

Anisotropic and Nonadditive Interactions of a Rydberg Impurity in a Quantum Bath

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(Received 26 November 2025; revised 28 April 2026; accepted 10 June 2026; published 13 August 2026)

We present a framework for treating mesoscopic anisotropic and nonadditive impurity–bath interactions, ubiquitous in realistic quantum impurity problems, which are often neglected in conventional approaches relying on additive, spherically symmetric pseudopotentials. To illustrate this general approach, we focus on the specific impurity problem of a Rydberg atom immersed in a Bose-Einstein condensate, where the internal-state degeneracy of the Rydberg impurity gives rise to configuration-dependent nonadditive potentials. With increasing interaction strength, anisotropy-induced partial-wave mixing generates distinct polaron and molaron resonances, allowing for radially and angularly excited bound states to become accessible. This approach captures the anisotropy and nonadditivity characteristic of a Rydberg impurity immersed in a quantum bath, and is broadly applicable to a host of quantum impurity problems beyond the Fröhlich paradigm.

DOI: [10.1103/38vj-6fqc](https://doi.org/10.1103/38vj-6fqc)

Ultracold quantum impurity problems, such as the Bose or Fermi polaron, have been predominantly studied under the assumption that the interaction between the impurity and particles from the surrounding bath is spherically symmetric and additive. This approximation has been successful in diverse physical systems ranging from ultracold neutral atoms [1–9], where a description using the contact zero-range interaction yields good agreement with experimental observations [10–14], to ionic [15–22] and Rydberg [23,24] impurities exhibiting a more intricate structure due to their long-ranged interactions and sundry molecular states [25–28]. However, interparticle interactions are often neither isotropic nor additive. While nonadditivity emerges naturally when the impurity’s internal state evolves dynamically so that its interaction potential depends explicitly on the configuration of surrounding bath particles [29–38], anisotropy appears, for example, in condensates composed of magnetic atoms or dipolar molecules [39–45] or in the study of rotating impurities [46–50].

Despite the ubiquity of such rich interactions, a comprehensive treatment of even the most idealized scenario—a Bose polaron at zero temperature in a homogeneous Bose-Einstein condensate (BEC)—that fully incorporates this physics is still missing. Progress in this direction requires a controllable platform where the relevant effects can be isolated, tuned, and observed. A Rydberg atom embedded in an ultracold quantum gas provides precisely this controllability and localization, as the Rydberg impurity’s internal structure and its interaction with the environment can be spectroscopically engineered [51–55]. Rydberg excitations with finite electronic angular momentum possess degenerate magnetic sublevels [56], which hybridize under perturbation from the bath to produce nonadditive interactions [29,57].

In this Letter, we develop a theoretical framework for the Bose polaron problem that incorporates an anisotropic and nonadditive impurity interaction, utilizing the versatility of Rydberg atoms as a platform for engineering mesoscopic inter-particle interactions. Figure 1 shows an exemplary absorption spectrum $A(\omega)$ of a Rydberg impurity immersed in an ideal BEC. Nonadditivity manifests in two closely related ways. First, the intrinsically many-body energies of multiply occupied impurity bound states are not additively related, and every molecular state must be computed independently and in full rather than additively constructed from lower-order clusters [29]. Second, the bath couples differently to a bare impurity than to a molecule formed with one or more bath particles, thereby endowing each dressed state—polaron or molaron, respectively—with a distinct spectral signature. These facets of nonadditivity

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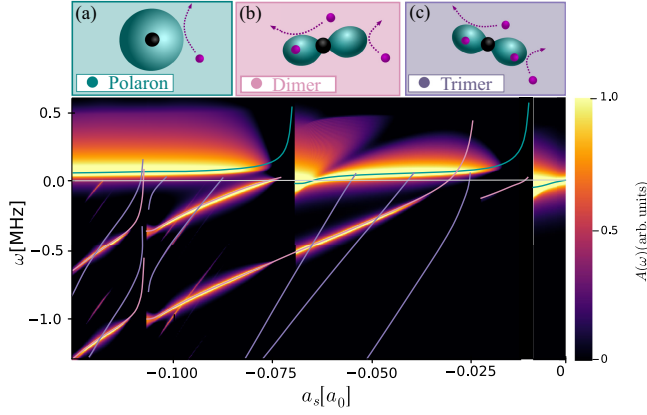


FIG. 1. Absorption spectrum of a Rb|25p> Rydberg impurity in a BEC of density $\rho = 2 \times 10^{14} \text{ cm}^{-3}$, shown as a function of the electron-atom scattering length a_s and detuning ω . We highlighted all polaron (teal) and dimer molaron (pink) spectral features with solid lines as a guide to the eye; as the trimer features have weak spectral weight we only highlighted a selection (purple) so that the remaining weak trimer features are visible. (a) In the polaron regime the interaction is effectively spherical. (b),(c) In the molecular regime, the electron localizes on specific bosons, forming a molecule and creating an anisotropic potential for additional bath atoms. Contributions to the absorption spectrum are truncated at trimers.

arise from the same physical origin: the backaction of the medium upon the impurity's electronic state. Our analysis additionally demonstrates how anisotropy influences the energy shifts induced by condensate density modulations.

We consider a Rydberg impurity with principal quantum number n and angular momentum l in a BEC of N particles at density ρ . Written in atomic units and in the frame comoving with the impurity [58] [59], the Hamiltonian reads

$$\hat{H} = \hat{H}_{\text{Ryd}}(\vec{r}) + \sum_{i=1}^N \left(-\frac{\nabla_{\vec{R}_i}^2}{2\mu} + \hat{V}_{\text{tr}}(\vec{r}, \vec{R}_i) \right), \quad (1)$$

where μ is the reduced mass of the Rydberg atom and one bath atom and $\vec{r}$ ($\vec{R}_i$) denotes the electron (i th bath particle) position operator [51,60–62]. The Hamiltonian for the Rydberg atom satisfies $H_{\text{Ryd}}|nlm\rangle = -[1/2(n - \mu_l)^2]|nlm\rangle$ where μ_l is a quantum defect [63] and the Rydberg wave function is $\langle \vec{r}|nlm\rangle = [u_{nl}(r)/r]Y_{lm}(\hat{r})$. The Rydberg electron interacts with each boson via the contact interaction $\hat{V}_{\text{tr}}(\vec{r}, \vec{R}_i) = 2\pi a_s \delta^3(\vec{r} - \vec{R}_i)$ with strength a_s , the electron-bath atom s -wave scattering length [64].

We employ the Born-Oppenheimer (BO) ansatz for the wave function, $\Phi_\alpha(\{\vec{R}\}_N, \vec{r}) = \phi_\alpha(\vec{r}; \{\vec{R}\}_N)\psi_\alpha(\{\vec{R}\}_N)$, where the electronic states $\phi_\alpha(\vec{r}; \{\vec{R}\}_N)$ depend parametrically on the set of bath coordinates $\{\vec{R}\}_N = \{\vec{R}_1, \dots, \vec{R}_N\}$. The electronic states are determined by diagonalizing $\hat{V}^{\text{el}} = \sum_i^N \hat{V}_{\text{tr}}(\vec{r}, \vec{R}_i)$ in the degenerate subspace of magnetic sublevels such that the eigenvalues of

$$\langle m|\hat{V}_{nl}^{\text{el}}|m'\rangle = \sum_{i=1}^N 2\pi a_s \frac{|u_{nl}(\vec{R}_i)|^2}{R_i^2} Y_{lm}^*(\Omega_i) Y_{lm'}(\Omega_i), \quad (2)$$

give the BO potential energy surfaces $V_\alpha^{\text{BO}}(\{\vec{R}_i\})$. The resulting Rydberg-Bose Hamiltonian is $\hat{H}^{(N)} = \sum_{\alpha=1}^{2l+1} \hat{H}^{N,\alpha}|\alpha\rangle\langle\alpha|$, where

$$\hat{H}^{K,\alpha} = -\sum_{i=1}^K \frac{\nabla_{\vec{R}_i}^2}{2\mu} + V_\alpha^{\text{BO}}(\{\vec{R}\}_K), \quad (3)$$

which shows explicitly how the Rydberg electron mediates the impurity-bath interaction with nonadditivity arising from the degeneracy of the electronic m states.

The absorption spectrum $A(\omega)$ shown in Fig. 1 is obtained from Eq. (3) through the Fourier transform of the Loschmidt echo, $S(t) = \langle \Psi_0|e^{i\hat{H}_0^{(N)}t}e^{-i\hat{H}^{(N)}t}|\Psi_0\rangle$, where $\hat{H}_0^{(N)}$ denotes the Hamiltonian of the initial noninteracting bath with ground-state $|\Psi_0\rangle$. The individual contributions from different few-body complexes (bare atom, dimer, and trimer) with their respective many-body dressing into quasiparticles (polarons and molarons) are highlighted with solid curves [2,25]. The bright feature, typically at small positive detuning and highlighted by the teal line, heralds the formation of a polaron. Here no bath particle occupies a bound state of the interaction potential, and so the Rydberg electron remains unperturbed, and the interaction is additive and effectively spherically symmetric within the BO approximation.

By contrast, the Rydberg molecule localizes the electronic wave function (in the molecular frame) in the vicinity of the perturber bath particle. This creates an anisotropic potential for bath particles to scatter off, dressing the bound complex into a quasiparticle—a molaron [65–67]. Because each molecular state is dressed differently—and differently from the bare atom—the scattering resonances at the dimer (pink) and trimer (purple) thresholds occur at electronic scattering lengths (e.g., $a_s \approx [-0.11, -0.025]$) where the polaron branch remains smooth, while the polaron exhibits resonances (e.g., $a_s \approx [-0.075, -0.02]$) in regimes where the molecular lines show no such behavior. Finally, weakly bound molecules can appear as molarons at positive energies, reflecting the repulsive many-body dressing characteristic of long-range interactions [25].

To capture that each molecule can interact differently with its surroundings, we partition the N -particle Hilbert space of the BEC such that exactly k particles occupy the bound-state subspace B while the remaining $(N - k)$ particles occupy the continuum subspace C . The total Hilbert space then decomposes as

$$\mathcal{H}^{(N)} = \bigoplus_{k=0}^N \mathcal{H}_B^{(k)} \otimes \mathcal{H}_C^{(N-k)}. \quad (4)$$

For this we define a projection P_k onto each partition as

$$\hat{P}_k = \sum_{\Psi} |\Psi\rangle\langle\Psi| \quad \text{with} \quad \hat{N}_B \Psi = k\Psi. \quad (5)$$

Here $\hat{N}_B = \sum_{\beta \in H_B} b_{\beta}^{\dagger} b_{\beta}$, with $b_{\beta}^{\dagger}$ (b_{β}) the creation (annihilation) operator of a boson in state β . This automatically ensures that exactly k bath particles occupy subsystem B and hence $N - k$ particles occupy subsystem C . Inserting the completeness relation $\sum_k \hat{P}_k = \mathbb{1}$ into the Loschmidt echo, we get the natural decomposition

$$S(t) = \sum_{k=0}^N \underbrace{\langle \Psi_0 | e^{i\hat{H}_0^{(N)} t} \hat{P}_k e^{-i\hat{H}^{(N)} t} | \Psi_0 \rangle}_{s_k(t)}. \quad (6)$$

To make use of this, we approximate the total N -particle Hamiltonian as a decomposition into bound $\hat{H}_B$ and continuum $\hat{H}_C$ parts. We do this on phenomenological grounds. The bound Hamiltonian for the α^{th} molecular eigenstate $\Phi_{\alpha,\gamma}(\{\vec{R}\}_k, \vec{r})$ is $\hat{H}^{k,\alpha}$ [Eq. (3)]. In the adiabatic picture, the Rydberg molecule interacts with the remaining $(N - k)$ bath particles through the Rydberg electron whose configuration $\phi_{\alpha,\gamma}(\vec{r}; \{\vec{R}\}_k)$ is determined by the equilibrium position of the bound particles $\{\vec{R}^{\text{eq}}\}_k$. As this state is no longer degenerate, the resulting mediated interaction is additive and can be determined directly from $\langle \phi_{\alpha,\gamma}(\vec{r}; \{\vec{R}^{\text{eq}}\}_k) | \hat{V}^{\text{el}} | \phi_{\alpha,\gamma}(\vec{r}; \{\vec{R}^{\text{eq}}\}_k) \rangle$, leading to

$$\hat{H}_C^{k,\alpha,\gamma} = \sum_{i=k+1}^N \left[-\frac{\nabla_{\vec{R}_i}^2}{2\mu} + 2\pi a_s \left| \phi_{\alpha,\gamma}(\vec{R}_i; \{\vec{R}^{\text{eq}}\}_k) \right|^2 \right].$$

In practice, we truncate the expansion in Eq. (6) at the trimer level ($k = 2$). This captures the dominant non-additive effects while remaining computationally feasible. As illustrated in Fig. 2, this level of description is already sufficient to reveal clear signatures of non-additivity: trimer lines deviate from additive dimer sums [panel (a)], and their resonance energies can even exceed those of polarons [panel (b)]. The latter inversion of the spectral hierarchy highlights the distinct many-body dressing of molaron and polaron states, a phenomenon absent in systems with purely additive interactions. Moreover, as the polaron's symmetry matches that of the zero-temperature BEC wavefunction, it couples far more strongly to the bath than any molaron, producing the intense, broadened spectral feature observed in Fig. 2(a).

This framework captures non-additive effects on polaron and molaron structures (further details of this derivation and on the orthogonality of states are given in the End Matter and Supplemental Material [68]) and treats each molecular state independently, mirroring absorption spectroscopy experiments where a fixed-energy laser excites a single molecule at its corresponding energy in regions that permit its formation,

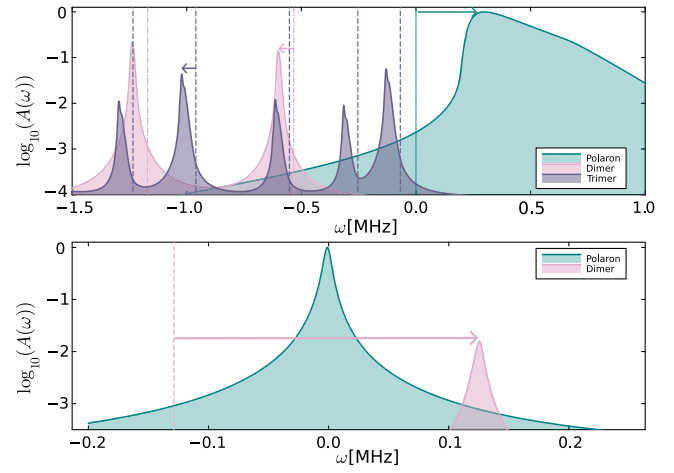


FIG. 2. Absorption spectra for a Rydberg impurity in $\text{Rb}|25p\rangle$. The shifts from the bare resonances (vertical lines) to the observed peak positions, induced by many-body dressing, are indicated by arrows. (a) At density $\rho = 2 \times 10^{15} \text{ cm}^{-3}$ and scattering length $a_s = -0.1a_0$, trimer peaks exhibit smaller binding energies compared to dimers, deviating from additive dimer sums and highlighting nonadditive effects. (b) At $\rho = 1 \times 10^{13} \text{ cm}^{-3}$ and $a_s = -0.028a_0$, the dimer branch crosses a resonance and consequently lies above the polaron branch, highlighting the nonadditivity in the many-body dressing. Both panels employ a logarithmic intensity scale.

such as a specific trimer configuration. However, to accurately capture quench dynamics, higher order processes such as a dimer scattering into a trimer state would need to be included. We expect that incorporating these processes will close the gaps in the parameter scans and mitigate the abrupt spectral changes at resonances evident in Fig. 2.

As the density increases, the molecular features become densely spaced and highly occupied such that any individual peaks are washed out [23,69]. In this limit, we reach a classical regime amenable to treatment via Monte Carlo sampling of the interaction potential [70,71], which has been shown to reproduce the distributions of deeply bound molecular states with high accuracy [57]. We have seen that such a limit is reached even once the typical number of particles within the Rydberg orbit reaches $k = 5$, signaling that our truncation to trimers is sufficient to capture the dominant quantum nonadditive features.

Although our full nonadditive model already includes anisotropic interactions, their subtle fingerprints can be easily obscured. To highlight the role of anisotropic interaction individually, we add a magnetic term $B\hat{L}_z$ in Eq. (1) to lift the m -level degeneracy. The $(2l + 1)$ resulting BO potentials are anisotropic but, as they are uncoupled, become additive. To calculate the interacting eigenstates needed to evaluate the Loschmidt echo [23,25], the two-body nuclear wave function is expanded into spherical harmonics $\psi(\vec{R}) = R^{-1} \sum_{L,M} \chi_{LM}(R) Y_{LM}(\Omega)$, which leaves the set of coupled radial Schrödinger equations

$$0 = \left(-\frac{1}{2\mu} \frac{d^2}{dR^2} + \frac{L(L+1)}{2\mu R^2} - \varepsilon \right) \chi_{LM}(R) + \sum_{L',M'} 2\pi a_s \underbrace{\left| \frac{u_{nl}(R)}{R} \right|^2 \langle LM | Y_{lm}^* Y_{lm} | L'M' \rangle}_{V_{L,L'}} \chi_{L'M}(R) \quad (7)$$

to be solved. The contributions of the potential matrix $V_{L,L'}$ [Fig. 3(b)] show the characteristic oscillations of the Rydberg potential persisting in all channels as well as the effect of the centrifugal barrier on higher partial wave contributions.

The corresponding absorption spectrum—for the specific example of an $\ell = 1$ Rydberg state—is shown in Fig. 3(c). As with nonadditive interactions, a shifted bare-atom resonance indicates the emergence of a predominantly repulsive polaron. However, owing to the additivity of the interaction, the spectrum displays molaron features at larger detunings whose energies are multiples of the underlying dimer levels—each two-body bound state can be occupied independently [24,69,72–74]. While the polaron energy for isotropic interactions can be captured within a mean-field description using only the Rydberg atom-boson s -wave scattering length from a single-channel calculation [25]—corresponding only to the $L = 0, M = 0$ term in Eq. (7)—this fails for anisotropic potentials.

We instead employ a comprehensive multichannel scattering treatment to obtain the K matrix [75], whose

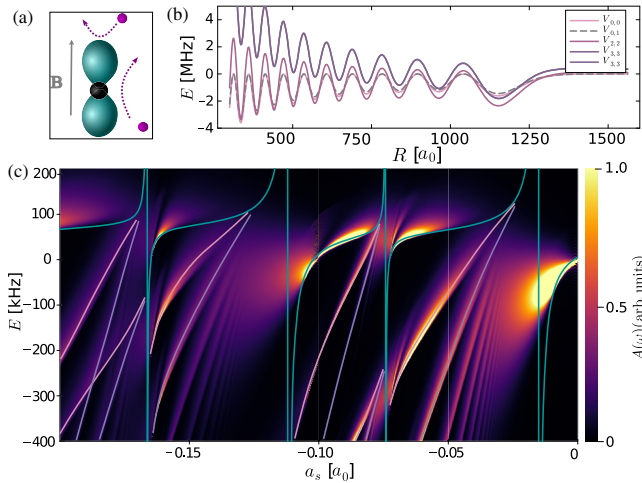


FIG. 3. (a) Sketch of a Rydberg atom aligning in a magnetic field. (b) Partial wave components of the anisotropic Rydberg p -state impurity potential in Eq. (7), highlighting its oscillations and centrifugal barriers. (c) The absorption spectrum of the Rb Rydberg atom in $|25p, m = 0\rangle$ within a BEC of density $\rho = 7 \times 10^{14} \text{ cm}^{-3}$ exhibits additive behavior in the molaron features: their energies are additive, and all states share the same many-body dressing, as reflected in the identical resonance structure. The dimer (pink) and trimer (purple) molarons are marked similar to Fig. 1. The role of anisotropy emerges only in the detailed form of the many-body dressing, which can be captured by the mean-field polaron energy with the effective scattering length [Eq. (9)], shown as the teal line.

diagonal $L = 0, M = 0$ element $K_{00,00}(\varepsilon)$ contains contributions from all scattering channels. By extracting this element and converting it into a zero-energy scattering length via the relation

$$a_{\text{Ryd}} = \frac{K_{00,00}(\varepsilon)}{\sqrt{2\mu\varepsilon}} \bigg|_{\varepsilon \rightarrow 0}, \quad (8)$$

we determine the mean field polaron energy

$$E_{\text{pol}} = \frac{2\pi}{\mu} \rho \cdot a_{\text{Ryd}}, \quad (9)$$

appropriate for pointlike interactions [76]. As the interaction potential deepens and bound states emerge, this mean-field polaron energy exhibits resonances aligning with those visible in the absorption spectrum [Fig. 3(c)]. Because of channel coupling, a bath particle in the $L = 0$ scattering channel can effectively tunnel through the centrifugal barriers of higher- L channels. This coupling induces resonances, belonging to radially as well as angularly excited bound states, to appear in the s -wave scattering matrix element, analogous to Feshbach resonances. As the density increases, the response of the anisotropic Rydberg impurity to the many-body dressing is qualitatively similar to the isotropic case [15,25,77].

In summary, we have developed a systematic partitioning scheme that constructs the full many-body response of a bosonic medium to a long-range impurity interaction from few-body clusters. We demonstrated this general approach with a Rydberg impurity as a controllable platform for illustrating effects arising from non-additive and anisotropic impurity-bath interactions. While Rydberg $l = 0$ states reproduce the familiar additive, spherically symmetric limit, higher- l manifolds such as the $l = 1$ case examined in detail here enable controlled exploration of direction-dependent and nonadditive coupling—phenomena well known in Feshbach-resonance physics but not previously addressed in ultracold impurity systems. The Rydberg-bath interactions can be tuned with the principal quantum number n as a discrete control knob, changing the depth and range of the interaction potential, allowing for control over the positions of molecular bound states and hence the Rydberg-bath scattering length.

The formalism developed here could be applied to the standard Bose polaron problem, where nonadditivity arises from both interparticle interactions and the details of the Feshbach resonance itself. It also offers a natural path toward treating dipolar impurities, such as polar molecules in a dipolar BEC, where internal state degeneracy and anisotropic interactions play a central role. More generally, the role of anisotropy in bipolaron formation—where the interactions are mediated by the environment particles, and reflect the anisotropic response to the impurity—can be considered.

Acknowledgments—We thank P. Giannakeas, A. Christianen, and T. A. Yogurt for valuable discussions. A. A. T. D. is grateful to the DFG Priority Program No. SPP 1929 (GiRyd) for funding an exchange visit to ITAMP. S. T. R. acknowledges support from the National Science Foundation through Grants No. NSF PHY-2409110 and No. PHY-2409111 and NSF support through ITAMP.

Data availability—There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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- [1] R. Schmidt, M. Knap, D. A. Ivanov, J.-S. You, M. Cetina, and E. Demler, *Rep. Prog. Phys.* **81**, 024401 (2018).
 - [2] Y. E. Shchadilova, R. Schmidt, F. Grusdt, and E. Demler, *Phys. Rev. Lett.* **117**, 113002 (2016).
 - [3] L. A. Pena Ardila and S. Giorgini, *Phys. Rev. A* **94**, 063640 (2016).
 - [4] L. A. Pena Ardila and S. Giorgini, *Phys. Rev. A* **92**, 033612 (2015).
 - [5] J. Levinsen, L. A. Pena Ardila, S. M. Yoshida, and M. M. Parish, *Phys. Rev. Lett.* **127**, 033401 (2021).
 - [6] P. Massignan, N. Yegovtsev, and V. Gurarie, *Phys. Rev. Lett.* **126**, 123403 (2021).
 - [7] S. M. Yoshida, S. Endo, J. Levinsen, and M. M. Parish, *Phys. Rev. X* **8**, 011024 (2018).
 - [8] C. Baroni, B. Huang, I. Fritsche, E. Dobler, G. Anich, E. Kirilov, R. Grimm, M. A. Bastarrachea-Magnani, P. Massignan, and G. M. Bruun, *Nat. Phys.* **20**, 68 (2024).
 - [9] B. J. DeSalvo, K. Patel, G. Cai, and C. Chin, *Nature (London)* **568**, 61 (2019).
 - [10] N. B. Jørgensen, L. Wacker, K. T. Skalmstang, M. M. Parish, J. Levinsen, R. S. Christensen, G. M. Bruun, and J. J. Arlt, *Phys. Rev. Lett.* **117**, 055302 (2016).
 - [11] M.-G. Hu, M. J. Van De Graaff, D. Kedar, J. P. Corson, E. A. Cornell, and D. S. Jin, *Phys. Rev. Lett.* **117**, 055301 (2016).
 - [12] M. G. Skou, T. G. Skov, N. B. Jørgensen, K. K. Nielsen, A. Camacho-Guardian, T. Pohl, G. M. Bruun, and J. J. Arlt, *Nat. Phys.* **17**, 731 (2021).
 - [13] C. Kohstall, M. Zaccanti, M. Jag, A. Trenkwalder, P. Massignan, G. M. Bruun, F. Schreck, and R. Grimm, *Nature (London)* **485**, 615 (2012).
 - [14] A. Schirotzek, C.-H. Wu, A. Sommer, and M. W. Zwierlein, *Phys. Rev. Lett.* **102**, 230402 (2009).
 - [15] E. R. Christensen, A. Camacho-Guardian, and G. M. Bruun, *Phys. Rev. Lett.* **126**, 243001 (2021).
 - [16] G. E. Astrakharchik, L. A. P. Ardila, R. Schmidt, K. Jachymski, and A. Negretti, *Commun. Phys.* **4**, 94 (2021).
 - [17] R. Pessoa, S. A. Vitiello, and L. A. Pena Ardila, *Phys. Rev. Lett.* **133**, 233002 (2024).
 - [18] P. Massignan, C. J. Pethick, and H. Smith, *Phys. Rev. A* **71**, 023606 (2005).
 - [19] T. A. Yoğurt and M. T. Eiles, *Phys. Rev. A* **112**, 063313 (2025).
 - [20] P. Wysocki, M. Tylutki, and K. Jachymski, *New J. Phys.* **28**, 013201 (2026).
 - [21] Laurent H. A. Simons, M. Wouters, and J. Tempere, *Phys. Rev. A* **110**, 043301 (2024).
 - [22] U. Cavazos Olivas, L. A. Peña Ardila, and K. Jachymski, *Phys. Rev. A* **110**, L011301 (2024).
 - [23] R. Schmidt, J. D. Whalen, R. Ding, F. Camargo, G. Woehl, S. Yoshida, J. Burgdörfer, F. B. Dunning, E. Demler, H. R. Sadeghpour, and T. C. Killian, *Phys. Rev. A* **97**, 022707 (2018).
 - [24] F. Camargo, R. Schmidt, J. D. Whalen, R. Ding, G. Woehl, S. Yoshida, J. Burgdörfer, F. B. Dunning, H. R. Sadeghpour, E. Demler, and T. C. Killian, *Phys. Rev. Lett.* **120**, 083401 (2018).
 - [25] A. A. T. Durst and M. T. Eiles, *Phys. Rev. Res.* **6**, L042009 (2024).
 - [26] J. Sous, H. R. Sadeghpour, T. C. Killian, E. Demler, and R. Schmidt, *Phys. Rev. Res.* **2**, 023021 (2020).
 - [27] R. Schmidt, H. R. Sadeghpour, and E. Demler, *Phys. Rev. Lett.* **116**, 105302 (2016).
 - [28] G. E. Astrakharchik, L. A. P. Ardila, K. Jachymski, and A. Negretti, *Nat. Commun.* **14**, 1647 (2023).
 - [29] C. Fey, M. Kurz, and P. Schmelcher, *Phys. Rev. A* **94**, 012516 (2016).
 - [30] A. Y. Chuang, H. Q. Bui, A. Christianen, Y. Zhang, Y. Ni, D. Ahmed-Braun, C. Robens, and M. W. Zwierlein, *Phys. Rev. X* **15**, 021098 (2025).
 - [31] J. F. E. Croft, C. Makrides, M. Li, A. Petrov, B. K. Kendrick, N. Balakrishnan, and S. Kotochigova, *Nat. Commun.* **8**, 15897 (2017).
 - [32] Y. Wang, M. Mirahmadi, and J. Pérez-Ríos, *Phys. Chem. Chem. Phys.* **26**, 7264 (2024).
 - [33] I. G. Kaplan, R. Santamaria, and O. Novaro, *Mol. Phys.* **84**, 105 (1995).
 - [34] W. E. Baylis, *J. Phys. B* **10**, L477 (1977).
 - [35] L.-Y. Tang, Z.-C. Yan, T.-Y. Shi, J. F. Babb, and J. Mitroy, *J. Chem. Phys.* **136**, 104104 (2012).
 - [36] L. C. Balling and J. J. Wright, *J. Chem. Phys.* **79**, 2941 (1983).
 - [37] J. Levinsen, T. G. Tiecke, J. T. M. Walraven, and D. S. Petrov, *Phys. Rev. Lett.* **103**, 153202 (2009).
 - [38] G. Quémener, P. Honvault, J.-M. Launay, P. Soldán, D. E. Potter, and J. M. Hutson, *Phys. Rev. A* **71**, 032722 (2005).
 - [39] A. G. Volosniev, G. Bighin, L. Santos, and L. A. Peña Ardila, *SciPost Phys.* **15**, 232 (2023).
 - [40] J. Stuhler, A. Griesmaier, T. Koch, M. Fattori, T. Pfau, S. Giovanazzi, P. Pedri, and L. Santos, *Phys. Rev. Lett.* **95**, 150406 (2005).
 - [41] B. Kain and H. Y. Ling, *Phys. Rev. A* **89**, 023612 (2014).
 - [42] T. Lahaye, C. Menotti, L. Santos, M. Lewenstein, and T. Pfau, *Rep. Prog. Phys.* **72**, 126401 (2009).
 - [43] J. Sánchez-Baena, L. A. Pena Ardila, G. E. Astrakharchik, and F. Mazzanti, *Phys. Rev. A* **110**, 023317 (2024).
 - [44] Z. Yan, P. B. Patel, B. Mukherjee, R. J. Fletcher, J. Struck, and M. W. Zwierlein, *Phys. Rev. Lett.* **122**, 093401 (2019).
 - [45] Z. Z. Yan, Y. Ni, C. Robens, and M. W. Zwierlein, *Science* **368**, 190 (2020).
 - [46] R. Schmidt and M. Lemesko, *Phys. Rev. Lett.* **114**, 203001 (2015).
 - [47] R. Schmidt and M. Lemesko, *Phys. Rev. X* **6**, 011012 (2016).

- [48] E. Yakaboylu, B. Midya, A. Deuchert, N. Leopold, and M. Leshchko, *Phys. Rev. B* **98**, 224506 (2018).
- [49] Z. Zeng, E. Yakaboylu, M. Leshchko, T. Shi, and R. Schmidt, *J. Chem. Phys.* **158**, 134301 (2023).
- [50] Y.-Y. Liu, Y. Cui, X.-Z. Zhang, R.-B. Yang, Z.-Q. Li, and Z.-W. Wang, [arXiv:2305.03473](https://arxiv.org/abs/2305.03473).
- [51] M. T. Eiles, *J. Phys. B* **52**, 113001 (2019).
- [52] M. Kurz and P. Schmelcher, *J. Phys. B* **47**, 165101 (2014).
- [53] A. T. Krupp, A. Gaj, J. B. Balewski, P. Ilzhöfer, S. Hofferberth, R. Löw, T. Pfau, M. Kurz, and P. Schmelcher, *Phys. Rev. Lett.* **112**, 143008 (2014).
- [54] M. Mayle, S. T. Rittenhouse, P. Schmelcher, and H. R. Sadeghpour, *Phys. Rev. A* **85**, 052511 (2012).
- [55] I. Lesanovsky, P. Schmelcher, and H. R. Sadeghpour, *J. Phys. B* **39**, L69 (2006).
- [56] Note that here we consider only Rydberg levels possessing finite quantum defects, where the only relevant degeneracy is that of the magnetic sublevels. Rydberg states of a hydrogen atom also possess degenerate angular momentum levels, producing further nonadditive and anisotropic interactions.
- [57] C. Fey, J. Yang, S. T. Rittenhouse, F. Munkes, M. Baluktsian, P. Schmelcher, H. R. Sadeghpour, and J. P. Shaffer, *Phys. Rev. Lett.* **122**, 103001 (2019).
- [58] T.-D. Lee and D. Pines, *Phys. Rev.* **92**, 883 (1953).
- [59] While bath-bath interactions induced by this transformation are known to vanish for spherically symmetric interactions [23], their angular correlations in the presence of an anisotropic interaction are not in general zero. However, the large spatial extent of the Rydberg atom and the correspondingly small rotational constant allow us to neglect angular contributions to the induced interactions to a good approximation.
- [60] C. H. Greene, A. S. Dickinson, and H. R. Sadeghpour, *Phys. Rev. Lett.* **85**, 2458 (2000).
- [61] J. P. Shaffer, S. T. Rittenhouse, and H. R. Sadeghpour, *Nat. Commun.* **9**, 1965 (2018).
- [62] F. B. Dunning, S. K. Kanungo, and S. Yoshida, *J. Phys. B* **57**, 212002 (2024).
- [63] T. F. Gallagher, *Rydberg Atoms*, Cambridge Monographs on Atomic, Molecular and Chemical Physics (Cambridge University Press, Cambridge, England, 1994).
- [64] E. Fermi, *Il Nuovo Cimento* **11**, 157 (1934).
- [65] Y. E. Shchadilova, F. Grusdt, A. N. Rubtsov, and E. Demler, *Phys. Rev. A* **93**, 043606 (2016).
- [66] N. Mostaan, N. Goldman, and F. Grusdt, *Phys. Rev. Res.* **7**, 043349 (2025).
- [67] O. K. Diessel, J. Von Milczewski, A. Christianen, and R. Schmidt, *Phys. Rev. Res.* **6**, 023239 (2024).
- [68] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/38vj-6fqc> for additional derivations, numerical details, and convergence tests.
- [69] M. Schlagmüller, T. C. Liebisch, H. Nguyen, G. Lohead, F. Engel, F. Böttcher, K. M. Westphal, K. S. Kleinbach, R. Löw, S. Hofferberth, T. Pfau, J. Pérez-Ríos, and C. H. Greene, *Phys. Rev. Lett.* **116**, 053001 (2016).
- [70] M. T. Eiles, J. Pérez-Ríos, F. Robicheaux, and C. H. Greene, *J. Phys. B* **49**, 114005 (2016).
- [71] A. L. Hunter, M. T. Eiles, A. Eisfeld, and J. M. Rost, *Phys. Rev. X* **10**, 031046 (2020).
- [72] V. Bendkowsky, B. Butscher, J. Nipper, J. B. Balewski, J. P. Shaffer, R. Löw, T. Pfau, W. Li, J. Stanojevic, T. Pohl, and J. M. Rost, *Phys. Rev. Lett.* **105**, 163201 (2010).
- [73] A. Gaj, A. T. Krupp, J. B. Balewski, R. Löw, S. Hofferberth, and T. Pfau, *Nat. Commun.* **5**, 4546 (2014).
- [74] F. Engel, T. Dieterle, F. Hummel, C. Fey, P. Schmelcher, R. Löw, T. Pfau, and F. Meinert, *Phys. Rev. Lett.* **123**, 073003 (2019).
- [75] M. Aymar, C. H. Greene, and E. Luc-Koenig, *Rev. Mod. Phys.* **68**, 1015 (1996).
- [76] G. D. Mahan, *Many-Particle Physics* (Springer, Boston, MA, 2000).
- [77] The absorption spectra of the anisotropic Rydberg polaron at different densities are shown in Supplemental Material [68].

End Matter

Appendix A: Derivation of nonadditive $S(t)$ —We consider an N -particle Hilbert space $\mathcal{H}^{(N)}$ that can be decomposed into subspaces with exactly k particles confined to subsystem B and the remaining $N - k$ particles confined to subsystem C . Formally,

$$\mathcal{H}^{(N)} = \bigoplus_{k=0}^N \mathcal{H}_B^{(k)} \otimes \mathcal{H}_C^{(N-k)}, \quad (\text{A1})$$

where each subspace $\mathcal{H}_B^{(k)}$ ($\mathcal{H}_C^{(N-k)}$) represents a fully correlated Hilbert space of exactly k ($N - k$) indistinguishable particles restricted to subsystem B (C), with no assumption of single-particle factorization. We can introduce the orthogonal projector

$$P_k = \frac{1}{k!(N-k)!} \sum_{\sigma \in S_N} U(\sigma) [\mathbb{1}_B^{(k)} \otimes \mathbb{1}_C^{(N-k)}], \quad (\text{A2})$$

where $\mathbb{1}_B^{(k)}$ denotes the identity operator acting on the fully correlated subspace where exactly k particles reside in subsystem B . Similarly, $\mathbb{1}_C^{(N-k)}$ acts on the correlated subspace with exactly $N - k$ particles in subsystem C . To enforce indistinguishability, we sum over all particle permutations using the operator $U(\sigma)$, which rearranges particles among subsystems.

Since the initial state $\Psi_0(\{\vec{R}\}_N) = \prod_i^N \psi_0(\vec{R}_i)$ is a product state, it lies in the totally symmetric space $(\mathcal{H}^{(1)})^{\otimes N}$. Thus, the Loschmidt echo reads

$$S(t) = \langle \Psi_0 | e^{iN\epsilon_0 t} e^{-i\hat{H}_N t} | \Psi_0 \rangle, \quad (\text{A3})$$

with

$$\hat{H}_0 | \Psi_0 \rangle = N\epsilon_0 | \Psi_0 \rangle. \quad (\text{A4})$$

Inserting the completeness relation $\sum_{k=0}^N P_k = 1$ yields

$$S(t) = \sum_{k=0}^N \underbrace{\langle \Psi_0 | e^{iN\epsilon_0 t} P_k e^{-i\hat{H}_N t} | \Psi_0 \rangle}_{s_k(t)} = \sum_{k=0}^N s_k(t). \quad (\text{A5})$$

This decomposition is the most general exact form of the autocorrelation function. We can, however, exploit the fact that the remaining $N - k$ particles in the continuum still interact independently with the impurity, allowing us to decompose the continuum into single-particle contributions. Because the k -body molecular state $|\Psi_{\alpha,\gamma}^{(k)}\rangle_B$ cannot be represented as product states, grouping these continuum single-particle states $|\psi_{\alpha,\gamma,\gamma}\rangle_C$ into k -body clusters $|\Psi_0^{(k)}\rangle_C$ allows for a fully orthonormal basis. We can therefore write the full many-body Hilbert space as

$$\mathcal{H}^{(N)} \approx \mathcal{H}_B^{(k)} \otimes [\mathcal{H}_C^{(k)}]^{\otimes \lfloor \frac{N-k}{k} \rfloor}. \quad (\text{A6})$$

For small cluster sizes k , the approximation of rounding $(N - k/k)$ becomes asymptotically exact in the large- N limit. By writing the initial state as a product of k -particle clusters

$$|\Psi_0\rangle = |\Psi_0^{(k)}\rangle_B \otimes (|\Psi_0^{(k)}\rangle_C)^{\otimes \lfloor \frac{N-k}{k} \rfloor}, \quad (\text{A7})$$

the Loschmidt echo simplifies considerably to

$$s_k(t) = e^{iN\epsilon_0 t} \binom{N}{k} \sum_{\alpha} \left[\left| \langle \Psi_0^{(k)} | e^{-i\hat{H}_B^{k,\alpha} t} | \Psi_0^{(k)} \rangle_B \right|^2 \times \sum_{\gamma} \left(\left| \langle \psi_0 | e^{-i\hat{H}_C^{k,\alpha,\gamma} t} | \psi_0 \rangle_C \right|^2 \right)^{\frac{N-k}{k}} \right]. \quad (\text{A8})$$

Until now, we have endeavored to keep the description of this framework for computing the many-body response of a Bose-Einstein condensate to an impurity with nonadditive interactions generic to any type of impurity. Of course, the specific forms of the different bound-state and continuum Hamiltonians depend on the impurity and microscopic details of its interaction with the bath and formation of two-body, three-body, and larger clusters. Therefore, the next Section details the molecular Hamiltonians and approximations we have employed in describing the few-body sector specific to the Rydberg impurity.

Appendix B: Hamiltonian of Rydberg impurity—We analyze the Rydberg impurity using an adiabatic BO framework that captures both the strong perturbations of a few atoms inside the Rydberg orbit and the weak perturbations of the surrounding bath. This is done by projecting the Hamiltonian Eq. (1) into the different k -subspaces via $\hat{P}_k \hat{H} \hat{P}_k$. This allows us to treat the k atoms forming a $(k + 1)$ -mer as a strong perturbation

on the electronic state, while the remaining $N - k$ atoms remaining in the continuum interact with the effective molecule-bath potential created by the bound complex.

The bound-state Hamiltonian for the $(k + 1)$ -mer is

$$\hat{H}_B^{k,\alpha} = - \sum_{i=1}^k \frac{\nabla_{\vec{R}_i}^2}{2\mu} + V_{\alpha}(\{\vec{R}\}_k), \quad (\text{B1})$$

where the BO potentials V_{α} are obtained by diagonalizing Eq. (2) for the k -particle configuration. The corresponding electronic eigenstates are

$$\phi_{\alpha,\gamma}(\vec{r}; \{\vec{R}\}_k) = \frac{u_{nl}(r)}{r} \sum_{m=-l}^l w_{\alpha,m}(\{\vec{R}\}_k) Y_{lm}(\hat{r}), \quad (\text{B2})$$

where $w_{\alpha m}^k$ are the expansion coefficients. Because of the m -level degeneracy, these BO surfaces depend on all $\{\vec{R}\}_k$, and in general there are $\max(2l + 1, k)$ nonzero surfaces. We solve $\hat{H}_B^{k,\alpha}$ for each potential surface to obtain the γ th vibrational state of the molecular wave function $|\Psi_{\alpha,\gamma}^{(k)}\rangle_B$. This state has energy $E_{k,\alpha,\gamma}$, which makes up the dominant contribution to the molaron energy for this configuration.

Each bound molecule interacts with the surrounding bath of $N - k$ particles in a manner that depends on its specific internal configuration. In the adiabatic picture, the Rydberg electronic wave function mediates the interaction between the Rydberg atom and the bath particles. This mediated interaction depends sensitively on the explicit electronic state, which in turn is determined by the nuclear coordinates $\{\vec{R}\}_k$ of the bound complex. To capture this dependence, we determine the equilibrium geometry of each molecular state,

$$\{\vec{R}_{\alpha,\gamma}^{\text{eq}}\}_k = {}_B \langle \Psi_{\alpha,\gamma}^{(k)} | \{\vec{R}\}_k | \Psi_{\alpha,\gamma}^{(k)} \rangle_B, \quad (\text{B3})$$

which defines the most probable nuclear configuration of that bound state and thereby fixes the corresponding electronic state mediating its interaction with the continuum particles.

The weakly perturbing particles move then in the effective potential generated by the bound complex. This is described by the continuum Hamiltonian

$$\hat{H}_C^{k,\alpha,\gamma} = - \sum_{i=k+1}^N \frac{\nabla_{\vec{R}_i}^2}{2\mu} + 2\pi a_s \sum_{i=k+1}^N \left| \phi_{\alpha,\gamma} \left(\vec{R}_i; \{\vec{R}^{\text{eq}}\}_k \right) \right|^2. \quad (\text{B4})$$

For $2l + 1 > k$, there exist $2l + 1 - k$ electronic states that do not support a $(k + 1)$ -mer bound state; for these, the adiabatic separation into $\hat{H}_C$ and $\hat{H}_B$ is ill defined, and within our approximation we retain only the states associated with bound molecules.