Resonant enhancement of three-body loss between strongly interacting photons

Marcin Kalinowski [1,2,3,*] Yidan Wang,[1,3,*] Przemyslaw Bienias,[1,4] Michael J. Gullans [1,4,5] D. P. Ornelas-Huerta,[1]
Alexander N. Craddock [1] Steven L. Rolston,[1] J. V. Porto,[1] Hans Peter Büchler,[6] and Alexey V. Gorshkov [1,4]

[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]*Department of Physics, Harvard University, 17 Oxford Street Cambridge, Massachusetts 02138, USA*
[4]*Joint Center for Quantum Information and Computer Science, NIST/University of Maryland, College Park, Maryland 20742, USA*
[5]*Department of Physics, Princeton University, Princeton, New Jersey 08544, USA*
[6]*Institute for Theoretical Physics III and Center for Integrated Quantum Science and Technology,
University of Stuttgart, 70550 Stuttgart, Germany*

Rydberg polaritons provide an example of a rare type of system where three-body interactions can be as strong as or even stronger than two-body interactions. The three-body interactions can be either dispersive or dissipative, with both types possibly giving rise to exotic, strongly interacting, and topological phases of matter. Despite past theoretical and experimental studies of the regime with dispersive interaction, the dissipative regime is still mostly unexplored. Using a renormalization group technique to solve the quantum three-body problem, we show how the shape and strength of dissipative three-body forces can be universally enhanced for Rydberg polaritons. We demonstrate how these interactions relate to the transmission through a single-mode cavity, which can be used as a probe of the three-body physics in current experiments.

Introduction. Systems exhibiting strong interactions between single photons are an exciting frontier of quantum optics [1]. They are practically relevant for quantum networks [2] and can give rise to new exotic states of matter [3–5]. Obtaining better control and understanding of these systems in the quantum few-body limit is central to realizing this potential in near-term experiments. An important step in this direction is the mastery of the three-body problem. Although in general not analytically solvable, the three-body problem has emergent universal properties, such as the existence of Efimov bound states [6]. Moreover, three-body forces can greatly influence the properties of quantum many-body systems as in the case of nuclear systems [7], neutron stars [8], and fractional quantum Hall states [9].

By coupling photons to Rydberg atoms using electromagnetically induced transparency (EIT) [10], strong and tunable pairwise interactions between photons are achievable [11–29]. Recently, it has been demonstrated that three-body forces between Rydberg polaritons can be very strong as well [23,30–33], which distinguishes them from weaker three-body forces engineered with ultracold atoms [34] and molecules [35–38]. However, those previous studies considered dispersive three-body interactions in the regime of unitary evolution, whereas dissipative interactions in open Rydberg-EIT systems have

only begun to be explored [39]. Dissipative forces are of interest as they often lead to exotic nonequilibrium dynamics in driven-dissipative systems [12,40–43], while also finding applications in engineering topological phases of matter such as the Pfaffian state [44].

In this work, we study the influence of dissipative three-body interactions on the physics of Rydberg cavity polaritons. Pure three-body scattering processes in Rydberg-EIT systems are strong and often comparable to two-body effects [23,24,30,31,45]. There is also evidence that effective three-body interactions are enhanced in this system [23,30,31] due to Rydberg blockade effects [46]. Here, by studying a simplified cavity model that can be treated with a rigorous renormalization group technique, we clearly establish the existence of a regime where both dispersive and dissipative three-body forces can be universally enhanced in a tunable fashion. This enhancement appears due to a near-resonant process when the incoming state can conserve energy and momentum by scattering to a large manifold of intermediate lossy states. Due to the role played by an intermediate resonant channel, this effect has similarities to Feshbach resonances [47]. The interaction can be tuned using the strength and the frequency of the classical control fields. We show how these effects can be probed in current experiments by studying the cavity transmission.

Because of the multicomponent nature of the Rydberg polaritons, the theoretical description of the three-body problem is nuanced and complex. To make progress, we concentrate on cavity models for interacting Rydberg atoms under EIT conditions [14,48–52]. We derive analytical formulas for the interaction-induced shifts in energies and decay rates of three dark-state polaritons (DSPs). We develop a method to derive

an effective Hamiltonian for the DSPs alone by solving a simplified version of the Faddeev equations. It is only with the considerable simplifications afforded by the Faddeev equation formalism that we have been able to rigorously solve for the three-body force in the relevant parameter regimes [53].

Prior work. We now compare our results to previous theoretical work on the three-body Rydberg polariton problem [23,30,31,45]. The present work has the most direct overlap with Refs. [30,31], but, as mentioned above, those works consider the limit $\Omega \ll |\Delta|$, where only one spin-wave branch contributes. Here, we must consider the more general problem that includes both spin-wave branches, significantly increasing the technical challenges. Moreover, Refs. [30,31] made different heuristic approximations, which led to small quantitative differences between all three solutions for the effective three-body force: Ref. [30], Ref. [31], and the present work. The approach presented here is more systematic and can be rigorously derived as an asymptotic perturbative expansion of the solutions to the three-body Schrödinger equation. We present further extensions of these results to general multi-mode cavities in our upcoming work [54]. Generalizing these cavity solutions to the free-space problem remains an outstanding challenge.

In Ref. [23], an alternative approach for free-space was developed based on defining effective three-body parameters through nonperturbative matching. In this approach, the three-body parameters are tuned in an effective field theory to reproduce low-energy observables (e.g., the dimer-polariton scattering length) obtained from the solution to the microscopic model. This method is based on exact numerical solutions of the three-body Faddeev equations, which increases accuracy, but can be difficult to implement.

Finally, Ref. [45] studies an enhanced three-body loss feature in free space that occurs in a similar parameter regime to the one we consider here, but has a richer physical origin due to the complexity of the free-space problem. It is argued in Ref. [45] that the observed resonant enhancement of three-body loss can be explained through a perturbative analysis of the effective interactions. In the analysis of Ref. [45], the enhanced three-body loss arises from a resonant enhancement of the effective two-body potential when the blockade radius goes to zero. In contrast, the enhancement found in the present work arises from a resonant three-body process that does not play a role in the two-body problem.

System. The medium we consider consists of three-level atoms with ground state $|g\rangle$ and an intermediate state $|p\rangle$ coupled to Rydberg state $|s\rangle$ by a coherent laser, with Rabi frequency Ω and a complex detuning $\Delta = \delta - i\gamma$ [Fig. 1(a)], which captures the $|p\rangle$ state's decay rate 2γ. The atomic cloud is suspended in a single-mode running-wave cavity. The quantum photon field, with collective coupling g, is tuned to the EIT resonance with the noninteracting Hamiltonian

$$H_0 = \int dz\, \vec{\psi}^\dagger(z) \begin{pmatrix} 0 & g & 0 \\ g & \Delta & \Omega \\ 0 & \Omega & 0 \end{pmatrix} \vec{\psi}(z), \qquad (1)$$

where $\vec{\psi}^\dagger(z) = [u_0^*(z)a^\dagger, P^\dagger(z), S^\dagger(z)]$ is a vector of bosonic creation operators for the cavity field a with mode function $u_0(z)$ and atomic states $|p\rangle$, $|s\rangle$ at position z. We set
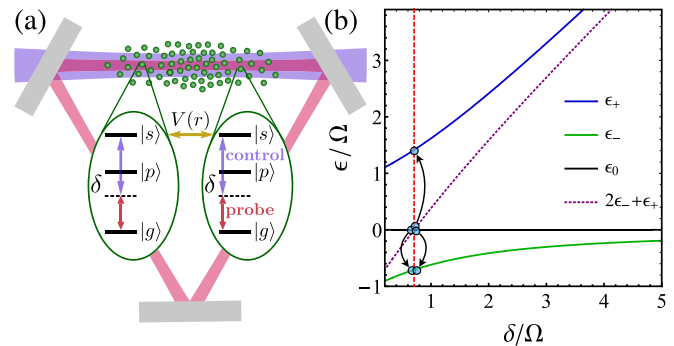

FIG. 1. (a) Gas of neutral atoms is suspended in an optical cavity. Each atom is a three-level system with the ground state $|g\rangle$, intermediate lossy state $|p\rangle$ with half-width γ, and a high-lying Rydberg state $|s\rangle$, which experiences strong interactions. Classical control field with Rabi frequency Ω and detuning δ couples states $|p\rangle$ and $|s\rangle$. Quantum photon field with collective coupling g drives the $|g\rangle$ - $|p\rangle$ transition and is tuned to the two-photon resonance between states $|g\rangle$ and $|s\rangle$ (via $|p\rangle$). (b) Energy of the upper (blue) and lower (green) branches of spin waves as a function of the single-photon detuning. At $\delta = \Omega/\sqrt{2}$ scattering of three DSPs (black) into spin waves (dotted purple) is on resonance.

$\hbar = 1$ throughout. We assume that the energy splitting to other modes of the cavity is much larger than g, which is, in turn, much larger than all the other energy scales. In this single-mode limit, H_0 couples the cavity field to one $|p\rangle$ mode and one $|s\rangle$ mode, both with the same mode function $u_0(z)$. Diagonalizing the resulting 3×3 matrix leads to three eigenmodes. The zero-energy mode is the DSP, which has no overlap with the lossy intermediate state. The two "bright-state" polariton modes are energetically separated and do not influence the DSP behavior in the experimentally relevant limit of strong coupling (large g) considered here. In practice, $g = \sqrt{\gamma c\, OD/2L}$, and it is sufficient to have $OD/L \geqslant 0.1\ \mu\text{m}^{-1}$ for our analysis to apply, which is readily achieved in experiment [29]. The remaining eigenstates of H_0 (spin waves) correspond to the excitations of the atomic cloud, have no photonic component, and couple to the polaritons only via atom-atom interactions. In the presence of Rydberg interactions $H_{\text{int}} = \frac{1}{2}\int dz dz'\, S^\dagger(z)S^\dagger(z')V(z-z')S(z')S(z)$, polaritons experience an effective two-body potential [55–57]

$$U_2(\omega; r) = \frac{V(r)}{1 - \chi(\omega)V(r)}, \qquad (2)$$

where $\chi(\omega)$ is a function of Δ, Ω, and the energy ω of the incoming polaritons. The bare interaction $V(r) = C_6/r^6$ is the van der Waals potential between two atoms separated by distance r. We see that, at large distances, the $1/r^6$ tail is transferred onto the polaritons, while, for r below the blockade radius $r_b = |\chi(0)C_6|^{1/6}$, the two Rydberg states are shifted out of resonance (the so-called Rydberg blockade mechanism) leading to a saturation of the effective potential.

To gain insight into few-body interactions, we consider a cavity as our setup, since its treatment requires only a finite number of photonic modes. When there are only a few relevant photonic modes, there is a natural separation of

scales that appears between low-energy polaritons and high-energy atomic excitations (spin waves). We take advantage of this energy separation to obtain an effective theory for the polaritons—renormalized by the influence of high-energy spin waves.

For simplicity, we consider an effectively one-dimensional running-wave cavity with a single, fixed-momentum photonic mode on EIT resonance and a uniform density of atoms filling the entire cavity mode. We present the generalization of our results to nonuniform setups, e.g., as in Fig. 1(a), in our upcoming work [54]. We focus on this model because it captures generic features of multimode systems, while simplifying technical aspects of the calculations.

Independent of the geometry, in such cavity models, the interactions between polaritons most simply appear as shifts in the energies and decay rates of the polariton modes. To calculate these shifts, we use a master equation description of the problem in the weak-driving regime, such that the anti-Hermitian part of the non-Hermitian Hamiltonian is sufficient to account for losses in the system [58]. Complex energy shifts for the two- and three-body problems coincide with the value of poles in the corresponding two- and three-body T matrix describing correlation functions in this system.

Qualitative picture. Before moving to the main derivations, we discuss the physical origin of the enhanced three-body dissipation in our system as it has an intuitive explanation. Three dark-state polaritons propagating at EIT resonance have zero energy. At the same time, upper and lower branches of spin waves have energies $\epsilon_\pm = \frac{1}{2}(\Delta \pm \sqrt{\Delta^2 + 4\Omega^2})$. Strong losses occur at $\delta = \Omega/\sqrt{2}$ because, at this point, $2\epsilon_- + \epsilon_+ = 0$, which means that three-body scattering into lossy atomic excitations is on energetic resonance [Fig. 1(b)].

Two-body problem. First, we turn to the two-body problem. Consider an incoming state of two polaritons (labeled 1 and 2) initially located at positions $\vec{x} = (x_1, x_2)$ and later measured at positions $\vec{x}'$ after interactions take place. The amplitude for this process can be described within the framework of scattering theory. The multicomponent nature of the polariton problem means that the full (bold) two-body T matrix $\hat{T}_2(\omega)$ is a 3×3 operator-valued matrix. However, only the Rydberg ($|s\rangle$) component experiences interactions. Therefore, we can restrict our considerations to the ss component $\hat{T}_2(\omega)$ of the full two-body T matrix $\hat{\boldsymbol{T}}_2(\omega)$ [55,59,60]. $\hat{T}_2(\omega)$ is governed by the Lippmann-Schwinger equation; see Fig. 2(a). In these cavity problems, one can equivalently study the integrated T matrix $T_2(\omega) \equiv \int d\vec{x} d\vec{x}' \, T_2(\omega; \vec{x}, \vec{x}')$, where $T_2(\omega; \vec{x}, \vec{x}')$ is the matrix element of $\hat{T}_2$ in coordinate space. Then, the solution of the Lippmann-Schwinger equation is

$$T_2(\omega) = \frac{\omega \, U_2(\omega)}{\omega - U_2(\omega)[1 - \omega \, \chi(\omega)]}, \qquad (3)$$

where $U_2(\omega) \equiv \int \frac{dr}{L} U_2(\omega; r)$ and L is the mode volume of the DSP. The two-body energy shift is given by the poles of $T_2(\omega)$.

Three-body problem. Previous works on the three-body problem for Rydberg polaritons considered the restricted limit $\Omega \ll |\delta|$ [30–32]; here, we extend the regime of applicability to $\Omega > |\delta|$, which allows for a more general description of the system, including repulsive photons [55] and dissipative behavior.
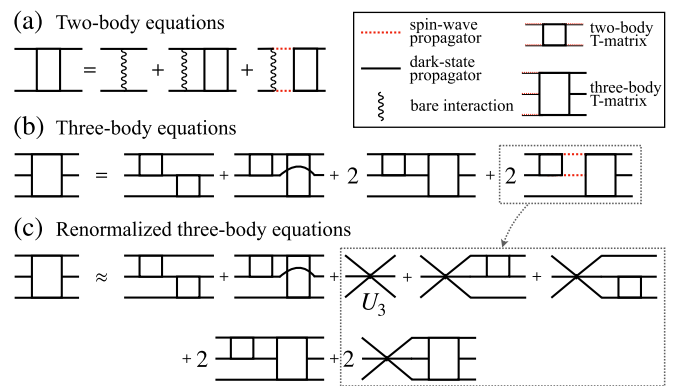

FIG. 2. (a) Diagrammatic representation of the Lippmann-Schwinger equation for the scattering of two dark-state polaritons. Processes with only one spin wave are forbidden by momentum conservation. (b) Schematic representation of Faddeev equations for the three-body T matrix $T_3^{12}(\omega)$, where particles 1 and 2 interact first. (c) Truncation of the three-body equations (b) to second order in r_b/L allows us to express the spin-wave contribution using an effective three-body potential U_3 between DSPs.

Although dramatically simplified, the three-body problem cannot, to the best of our knowledge, be solved exactly in the single-mode cavity model considered here. Instead, we approximate the full result by a power series in the small parameter r_b/L, which is effectively the product of the blockade radius and the density of polaritons in this few-body limit. We stress that this approach is still nonperturbative in the bare interaction $V(r)$. Our final expression for the three-body energy shift to second order in r_b/L is given by [53]

$$
\begin{aligned}
\delta E_3 &= \frac{r_b}{L} E_3^{(1)} + \frac{r_b^2}{L^2} E_3^{(2)} + O\big(r_b^3/L^3\big) \\
&= \underbrace{3U_2}_{O(r_b/L)} + \underbrace{3U_3 + 3U_2(3U_2' - \chi \, U_2)}_{O(r_b^2/L^2)},
\end{aligned} \qquad (4)
$$

where U_3 is a universal effective three-body force (Fig. 2) and $U_2' \equiv dU_2(\omega)/d\omega|_{\omega=0}$. U_3 has a similar expression to U_2 as an integral over a local three-body potential and arises in any cavity geometry, even if the three photons were to occupy different modes, as long as the modes spatially overlap [53,54]. The other terms at order r_b^2/L^2 represent additional nonlocal, nonperturbative corrections that are specific to the cavity geometry we consider.

In contrast to the two-body problem [55], the three-body Rydberg polariton problem cannot be reduced to a single scalar equation—even in the perturbative expansion in r_b/L. Inspired by the seminal work of Faddeev on three-body quantum systems [61], we take the Faddeev equation approach to solving the Schrödinger equation. An indispensable advantage in the present case is that the Faddeev equations can be expressed entirely in terms of Rydberg spin-wave correlation functions, which simplifies the theoretical treatment of the multicomponent nature of DSPs. In this formalism, the three-body problem can be recast as an infinite series of two-body interactions. All scattering processes are grouped depending on which pair of particles interacts first. Crucially, the T matrix separates into the sum $\hat{T}_3(\omega) = \frac{1}{3} \sum_{i<j} \hat{T}_3^{ij}(\omega, \epsilon_k)$, where
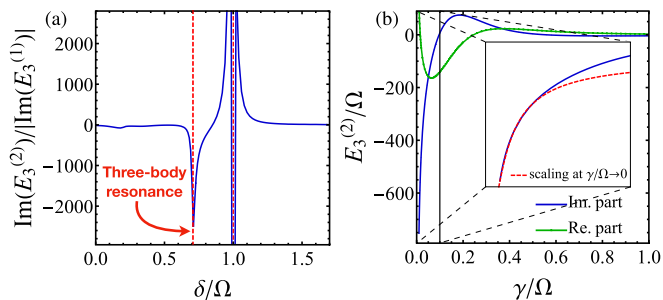
FIG. 3. (a) Ratio of imaginary parts of $E_3^{(2)}$ and $E_3^{(1)}$ at $\gamma/\Omega = 0.01$. Near $\delta/\Omega = 1/\sqrt{2}$, we observe enhancement caused by a three-body resonance. $\delta = \Omega$ is a singular point where $r_b \to 0$. (b) Real and imaginary parts of $E_3^{(2)}$ as a function of γ/Ω at $\delta/\Omega = 1/\sqrt{2}$. Inset: Our numerical results (blue) agree with analytical scaling arguments (dashed red) suggesting that the ratio $\mathrm{Im}(E_3^{(2)})/\mathrm{Re}(E_3^{(2)})$ diverges as Ω/γ in the limit $\gamma/\Omega \to 0$.

FIG. 4. (a) Three-body loss parameter r_3 in units of Γ^{-2} as a function of real and imaginary parts of u_3/Γ for the case of $u_2 = 0$, which corresponds to negligible two-body correlations in the microscopic model. (b) Growth of the three-body correlation function $\eta_3(0,0)$ (red) and that of three-body loss (blue). The parameters used are $\Omega/2\pi = 30$ MHz, $2\gamma/2\pi = 6.1$ MHz, $L = 100\ \mu m$, $OD/L \geqslant 0.1\ \mu m^{-1}$, $\Gamma/\Omega = 2/3$, and $C_6/2\pi = 1.8$ THz μm^6.

$\hat{T}_3^{ij}(\omega, \epsilon_k)$ denotes the T matrix for scattering where particles i, j interact first and the third particle $k \neq i, j$ has incoming energy ϵ_k. Similarly to the two-body case, we consider just the *sss* component $\hat{T}_3$ of the full three-body T matrix $\boldsymbol{\hat{T}_3}$. The equation for $\hat{T}_3^{12}(\omega, \epsilon)$, when all outgoing states are DSPs, is

$$\hat{T}_3^{12}(\omega, \epsilon) = \hat{T}_2^{12}(\omega - \epsilon)\hat{g}_s(\omega)\big[\hat{T}_2^{23}(\omega) + \hat{T}_2^{13}(\omega)\big]$$
$$+ \int d\tilde{\epsilon}\, \hat{T}_2^{12}(\omega - \epsilon)\hat{g}_s(\tilde{\epsilon})\hat{g}_s(\omega - \epsilon - \tilde{\epsilon})$$
$$\times \big[\hat{T}_3^{23}(\omega, \tilde{\epsilon}) + \hat{T}_3^{13}(\omega, \tilde{\epsilon})\big], \qquad (5)$$

where $\hat{T}_2^{ij}$ describes the two-body scattering of particles labeled i, j. The Rydberg-component propagator $\hat{g}_s$ is a complex object that involves contributions from different spin-wave branches and the DSP mode. Note that the simple form of Eq. (5) is thanks to the use of abstract operators. The representation in, e.g., a coordinate basis is more involved [53].

To derive an effective DSP theory, we separate spin-wave and DSP components in $\hat{g}_s$, which will allow us to perform the expansion in r_b/L. The equation for the T matrix describing DSP-to-DSP scattering $\hat{T}_3(\omega) \equiv \hat{T}_3^{12}(\omega, 0)$ is represented diagrammatically in Fig. 2(b), where we explicitly showed separated spin-wave (red) and DSP (black) propagators. Next, we restrict both sides of Eq. (5) to the second 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. Finally, we rewrite the original Faddeev equations in an approximate form shown in Fig. 2(c)—without any spin-wave degrees of freedom. We provide the full set of equations in the Supplemental Material [53].

In Fig. 3(a), we characterize the strength of three-body loss using the ratio of the expansion coefficients $\mathrm{Im}(E_3^{(2)})/\mathrm{Im}(E_3^{(1)})$. The denominator $\mathrm{Im}(E_3^{(1)})$ from Eq. (4) contains contributions to three-body loss from disconnected two-body processes only. We see the expected enhancement at the resonance condition $\delta = \Omega/\sqrt{2}$ [62]. There is another resonant feature at $\delta = \Omega$ that arises due to the vanishing (in
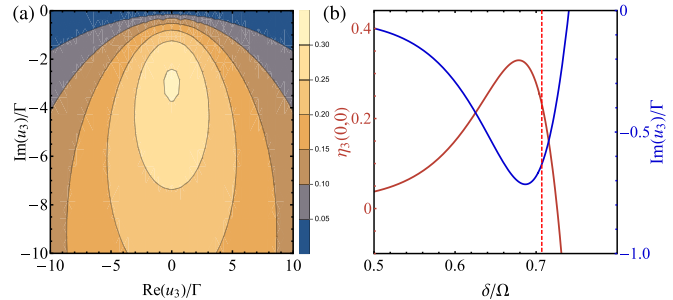
the limit of small γ) of the two-body susceptibility χ and, consequently, r_b. This phenomenon appears even for a two-body problem, and in our case leads to an overall enhancement of both two and three-body interactions, which is not desired. In contrast, for the three-body resonance condition $\delta = \Omega/\sqrt{2}$, there are no resonant features that appear in the two-body problem.

In Fig. 3(b), we show the dependence of $E_3^{(2)}$ on the decay rate γ at the resonance condition $\delta = \Omega/\sqrt{2}$. We find a divergence as $\gamma/\Omega \to 0$ (see inset) indicating that the enhancement factor for three-body loss can be made arbitrarily large. This behavior is in agreement with analytical scaling arguments that predict $\mathrm{Im}(E_3^{(2)}) \sim 1/\gamma$, which stems from the fact that, for finite γ, we have $\mathrm{Im}(2\epsilon_- + \epsilon_+) \sim \gamma$.

Experimental probing. In order to relate our microscopic description to experimentally measurable quantities, we now study transmission of photons resonant with the DSP modes in the cavity, which is essential for experimental relevance of this work. Our analysis follows directly from the prior sections upon including a waveguide as an additional element in the model. Taking a weak-coupling limit between the cavity and this waveguide leads to an effective low-energy model for the transmission where the only excitations in the cavity are the DSPs [53,63,64],

$$H = -i(\Gamma + \kappa)b^\dagger b + u_2(b^\dagger)^2 b^2 + u_3(b^\dagger)^3 b^3, \qquad (6)$$

where $b^\dagger$ is a bosonic creation operator for the DSPs, 2Γ is the decay rate of DSPs from the cavity into the waveguide, 2κ is the decay rate to other modes, and the coefficients u_2, u_3 are chosen to match the energy shifts $\delta E_2, \delta E_3$, calculated from the full microscopic theory, through $u_2 = \delta E_2$ and $u_3 = \delta E_3 - 3\delta E_2$. First, we focus on the limit where three-body effects dominate by taking $u_2 = 0$ in Eq. (6). We use a measure of three-body loss, $r_3 = \int d\tau_1 d\tau_2 [1 - g^{(3)}(\tau_1, \tau_2)]$, that is appropriate when all decay is into the waveguide ($\kappa = 0$) and when two-body interactions u_2 are negligibly small. Here, $g^{(3)}(\tau_1, \tau_2)$ is the three-photon correlation function at the output of the waveguide and $\tau_{1,2}$ are relative coordinates. The r_3 parameter measures the probability that three photons are lost from the pulse due to the interactions. In Fig. 4(a),

we show a contour plot of r_3 as a function of the real and imaginary parts of the three-body interaction u_3. Interestingly, r_3 does not increase arbitrarily as the three-body loss rate is increased, but instead has a maximum value at $\text{Im}(u_3) \sim \Gamma$ due to quantum Zeno-like effects. When u_2 is nonzero, a more appropriate measure of three-body loss is a positive peak in the connected correlation function $\eta_3(0,0) = 3g^{(2)}(0) - 2 - g^{(3)}(0,0)$ [45]. In Fig. 4(b), we plot $\eta_3(0,0)$ for parameters close to those of the usual Rydberg experiments [29]. We observe a peak in $\eta_3(0,0)$ near the resonance, indicating a strong enhancement of the three-body loss.

Outlook. In this work, we showed the existence of a parameter regime for Rydberg polaritons where three-body loss can be resonantly enhanced. We focused on dissipative dynamics because, for currently accessible experimental parameters [25–28], the dissipative interactions can be strongly enhanced by working close to the resonance. Through further experimental improvements and by tuning slightly away from the resonance, one could also operate in a regime of enhanced dispersive three-body interactions. We would like to stress that although our results are based on a perturbative expansion, this does not mean the interactions are weak. On the contrary, the asymptotic expansion in r_b/L means that our results hold for arbitrary optical depths and can give rise to strong effects on the correlations between few photons [65]. The extension of the presented work to free space is another important direction to explore. Our work clearly demonstrates the possibilities offered by Rydberg-EIT to tune the properties of multibody interactions. This motivates further exploration of possible interactions, which might give rise to different exotic phases of matter [33,66].

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