Creation of Rydberg Polarons in a Bose Gas

F. Camargo,1 R. Schmidt,2,4,7 J. D. Whalen,1 R. Ding,1 G. Woehl, Jr.,1,5 S. Yoshida,6 J. Burgdörfer,6 F. B. Dunning,1 H. R. Sadeghpour,2 E. Demler,3 and T. C. Killian1,*

1Department of Physics & Astronomy, Rice University, Houston, Texas 77251, USA
2ITAMP, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts 02138, USA
3Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA
4Institute of Quantum Electronics, ETH Zürich, CH-8093 Zürich, Switzerland
5Instituto de Estudos Avançados, São José dos Campos, São Paulo, 12.228-001, Brazil
6Institute for Theoretical Physics, Vienna University of Technology, A-1040 Vienna, Austria, EU
7Max-Planck-Institute for Quantum Optics, 85748 Garching, Germany

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We report spectroscopic observation of Rydberg polarons in an atomic Bose gas. Polarons are created by excitation of Rydberg atoms as impurities in a strontium Bose-Einstein condensate. They are distinguished from previously studied polarons by macroscopic occupation of bound molecular states that arise from scattering of the weakly bound Rydberg electron from ground-state atoms. The absence of a p-wave shape resonance in the low-energy electron-atom scattering in Sr introduces a universal behavior in the Rydberg spectral line shape and in scaling of the spectral width (narrowing) with the Rydberg principal quantum number, n. Spectral features are described with a functional determinant approach (FDA) that solves an extended Fröhlich Hamiltonian for a mobile impurity in a Bose gas. Excited states of polyatomic Rydberg molecules (trimers, tetrameters, and pentamers) are experimentally resolved and accurately reproduced with a FDA.

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The interaction of an impurity with a deformable medium can lead to a collective response and formation of quasiparticles known as polarons, consisting of the impurity dressed by excitations of the background medium. Polarons play important roles in conduction in ionic crystals and polar semiconductors [1], spin-current transport in organic semiconductors [2], optical absorption of two-dimensional materials [3–5], and collective excitations in strongly interacting fermionic and bosonic ultracold gases [6–8]. Here, we report spectroscopic observation of Rydberg polarons formed through excitation of Sr Rydberg atoms in a strontium Bose-Einstein condensate (BEC), which represent a new class of impurity states beyond those typically seen in condensed matter settings [9]. Rydberg polarons are distinguished by macroscopic occupation of bound molecular states that arise from interaction of ground-state atoms with the Rydberg electron. A functional determinant approach (FDA) [10], adapted to include effects of impurity recoil, solves an extended Fröhlich Hamiltonian for an impurity in a Bose gas and accurately reproduces the observed excitation spectrum.

Rydberg-excitation experiments have been carried out previously using a Rb BEC [11,12]. In Rb, however, a p-wave shape resonance [13–15] for e-Rb scattering introduces highly nonuniversal density and n dependence of Rydberg absorption spectra, complicating the identification of polaron features. In Sr, such a shape resonance is absent. Deviations in the Sr excitation spectrum from a mean-field prediction reveal that in the s-wave-dominated regime, the shape of the polaron spectrum and the scaling of the spectral width with n are insensitive to quantitative details of the underlying potential, and, in this sense, universal. Accessing this window of universality is new in Rydberg physics.

The formation of Rydberg and typical condensed-matter polarons [9] can both be understood as dressing of the...
impurity by excitations of the bosonic bath, which entangles
the impurity momentum with that of the bath excitations
[16], but there are important differences. Most importantly,
the spectral width for a region of uniform density shows that
Rydberg polarons exist not as the ground state of the many-
body system but rather as a set of excited states, which is
unusual in polaron physics. This highlights the special nature
of Rydberg polaron formation where no single many-body
state dominates the nonequilibrium dynamics at late times.
Furthermore, the description of Rydberg polarons relies on
multiple excitations from the BEC—on the order of the
number of atoms within the Rydberg orbit, while other
polaron states are often well described by including only
single-excitation terms. In addition, whereas most polarons
in condensed matter systems can be described fully with
excitations to unbound states such as free particle or phonon
modes, for Rydberg polarons it is essential to include
negative-energy bound states.

Bound states are also important for Fermi [6,17–21]
and Bose polarons [7,8,22–28] consisting of ground-state
impurity atoms interacting with atoms in a background
Fermi gas or BEC, respectively. In these cases, contact
interactions are tuned with a Feshbach resonance, while
Rydberg impurities introduce long-range interactions with
strength tuned by changing \( n \).

The interaction between a Rydberg atom and a bosonic
medium can be described with the Born-Oppenheimer
potential for a ground-state atom a distance \( r \) from the
Rydberg impurity [13,14,29–31]

\[
V(r) = \frac{2\pi\hbar^2}{m_e} A_s |\Psi(r)|^2 + \frac{6\pi\hbar^2}{m_e} A_p |\nabla \Psi(r)|^2, \tag{1}
\]

where \( \Psi(r) \) is the Rydberg electron wave function, \( A_s \)
and \( A_p \) are the momentum-dependent s-wave and p-wave
scattering lengths, and \( m_e \) is the electron mass. When
\( A_s < 0 \), \( V(r) \) can support molecular states with one or more
ground-state atoms bound to the impurity [30,32,33].
The typical form of such a potential is shown in Fig. 1,
which illustrates schematically the formation of a Rydberg
polaron. Laser excitation of an atom in the BEC creates a
Rydberg impurity and projects the system into a super-
position of bound and scattering states \( |\beta_i\rangle \). While bound
states are confined in the potential \( V(r) \), scattering states
can encompass the entire BEC.

The Hamiltonian for a mobile impurity with mass \( M \)
at position \( \vec{R} \) and momentum \( \vec{p} \), interacting with a gas of
bosons is

\[
\hat{H} = \frac{\hat{r}^2}{2M} + \sum_k c_k \hat{a}^+_k \hat{a}_k + \frac{1}{V} \sum_{kq} V(q) e^{-i\vec{q} \cdot \vec{R}} \hat{a}^+_k \hat{a}_{k+q}, \tag{2}
\]

where \( V \) is the quantization volume, and \( \hat{a}_k \) (\( \hat{a}^+_k \)) are the
annihilation (creation) operators for the bosons with
dispersion \( c_k = \sqrt{\hbar^2 k^2/(2m)} \) (for details see Ref. [16]). The last term, which gives rise to polaron
formation, describes the Rydberg impurity-boson interaction
where \( V(q) \) is the Fourier transform of Eq. (1). In our
experiments, the impurity-boson interaction is much
stronger than the interaction between bosons, which hence,
is omitted when calculating the excitation spectrum from a
region of constant density.

The Hamiltonian of Eq. (2) is the basis for various
polaron models [16]. For instance, applying the
Bogoliubov approximation and keeping only terms that
are linear in bosonic field operators yields the widely
studied Fröhlich Hamiltonian [34]. However, the Fröhlich
Hamiltonian does not account correctly for scattering,
including formation of bound states [7,8,22–28,35,36].
Retaining all impurity-boson interaction terms yields an
extended Fröhlich Hamiltonian, which is required (and
used here) to account for the role that bound molecular
states play in Rydberg polaron formation.

The Rydberg excitation spectrum in the linear response
is given by \( A(\nu) \propto \sum_j |\langle \Psi_j | \Psi_{\text{BEC}} \rangle|^2 \delta \nu - (E_j - E_{\text{BEC}})/\hbar \),
for laser detuning \( \nu \). Here \( |\Psi_{\text{BEC}}\rangle \) is the BEC ground state
before excitation of the impurity, with energy \( E_{\text{BEC}} \), and the
sum is over all many-body eigenstates \( |\Psi_j\rangle \) of Eq. (2)
describing the system after impurity excitation, with energy
\( E_j \) [16]. The states \( |\Psi_j\rangle \) are constructed by the occupation
of single-particle bound and scattering states \( |\beta_i\rangle \), which are
modified by the Rydberg potential (see Fig. 1). \( A(\nu) \) is
evaluated with a FDA that was previously developed to
describe only the case of immobile impurities [10]. Here,
we extend the previous work with a canonical trans-
formation that transforms the system into the frame
comoving with the impurity [16,37,38] and decouples
the impurity and boson Hilbert spaces, mapping the
problem onto a purely bosonic system [26]. This allows
treatment of impurity motion, which is crucial for a
accurate description of the observed spectra. Spectra are
obtained from the unitary time evolution of the
Hamiltonian in Eq. (2) including all orders of the impu-
rit-y-Bose interaction. Finally, we average the spectrum over
the atomic density profile [16].

The Rydberg excitations are carried out in a \(^{86}\text{Sr} \) BEC in a
crossed-beam, optical-dipole trap [39,40]. The BEC is
close to spherically symmetric, with \( 3.5 \times 10^5 \) atoms
atoms, a peak condensate density \( \rho_{\text{max}} = 3.6 \times 10^{14} \text{ cm}^{-3} \),
and a Thomas-Fermi radius \( R_{TF} = 8 \mu \text{m} \). The condensate
fraction is \( \eta \sim 75\% \) and the temperature is \( T \sim 150 \text{ nK} \).
These parameters are determined from time-of-flight
measurements using resonant absorption imaging on the
\(^1\text{S}_0 \rightarrow ^1\text{P}_1 \) transition with 461 nm light. The average
nearest-neighbor atom separation at \( \rho_{\text{max}} = 80 \) nm.

Rydberg spectra are obtained by driving the \( 5s^2 \ ^1\text{S}_0 \rightarrow
5s5p \ ^3\text{P}_1 \rightarrow 5s6s \ ^3\text{S}_1 \) two-photon transition using 689 nm
and 319 nm light [33,41]. The 689 nm light is blue-detuned
from the intermediate state by 80 MHz while the frequency
of the 319 nm laser is scanned. The laser excitation is
applied in a 2 \( \mu \text{s} \) pulse, immediately followed by an
electric-field ramp that ionizes Rydberg atoms and molecules in about 1 μs and directs the product electrons towards a micro-channel-plate detector. The time from excitation to detection is kept short to avoid density-dependent Rydberg-atom loss [41,42], which would distort the spectrum. Typical intensities of the excitation beams are 200 mW/cm² and 6 W/cm² for the 689 nm and 319 nm beams, respectively, and they are uniform across the BEC. This results in a Rydberg excitation rate well below one per pulse, avoiding Rydberg-blockade effects and ensuring a linear-response measurement. Excitation and detection are repeated at a 4 kHz rate. For each frequency point in the spectrum, a new trapped sample is used and the signal from 1000 pulses is acquired. The peak atom density varies by less than 5% for each point.

At low principal quantum numbers, resolved Rydberg-molecular lines dominate the spectrum and are well understood [43]. This is shown for Sr(5s38s) excitation in Fig. 2, for which on average three atoms are within the Rydberg orbital \( N_{\text{orb}} = \rho_{\text{max}} 4 \pi r_p^2 / 3 \), where \( r_p \) is the radius of the outer lobe of the wave function. Individual spectral lines correspond to Rydberg molecular states with one [dimer (D)], two [trimer (Tr)], three [tetramer (Te)], and four [pentamer (P)] bath atoms bound in the available levels. The intricate structure of the molecular lines is reminiscent of the nuclear shell model.

We find remarkable agreement between theory and experiment over several orders of magnitude of spectral strength. For theoretical calculations, the sample temperature, total atom number, trap frequencies, and overall signal intensity are taken as adjustable parameters and lie within experimental uncertainty [16].

For higher \( n \), the average number of ground-state atoms per Rydberg-orbital volume increases to 13, 50, and 160 atoms for \( n = 49, 60, \) and 72, respectively, and many-body effects become relevant (Fig. 3). The interplay of few- and many-body physics is particularly evident for \( n = 49 \). Here, a series of up to sevenfold-occupied molecular bound states can be identified for scaled detuning between zero and \(-0.5\). With increasing \( n \), the spectrum evolves into a broad continuum, which is the signature of many-body dressing of the impurity. The peak in spectral support at zero detuning, comes almost exclusively from the low-density, non-condensed atoms. The FDA, which accounts for quantum many-body effects, describes the data well (details in [16]).

A mean-field treatment using the local density approximation helps to elucidate the spectral signatures of polaron excitation in the regime of large numbers of atoms per Rydberg orbital. This treatment is reminiscent of Fermi’s original description of Rydberg excitation in dense thermal gases [29,43] and predicts an excitation spectrum of the form \( A(\nu) \propto \int d^3 r \rho(r) \delta[\nu - \Delta(r)] \). An atom in a spatial region with density \( \rho(r) \) contributes to the spectrum at a detuning from unperturbed atomic resonance of \( \Delta(r) = \rho(r) \int d^3 r' V(r - r') / h \). This mean-field shift reflects a spatial average of the interaction between

![FIG. 2. Spectrum for excitation of the 5s38s3S1 Rydberg state in the few-body regime. Pure atomic excitation is at zero detuning, and lines for dimers (\( X = D \)) through pentamers (\( X = P \)) are visible. Subscripts (\( X_{ij} \)) indicate the molecular levels of the Rydberg potential occupied by atoms after laser excitation. The FDA matches the data over several orders of magnitude in signal strength, which is normalized to the peak at zero detuning.](image)

![FIG. 3. Rydberg excitation in the many-body regime for \( n = 49, 60, 72 \). The mean-field prediction is for the condensate atoms only. Deviations between the mean-field result and data at small detuning (\( \nu / \Delta_{\text{max}} < 0.3 \)) represent contributions from the noncondensed thermal atoms. The shaded regions represent the predicted polaron response from the center of the atom cloud as obtained from the FDA. Theory for this high-density region matches the tail of the experimental data and represents the Rydberg-impurity spectral function for a nearly uniform-density region. For uniform density, the width of the spectrum scales as \( n^{-3} \).](image)
Rydberg and ground-state atoms [cf. Eq. (1)]. At fixed density, Δ varies slightly with n [16].

In Fig. 3, the frequency axis is scaled by the fit value of the mean-field shift for the peak density in the BEC (-Δ\text{max}) [16], and the Rydberg-excitation intensity is normalized to its integral neglecting the thermal contribution. With this scaling, the mean-field prediction is independent of n. The rough agreement between experiment and the mean field prediction implies that the shape of the full experimental spectrum is dominated by broadening due to the inhomogeneous, in-trap density distribution. While the mean-field matches the response at intermediate frequencies, it fails to describe the resolved few-body features and the red-detuned tails of the spectra in Fig. 3.

It is the deviations from mean-field theory that provide key insight into the nature of Rydberg polarons. While polarons in condensed matter systems [6] are often described through dressing by long wavelength excitations of the medium such as phonons, the formation of Rydberg polarons relies on the dressing by localized states bound to the impurity with discrete, negative energies. These molecular levels are revealed as resolved features at small detuning for n = 49. Massive, many-body occupation of these bound states gives rise to the formation of Rydberg polarons and the broad tail at far red detuning.

The FDA predicts that Rydberg polaron formation leads to a Gaussian profile of the excitation spectrum for systems of uniform density, with a width that decreases with increasing n, which are both beyond-mean-field effects. To investigate these predictions, we focus on the spectral tail at large red detuning where a clear variation between the different data sets is found. Using the FDA, we are able to isolate the spectral contribution from a small, central region with the highest density (shaded regions in Fig. 3). Comparison between experiment and the FDA shows that the tail arises entirely from this high-density region. In this region the average density is \( \langle \rho \rangle = 0.92 \rho_{\text{max}} \). The small standard deviation \( \sqrt{\langle (\Delta \rho)^2 \rangle} / \langle \rho \rangle = 0.04 \) shows that the spectral width reflected in the tail cannot be explained by density inhomogeneity. Since the observed spectral width likewise cannot be explained by laser resolution (400 kHz), we attribute it to the intrinsic excitation of Rydberg polarons. The agreement between the simulated Rydberg polaron response and the observed data over an order of magnitude variation in the signal strength confirms the FDA predictions.

The emergence of a Gaussian profile is one of the defining properties of Rydberg polaron formation. Its presence can be understood by decomposing the spectral function \( A(\nu) \) in terms of excitations from the BEC ground state to interacting single-particle states. The resulting multinomial distribution is dominated by a few terms with large probability \( p_i = |\langle \beta_i | s \rangle|^2 \) for bath particles in state \( |s\rangle \) to scatter, via interaction with the Rydberg atom, into states \( |\beta_i\rangle \) (see Fig. 1). For a model with \( N \) particles and only two interacting single-particle states, i.e., one bound state (e.g., \( |\beta_0\rangle \), Fig. 1), with energy \( e_B \) and overlap \( p_0 \ll 1 \), and one low-energy scattering state (e.g., \( |\beta_1\rangle \equiv |\beta_i\rangle \)) with energy \( e_1 \approx 0 \) and \( p_1 = 1 - p_0 \), a binomial distribution results,

\[
A(\nu) = \sum_{j=1}^{N} \binom{N}{j} p_0^j (1 - p_0)^{N-j} \delta(\nu - j e_B / h).
\]

For large \( N \) Eq. (3) becomes a Gaussian with mean energy \( \mu = N p_0 e_B \) and variance \( N p_0 (1 - p_0) e_B^2 \). Since the single-particle overlap of \( |s\rangle \) with the bound state \( |\beta_j\rangle \) is small, the Gaussian width becomes \( \sqrt{N p_0 e_B} \). We identify \( N p_0 = N_{\text{ob}} = 4 \pi \rho r_R^2 / 3 \), where \( r_R \sim n^2 \) is the Rydberg electron radius, and assume that the Gaussian mean energy corresponds to the mean-field energy \( h \Delta \). The Gaussian width then varies as \( \sim \sqrt{\rho / n^3} \), in agreement with experiment and numerical simulation of the full Hamiltonian (Fig. 3). The spectrum is sensitive to density-density correlations in the background gas, which display Poissonian fluctuations for a BEC. The decreasing width with decreasing density also explains why the data and mean-field prediction agree well at small detuning after subtracting the contribution from the thermal fraction. The Gaussian line shape and \( \sim \sqrt{\rho / n^3} \) scaling are universal features that emerge when s-wave electron-atom scattering dominates, and they are masked by sensitivity to quantitative details of the underlying interaction in the presence of a shape resonance, as in Rb [12].

The spectrum \( A(\nu) \) in the approximate expression Eq. (3) consists of delta-shaped peaks corresponding to quantized molecular levels. In the accurate FDA description, the binomial distribution is effectively replaced by a multinomial distribution that yields denser energy levels and includes coupling to scattering states, which, in principle, broadens molecular levels asymmetrically to positive energies. But this effect is too small to be resolved within our experimental resolution. The experimentally observed broadening to the blue of unperturbed atomic resonance, most clearly visible in Fig. 2, does not arise from known mechanisms connected to the presence of a shape resonance [12,44], nor can it be attributed to finite temperature, and it remains a subject for future studies.

When the spacings of molecular levels become comparable to the broadening due to scattering states or experimental resolution, the spectrum becomes a continuous distribution. In this limit the spectrum can also be well described by a classical statistical model as, e.g., used to describe Rydberg impurity excitations in a Rb condensate [12]. As shown in Ref. [16], this model corresponds to an approximation in which the motions of the impurity and bath atoms are neglected. Since this model “freezes” all dynamics in the system it cannot describe Rydberg molecules (visible as resolved lines in the spectrum), which are the underlying building blocks of Rydberg polarons.

In conclusion, we present experimental evidence for the creation of Rydberg polarons consisting of an impurity Rydberg atom dressed by a large number of bound...
excitations of Bose-condensed Sr atoms. By focusing on the red-detuned spectral tail, we show that a Gaussian line shape and $1/n^3$ scaling of the linewidth with principal quantum number are universal features of the Rydberg-polaron excitation spectrum in a BEC when $s$-wave electron-atom interactions dominate. The spectral width reflects that Rydberg polarons are created in excited states of the impurity-bath system that originate from the interplay of many-body dressing and few-body molecule formation. The spectral width can serve as a local probe of density-density correlations in the BEC. Our observations are in remarkable agreement with FDA predictions including impurity recoil, emphasizing the quantum nature of the polaron. Important questions remain to be explored, such as whether the interactions are strong enough to lead to self-localization of the impurity, and the extent to which notions of effective mass, mobility, and polaron-polaron interactions are applicable to Rydberg polarons. Ramsey interferometry experiments can probe the intrinsic quantum nature of the dressing of Rydberg impurities in real time [20,21,45,46].

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*Corresponding author.
killian@rice.edu