}^{(2)}(2k)=A=f_i(\vec{k}')=0$. From Eqs. (S36) and (S39), we can verify that $\bar{T}_c^{(3)}(\omega,\vec{k},\vec{k}')=\bar{V}^{(3)}$, which agrees with Eq. (S35).

B. r_3 and $\eta(0,0)$

Next, we calculate r_3 and $\eta_3(0,0)$ defined in the main text. To define r_3 and $\eta_3(0,0)$, we first introduce the two-photon and three-photon correlation functions at the output of the waveguide. The two-photon correlation function $g^{(2)}(\tau)$ is a function of the time difference $\tau=t_2-t_1$ between the photon number measurements at times $t_{1,2}$. For a weak and continuous coherent-state input with photon momentum k, $g^{(2)}(\tau)$ is related to the output two-photon wavefunction $\psi^{(2)}(z_1=t_1,z_2=t_2)$ in the dressed photon-basis:

$$g^{(2)}(\tau)=|\psi^{(2)}(z_1,z_2=z_1+\tau)|^2, \tag{S43}$$

where $\psi^{(2)}(z_1,z_2)$ is a Fourier transform of $S^{(2)}(k,k,k_1',k_2')$ with respect to the output momenta k_1',k_2'. In the center of mass coordinates $R=(z_1+z_2)/2,\tau=z_2-z_1$, $\psi^{(2)}$ is given by

$$\psi^{(2)}(\tau,R)=\exp(2ikR)[1-\phi^{(2)}(\tau)], \tag{S44}$$

where $\phi^{(2)}(\tau)$ is the Fourier transform of $2\pi iT^{(2)}(2k+i0,k,k,k_1'=k+q,k_2'=k-q)$ with respect to the relative momentum q.

Similarly, the three-photon correlation function $g^{(3)}(\tau_1,\tau_2)$ is a function of the time differences between the photon number measurements at times $t_{1,2,3}$: $\tau_1=t_2-t_3,\tau_2=t_3-t_1$. Again, for a weak and continuous coherent-state input with photon momentum k, $g^{(3)}(\tau_1,\tau_2)$ is related to the output three-photon wavefunction $\psi^{(3)}(z_1=t_1,z_2=t_2,z_3=t_3)$ in the dressed-photon basis, which is the Fourier transform of $S^{(3)}(\vec{k}=(k,k,k),\vec{k}')$ with respect to the output momenta $\vec{k}'$. In the center of mass coordinates $\tau_1=z_2-z_3$, $\tau_2=z_3-z_1$, $R=(z_1+z_2+z_3)/3$, we have

$$g^{(3)}(\tau_1,\tau_2)=|\psi^{(3)}(\tau_1,\tau_2,R)|^2,$$
$$\psi^{(3)}(\tau_1,\tau_2,R)=\exp(3ikR)[1-\phi^{(3)}(\tau_1,\tau_2)], \tag{S45}$$

where $\phi^{(3)}(\tau_1,\tau_2)$ is the Fourier transform of $2\pi iT^{(3)}(3k+i0,\vec{k},k_1'=k-q_2,k_2'=k+q_1,k_3'=k-q_1+q_2)$ with respect to q_1,q_2. Using Eq. (S36), $\phi^{(3)}(\tau_1,\tau_2)$ can be decomposed as

$$\phi^{(3)}(\tau_1,\tau_2)=\phi^{(2)}(\tau_1)+\phi^{(2)}(\tau_2)+\phi^{(2)}(\tau_1-\tau_2)+\phi_c^{(3)}(\tau_1,\tau_2), \tag{S46}$$

where the connected three-photon wavefunction $\phi_c^{(3)}(\tau_1,\tau_2)$ is the Fourier transform of $2\pi i T_c^{(3)}(3k+i0,\vec{k},k_1'=k-q_2,k_2'=k+q_1,k_3'=k-q_1+q_2)$ with respect to q_1,q_2.

In the case of zero one- and two-body losses, r_3 represents a good measure of three-body loss:

$$r_3=\int_{-\infty}^{+\infty} d\tau_1 d\tau_2\,[1-g^{(3)}(\tau_1,\tau_2)]. \tag{S47}$$

Using Eq. (S45), r_3 can be calculated using the dressed-photon three-body T-matrix:

$$\begin{aligned} r_3&=\int_{-\infty}^{+\infty} d\tau_1 d\tau_2\,[1-|1-\phi^{(3)}(\tau_1,\tau_2)|^2] &\text{(S48)}\\ &=\int_{-\infty}^{+\infty} d\tau_1 d\tau_2\,\{2\mathrm{Re}[\phi^{(3)}(\tau_1,\tau_2)]-|\phi^{(3)}(\tau_1,\tau_2)|^2\} &\text{(S49)}\\ &=2\mathrm{Re}[2\pi i T^{(3)}(K+i0,\vec{k},\vec{k}'=\vec{k})]-\int_{-\infty}^{+\infty} dq_1dq_2\left|2\pi i T^{(3)}(K+i0,\vec{k},k_1'=k-q_2,k_2'=k+q_1,k_3'=k-q_1+q_2)\right|^2. &\text{(S50)}\end{aligned}$$

When $u_2=0$, the dressed photon T-matrix only has the connected component $T^{(3)}(K+i0,\vec{k},\vec{k'})=T_c^{(3)}(K+i0,\vec{k},\vec{k'})=\bar{T}^{(3)}(\omega)e^*_{\vec{k}}e_{\vec{k'}}$, where $\bar{T}^{(3)}(\omega)$ is given by Eq. (S35). In this case, r_3 can be analytically calculated to be

$$r_3=4\pi\left(\frac{\Gamma}{\pi}\frac{1}{k^2+\Gamma^2}\right)^3\frac{\mathrm{Im}[-\frac{1}{u_3^*}]}{\left|\frac{1}{u_3}-\frac{1}{3k+3i\Gamma}\right|^2}\geq 0. \tag{S51}$$

Another good measure of three-body loss is $\eta_3(\tau_1,\tau_2)$, defined as

$$\eta_3(\tau_1,\tau_2)=g^{(2)}(\tau_1)+g^{(2)}(\tau_2)+g^{(2)}(\tau_1-\tau_2)-2-g^{(3)}(\tau_1,\tau_2). \tag{S52}$$

Hence, $\eta_3(0,0)$ is given by

$$\begin{aligned}\eta_3(0,0)&=3g^{(2)}(0)-2-g^{(3)}(0,0) &\text{(S53)}\\ &=2|\phi_c^{(3)}(0,0)||1-3\phi^{(2)}(0)|-6|\phi^{(2)}(0)|^2-|\phi_c^{(3)}(0,0)|^2, &\text{(S54)}\end{aligned}$$

where

$$\begin{aligned}\phi_c^{(3)}(0,0)&=\int_{-\infty}^{+\infty} dq_1dq_2\;2\pi i T_c^{(3)}(K+i0,\vec{k},k_1'=k-q_2,k_2'=k+q_1,k_3'=k-q_1+q_2), &\text{(S55)}\\ \phi^{(2)}(0)&=\int_{-\infty}^{+\infty} dq\;2\pi i T^{(2)}(2k+i0,k,k,k_1'=k+q,k_2'=k-q). &\text{(S56)}\end{aligned}$$

In Fig. (4) of the main text, we consider the case where incoming photons are resonant with the cavity; i.e., $k=0$.

[S1] P. Bienias, S. Choi, O. Firstenberg, M. F. Maghrebi, M. Gullans, M. D. Lukin, A. V. Gorshkov, and H. P. Büchler, Scattering resonances and bound states for strongly interacting Rydberg polaritons, Phys. Rev. A **90**, 053804 (2014).
[S2] L. D. Faddeev, Scattering theory for a three-particle system, Sov. Phys. JETP **12**, 1014 (1961).
[S3] Y. Wang *et al.*, (in preparation)