

Phenomenology of a Rydberg impurity in an ideal Bose-Einstein condensate

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We investigate the absorption spectrum of a Rydberg impurity immersed in and interacting with an ideal Bose-Einstein condensate. Here, the impurity-bath interaction range can greatly exceed the mean interparticle distance; this discrepancy in length scales challenges the assumptions underlying the universal aspects of impurity atoms in dilute bosonic environments. Our analysis finds three distinct parameter regimes, each characterized by a unique spectral response. In the low-density regime, we find that the Rydberg impurity is dressed by the surrounding bath similarly to the known Bose polaron. Transitioning to intermediate densities, the impurity response, given by sharp quasiparticle peaks, fragments into an intricate pattern bearing the hallmarks of a diverse molecular structure. Finally, at high density, a universal Gaussian response emerges as the statistical nature of the bath dominates its quantum dynamics. We complement this analysis with a study of an ionic impurity, which behaves equivalently. Our exploration offers insights into the interplay between interaction range, density, and many-body behavior in impurity systems.

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Polarons, quasiparticles that form when impurities are dressed by the collective response of a surrounding medium, are observed when few-body correlations and collective excitations dramatically alter particle dynamics. Originally introduced to describe the emergent behavior of electrons interacting with phonons in a solid [1–3], polaron theory now underpins our understanding of numerous fundamental phenomena, such as transport processes [4–6], ³He atoms in superfluid ⁴He [7], and ultracold mixtures [8–16]. However, even the dynamics of a single impurity immersed in a non-interacting bath poses a significant challenge to theoretical description, despite the apparent simplicity of the Hamiltonian

$$\hat{H} = \sum_k \frac{k^2}{2\mu} \hat{b}_k^\dagger \hat{b}_k + \sum_{k,q} V(q) \hat{b}_{k+q}^\dagger \hat{b}_k. \quad (1)$$

In Eq. (1), which is written in atomic units and in a frame centered on the zero-momentum impurity, $\hat{b}_k^\dagger$ and $\hat{b}_k$ denote the bath creation and annihilation operators at momentum k , $V(q)$ is the interspecies interaction, and μ is the reduced mass of the impurity and a bath atom [17–19]. $\hat{H}$ is commonly used to describe dilute gases where the mean interparticle distance $\rho^{-1/3}$, where ρ is the density, greatly exceeds the range of the potential $V(r)$. This justifies its replacement by a zero-range pseudopotential proportional to the bath-impurity scattering

length a [20]. Within this approximation, the physics of the system becomes universal, depending only on the scattering length, the dimensionality of the system, and the density and quantum statistics of the bath [21–31]. Measurements of repulsive and attractive polaron quasiparticles and weakly bound molecules in ultracold gases have provided strong evidence for this universal behavior [31–42].

However, this universality is not expected to apply when the interaction range is comparable to the typical interparticle distance. Such is the case for a Rydberg impurity, where the highly excited Rydberg electron with principal quantum number n mediates the impurity-bath interaction by scattering off of the bath particles. The s -wave electron-atom scattering length a_s (see Fig. 1) determines the overall strength of this interaction [43–45]. For a Rydberg $|nS\rangle$ state this leads to the isotropic potential

$$V_{\text{Ryd}}(r) = 2\pi a_s |\psi_{n00}(r)|^2, \quad (2)$$

which, unlike a zero-range potential, can in principle support several bound states [46–52]. The appearance of the Rydberg wave function, $\psi_{n00}(r)$, causes the range R_0 and depth V_0 of this highly oscillatory potential to vary as n^2 and n^{-6} , respectively [50]. Recently, the ultracold toolbox has been expanded to include other impurity systems with finite-ranged interactions, such as dipolar atoms [53–55] and ion-atom mixtures [56–59]. These are rarely compatible with the zero-range approximation and its universality and raise the question of whether or not a unified description of a finite-ranged impurity in a quantum environment exists.

In this Letter, we approach this question through an exploration of the behavior of a Rydberg impurity interacting with an ideal Bose-Einstein condensate (BEC) at zero temperature. Previous studies [60–63] claimed that such an impurity has qualitatively different properties than those known from the zero-range approximation. However, by additionally

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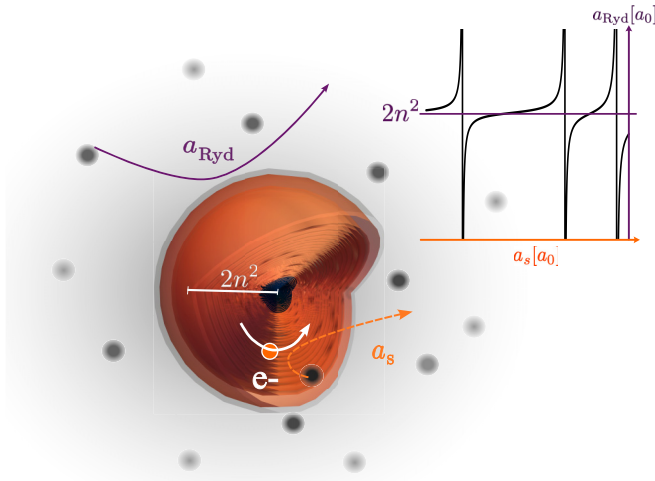


FIG. 1. Two scattering lengths characterize the interaction of a Rydberg impurity with its environment. Atoms probing the interior of the Rydberg atom collide with the highly excited electron directly; these interactions are characterized by the atom-electron scattering length a_s . In contrast, distant atoms interact with the Rydberg atom as a single entity and the atom-impurity scattering length a_{Ryd} is characteristic of this interaction.

considering an ionic impurity and studying a full parameter range for both systems, we show that different impurities share the same spectral response dictated by the dimensionless quantity $a\rho^{1/3}$. Even though the spectral responses—containing narrow polaron or dressed molecule (“molaron”) peaks as well as broad features indicating the participation of many bath particles—become structurally more complicated, each can be described independently of the type of impurity. This leads to an extension of the universal description of Bose polarons to a broader class of interactions. With this, we establish a common language for the phenomena observed in Bose-polaron physics and unify the disparate interpretations provided by the many approaches developed for such impurity problems. These include the field-theoretical quasiparticle methods describing polaron physics, the few-body picture of molecular physics, and semiclassical methods originating in the theory of pressure broadening.

To investigate and characterize the universal aspects of the Rydberg impurity, we performed a detailed numerical study of the absorption spectrum $A(\omega)$. This is obtained from the Fourier transform of the autocorrelation function

$$S(t) = \langle e^{i\hat{H}_0 t} e^{-i\hat{H} t} \rangle = \left(\sum_{\alpha} e^{i(\epsilon_0 - \omega_{\alpha})t} |\langle 0 | \alpha \rangle|^2 \right)^N, \quad (3)$$

where the expectation value is taken with respect to the noninteracting BEC state (the ground state of $\hat{H}_0$) of N bosons [62]. Since the ideal BEC is in a pure product state, the final expression of the many-body response only requires the eigenstates ($|0\rangle$), $|\alpha\rangle$ and energies (ϵ_0), ω_{α} of the (non-)interacting two-body Hamiltonian of the Rydberg atom and a single boson. We solve for the two-body physics by employing the eigenchannel R -matrix method [64], which yields the energy-dependent logarithmic derivative of the scattering wave function $|\alpha\rangle$ for $r > R_0$. This allows us to efficiently calculate thousands of box-continuum states, molecular bound

states, and the zero-energy scattering length a_{Ryd} . Figure 1 shows $a_{\text{Ryd}}(a_s)$, which is centered around the interaction range $R_0 \sim 2n^2$ and is typically one to two orders of magnitude larger than the scattering length between two ground state atoms. We study a ^{84}Sr Rydberg impurity with $n = 50$ as an illustrative and generic example and compute $A(\omega)$ as a function of a_s [65]. In this way we adjust V_0 independent of R_0 [66].

Figure 2 shows $A(\omega)$ when $R_0\rho^{1/3} \ll 1$. In this regime, we recover all of the features known from the limit of a zero-range impurity [9,27,67]. When $a_s > -0.06a_0$, V_{Ryd} does not support a two-body bound state and $A(\omega)$ exhibits a single peak at negative energy indicating the formation of an attractive polaron. The mean-field energy of the zero-range approximation of the Rydberg potential, $E_{\text{Zr}} = 2\pi a_{\text{Ryd}}\rho/\mu$, describes the position of this feature. However, this description fails dramatically near a scattering resonance. The mean-field description of the full Rydberg potential, $E_{\text{Ryd}} = \rho \int V_{\text{Ryd}}(r) d^3r = 2\pi a_s \rho / m_e$ [44,62,68], follows the center of spectral weight smoothly across unitarity, even as the spectral feature diffuses and cannot be associated with a well-defined quasiparticle [9,69]. Using the Born approximation for the Rydberg scattering length, $a_{\text{Ryd}}^{\text{B}} = a_s \mu / m_e$, one can rewrite $E_{\text{Ryd}} = 2\pi a_{\text{Ryd}}^{\text{B}} \rho / \mu$ to have the same structure as E_{Zr} .

To the red of the resonance, E_{Zr} again describes the brightest spectral feature, which is located at positive energy and therefore identified as a repulsive polaron. Below this feature lies a series of negative energy peaks associated with the molecular bound state, which can be multiply occupied to form dimers, trimers, and the like. These interact with the residual bath through the same Rydberg interaction as the bare atom and as a result they are dressed by bath excitations in the same fashion. Each molecule therefore possesses some quasiparticle character inherited from the repulsive polaron and forms a “molaron” with a binding energy shifted from that of the bare dimer, E_b , to $E_b + E_{\text{Zr}}$ [70–73].

Figure 3(a) displays $A(\omega)$ over a broader range of a_s and at ten times the density of Fig. 2, which causes the molaron peaks to accumulate appreciable spectral weight as

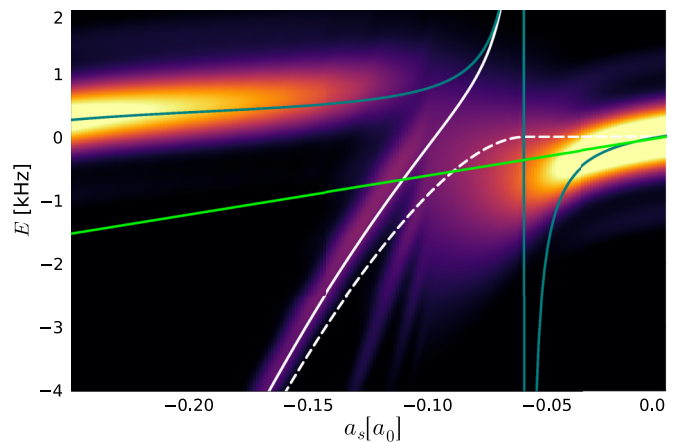


FIG. 2. $A(\omega)$ of a $50S$ Rydberg impurity in a BEC with $\rho = 10^{12} \text{ cm}^{-3}$. The mean-field energy shifts E_{Zr} (teal) and E_{Ryd} (green), as well as the bare and dressed dimer energies E_b (dashed white) and $E_b + E_{\text{Zr}}$ (solid white) are overlaid.

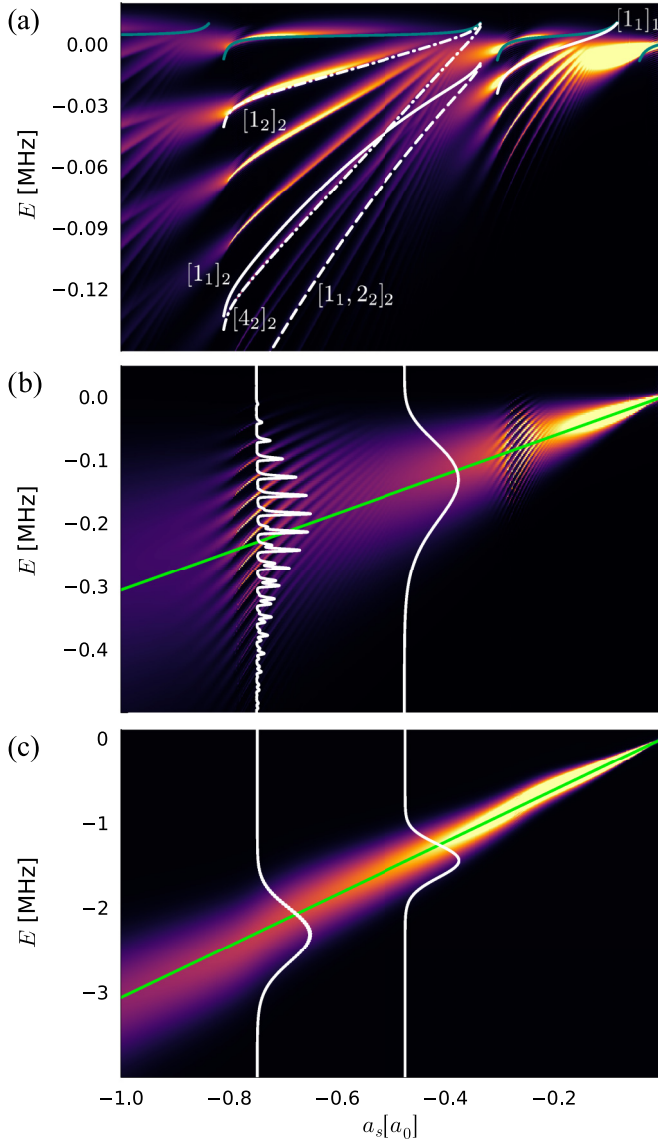


FIG. 3. $A(\omega)$ of a 50S Rydberg impurity in a BEC with (a) $\rho = 10^{13} \text{ cm}^{-3}$, (b) $\rho = 5 \times 10^{13} \text{ cm}^{-3}$, and (c) $\rho = 5 \times 10^{14} \text{ cm}^{-3}$. In (a), the mean-field energy E_x (teal) is shown alongside the mean-field energies of various molaron states. In panels (b) and (c) cuts of $A(\omega)$ at fixed a_s (white curves) show the line shape more clearly. E_{Ryd} is shown in green.

$R_0 \rho^{1/3} \sim 1$. This reveals an intriguing internal structure due to the appearance of additional two-body bound states as the potential deepens. This structure suggests a nomenclature, the $[n_0, \dots, n_i, \dots, n_M]_M$ molaron, where M is the total number of two-body bound states supported by the potential and n_i is the bosonic occupation number of the i th bound state, with $i = 0$ representing the bare atom. The peak position of the $[0_0]_M$ molaron, i.e., the polaron, coincides with E_{zf} . Exemplary molaron peaks are labeled in Fig. 3(a). At every scattering resonance each quasiparticle peak undergoes the same fragmentation seen in Fig. 2. For example, at the second scattering resonance, each of the $[n_i]_1$ states broadens and eventually splits into a multitude of states $[n_i, m_{i+1}]_2$.

An important consequence of the large extent of the Rydberg potential is that a_{Ryd} is large and positive except close to a

resonance, where it dips below zero. As a result, the attractive polaron exists only in a very limited parameter space. At the transition from a repulsive to an attractive polaron, a_{Ryd} vanishes at a Ramsauer-Townsend zero [74,75]. Despite its nonzero interaction potential, the Rydberg atom effectively does not interact with the bath—the scattering phase shift vanishes. Here, the molaron features sharpen, losing quasiparticle weight as they more closely resemble bare molecules.

At higher density [Fig. 3(b)] the spectral weight shifts entirely into molaron states. Their absorption peaks blur together and $A(\omega)$ takes on a Gaussian profile with mean values E_{Ryd} whose emergence is explained by the central limit theorem [60,62,76]. However, the regression to a Gaussian distribution does not occur at the same density for each a_s : the distinct molaron peaks, still resolvable in the vicinity of the Ramsauer-Townsend zeros in Fig. 3(b), merge to form a Gaussian spectral profile only at higher density [Fig. 3(c)].

This progression from an individual Lorentzian at zero density through asymmetric line shapes and polymer formation to a symmetric Gaussian distribution at high density is largely consistent with the semiclassical theory of pressure broadening [68,77–84]. This theory predicts the occurrence of “satellite” peaks at integer multiples of the extrema of the interaction potential, an obvious parallel to the molaron structure, as well as their blending together into a Gaussian response as the number of particles within the range of the potential, $R_0^3 \rho$, increases [76,79]. Our calculation shows that a more accurate condition takes into account the zero-energy scattering length and thus generalizes the above condition to $\rho^{-1/3} \ll a_{\text{Ryd}}$.

This more accurate condition is depicted in Fig. 4, which shows a qualitative estimate of the quasiparticle weight Z given by a temporal average of $S(t)$ in the long time limit ($\sim 1000 \mu\text{s}$). This estimate is reasonable, as the finite Rydberg lifetime ($\sim 100 \mu\text{s}$) prevents $S(t)$ from decaying to zero for every a_s value as is expected for a noninteracting BEC [85–87]. The curve $a_{\text{Ryd}} \rho^{1/3} = 1$ indicates the transition between a

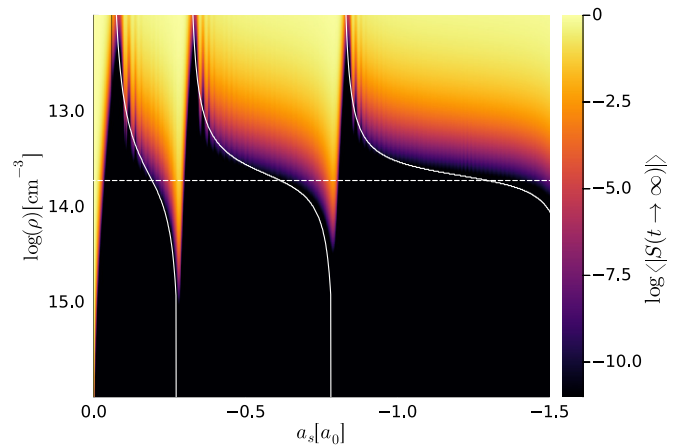


FIG. 4. Effective quasiparticle weight of the Rydberg impurity, calculated by averaging $S(t)$ over late times. The solid white curve shows $a_{\text{Ryd}} \rho^{1/3} = 1$, which demarcates the two phases: quasiparticle (molaron or polaron limit with $Z = 1$) and semiclassical/statistical ($Z = 0$). The dashed white line shows $R_0 \rho^{1/3} = 1$, the semiclassical transition.

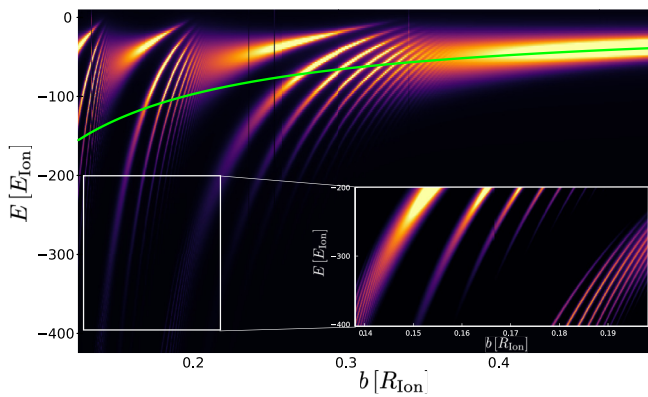


FIG. 5. $A(\omega)$ of an ionic impurity at $\rho = 2 R_{\text{Ion}}^{-3}$. The green line shows the mean-field energy shift E_{Ion} . The inset highlights the fragmentation of molaron states.

spectrum dominated by quasiparticle features [where $Z \gg 0$] to a purely statistical state without quasiparticle weight and characterized by significant broadening.

However, in the extreme limit $a_{\text{Ryd}} \rightarrow 0^{+/-}$, the above condition is never satisfied and $A(\omega)$ will show distinct peaks no matter the density. This explains the regions with large quasiparticle weight even at high densities seen in Fig. 4 where the impurity has a small but nonzero quasiparticle character due to its vastly reduced coupling to the dense environment.

In contrast, the region of approximately zero quasiparticle weight extends to very low densities when a_{Ryd} diverges. This corresponds to the parameter regime where the loss of quasiparticle characteristics and a broadening of the well-defined polaron peak into a diffuse continuum is known from zero-range impurities as well [21,70,88]. This indicates that the spectral broadening of the attractive polaron peak near resonance in the zero-range polaron and the emergence of the Gaussian feature in Rydberg polaron studies share the same underlying physical origin.

This collection of phenomena is not limited to a Rydberg atom, but is shared by all impurity systems: the quantitative differences between the absorption spectra of various impurities are a matter of degree rather than of kind. This can be seen by comparing the spectrum of a neutral impurity [67,70] or an ionic impurity [56] with the present results. In the latter case, the interaction is often taken to be a regularized polarization potential [56,89]

$$V_{\text{Ion}}(r) = -\frac{\alpha_P}{(r^2 + b^2)^2} \frac{r^2 - c^2}{r^2 + c^2}, \quad (4)$$

with a characteristic length scale $R_{\text{Ion}} = \sqrt{2\mu\alpha_P}$, determined by the atomic polarizability α_P . We calculated $A(\omega)$ for $c =$

$0.0023 R_{\text{Ion}}$, varying b to adjust the potential depth, as shown in Fig. 5. We observe polaron features and fragmentation of the molaron states as they cross a scattering resonance, familiar from Fig. 3(a). At the density of $\rho = 2 R_{\text{Ion}}^{-3}$, the molaron states possess significant spectral weight and hint at the emerging Gaussian line shape centered around the mean-field energy shift $E_{\text{Ion}} = \rho \int V_{\text{Ion}}(r) d^3r$, which becomes completely dominant at higher densities as in Fig. 3(c) [90]. As with the Rydberg impurity, by writing $E_{\text{Ion}} = 2\pi a_{\text{Ion}}^B \rho / \mu$ we see that this feature is simply characterized by the Born approximation for the scattering length, the reduced mass, and the bath density.

With the insights provided by these two impurity systems, we have shown that $a\rho^{1/3}$ determines the parameter regime, how well the zero-range approximation applies, and the qualitative behavior of the absorption spectra. This unites the phenomena of finite-ranged impurities with those known from the well-studied zero-range impurity (the “Bose polaron”). In the low density limit, even deeply bound molecular states, whose energies depend on the two-body interaction, respond universally to the influence of the bath and have nonzero quasiparticle weight. At sufficiently high densities, the details of the potential become irrelevant and, even though the zero-range approximation fails, the Gaussian responses are the same, depending only on the Born approximated scattering length in a mean-field description. This signifies the loss of quasiparticle character.

The huge length scale of the Rydberg atom is especially appropriate for the approximations made in our study: the neglect of bath interactions, kinetic energy correlations, and the effect of three-body recombination. Although the bath-induced density shift of the molaron peaks, $4\pi n^2 \rho / \mu$, is small relative to the binding energies, it is comparable to, or even much larger than, the polaron shift already measured in ultracold mixtures of ground state atoms [32,34]. Therefore, Rydberg impurities should serve as a more suitable platform for studying molarons, especially the repulsive polaron, and refining their theoretical description. Even though the additive structure of polyatomic long-range Rydberg molecules has already been observed [52,61,91–93], the presence of this density shift has not yet been searched for. Another exciting avenue would be to consider the effect of background defects on the Rydberg excitons observed in CuO_2 [94–97] or transition metal dichalcogenide semiconductors [98,99], which should again lead to spectral broadening and perhaps the spatial pinning of the excitons.

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