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PAPER

Imaging single Rydberg electrons in a Bose–Einstein condensate

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**Abstract**

The quantum mechanical states of electrons in atoms and molecules are distinct orbitals, which are fundamental for our understanding of atoms, molecules and solids. Electronic orbitals determine a wide range of basic atomic properties, allowing also for the explanation of many chemical processes. Here, we propose a novel technique to optically image the shape of electron orbitals of neutral atoms using electron–phonon coupling in a Bose–Einstein condensate. To validate our model we carefully analyze the impact of a single Rydberg electron onto a condensate and compare the results to experimental data. Our scheme requires only well-established experimental techniques that are readily available and allows for the direct capture of textbook-like spatial images of single electronic orbitals in a single shot experiment.

The wavefunction is a fundamental concept of quantum mechanics. Our current understanding of atoms, molecules and solids is based on the fact that the probability density of an electron is the absolute square of the electronic wavefunction. The theoretical description of electron orbitals was founded at the beginning of the last century. Yet, to date the spatial structure of most orbitals has not been observed directly. Most current techniques to study the wavefunction of electrons in atoms and molecules rely on tomographic reconstruction. Based on this technique the wavefunction of the energetically highest orbital of various molecules has been obtained using high harmonics generated in the interaction of intense femtosecond laser pulses with molecules [1] and photoemission spectroscopy [2]. Another method, which has been until now applied to larger polymers, is based on scanning tunneling microscopy [3–5]. Furthermore, an image of the electron wavefunction in single hydrogen atoms has been reconstructed only recently [6]. There, electronic states in a strong static electric field have been observed via photoionization and subsequent electron detection using a magnifying electrostatic lens.

Here, we propose a method to optically image the orbitals of electrons excited to a Rydberg state. These orbitals are larger in size than optical wavelengths. Moreover, due to the different sizable quantum defects and dipolar selection rules, they can be prepared in a well-defined quantum state providing clean s, p, d and f series. The proposed method is based on the interaction of the Rydberg electron with a dense ultracold gas [7]. Due to this interaction the probability density of a single Rydberg electron can be imprinted on the density of surrounding Bose–Einstein condensate (BEC) atoms. Thus, textbook-like optical images of hydrogenic states can be obtained using already well-established imaging techniques for cold atoms.

In order to model the Rydberg excitation dynamics and the phase imprint onto a finite-size BEC we develop a numerical model describing the probabilistic Rydberg excitation process and the subsequent interaction with the finite-size BEC. Our approach agrees well with available experimental data on Rydberg excitations in a BEC [7] and confirms electron–phonon coupling as the underlying mechanism, which has been studied previously in the framework of Bogoliubov approximation. We discuss the experimental requirements and challenges to

implement our proposal, including finite imaging resolution as well as the role of atomic and photonic shot noise for the expected images of Rydberg orbitals.

The $1/r^4$ interaction between the single electron of the Rydberg atom and polarizable ground state atoms can be well described by a pseudopotential [8, 9] resulting in an effective potential acting on the ground state atoms of the form

$$V_{\text{Ryd}}(\vec{r}) = \frac{2\pi\hbar^2 a}{m_e} |\Psi_{\text{Ryd}}(\vec{r})|^2, \quad (1)$$

where $\Psi_{\text{Ryd}}(\vec{r})$ is the Rydberg electron wavefunction, a denotes the electron–atom s-wave triplet scattering length, $a = -16.1$ a.u. for ^{87}Rb [10], and m_e is the electron mass. This well-known model has previously been used to make quantitative statements about the binding energy and excitation spectra of ultralong-range Rydberg molecules [11–14]. Higher partial waves are irrelevant for principal quantum number $n > 100$. The interaction between the ionic core and the BEC is ~ 300 times smaller and can be safely neglected [15]. Thus, the interaction between the Rydberg atom and ground state atoms creates a potential V_{Ryd} around the excited atom with a structure defined by the Rydberg electron orbital.

To model the effect of a single Rydberg electron on the BEC we introduce the pseudopotential term V_{Ryd} as a mean field component in the Gross–Pitaevskii equation (GPE). GPE describes the dynamics of the bosonic atomic field. We adopt a classical field approximation (CFA), where a long-wavelength atomic field is replaced by a classical complex function $\Psi(\vec{r}, t)$ satisfying the time-dependent GPE

$$i\hbar \frac{\partial}{\partial t} \Psi(\vec{r}, t) = \left[-\frac{\hbar^2}{2m} \nabla^2 + V_{\text{trap}}(\vec{r}) + g |\Psi(\vec{r}, t)|^2 + f(t) V_{\text{Ryd}}(\vec{r} - \vec{R}) \right] \Psi(\vec{r}, t), \quad (2)$$

where $f(t)$ is 1 for the finite time when the Rydberg atom is present in the BEC and 0 otherwise. On the right-hand side the first three terms are related to the kinetic energy, the trapping potential and the contact interaction with coupling constant g . CFA is a valid treatment for describing Bogoliubov–Popov excitations [16].

Before we turn to the investigation of the imaging of electron orbitals, we use our approach to model our recent experiment, where about a few hundred Rydberg atoms were excited successively at random positions inside a BEC. In the experiment [7] a Rydberg atom in an s-state with principal quantum numbers n ranging from 110 to 202 was created in a condensate of ^{87}Rb atoms. We used a $1 \mu\text{s}$ light pulse, during which the Rydberg atom got excited with a certain probability. $10 \mu\text{s}$ after the excitation pulse, we sent a $2 \mu\text{s}$ ionization pulse, which extracted the Rydberg atom unless it has not been lost before. Although the Rydberg blockade mechanism [17] ensures that at any moment there was not more than a single excitation within the BEC, we studied only the cumulative effect of many successive excitations on the BEC. In the finite-size BEC the resonance frequency is modified by the spatially varying energy shift $\delta E(\vec{R}, t)$ [18] due to local density. We model this complicated many-body excitation process by a stochastic model.

Each appearance and disappearance of the potential V_{Ryd} are examples of quantum jumps. We check if a randomly chosen atom on a grid representing the density distribution $\rho(\vec{r})$ is in a Rydberg state according to the excitation probability. The probability to find an atom at position \vec{R} in the Rydberg state, for sufficiently short times t and low single atom Rabi frequencies Ω_R , is given as

$$p(\vec{R}, t) = \frac{\Omega_R^2}{\Omega^2(\vec{R}, t)} \sin^2 \left[\Omega(\vec{R}, t) t/2 \right], \quad (3)$$

where $\Omega(\vec{R}, t) = \sqrt{\Omega_R^2 + \Delta^2(\vec{R}, t)}$ is the effective Rabi frequency, which accounts for a non-zero local detuning $\Delta(\vec{R}, t)$. This spatially varying detuning $\Delta(\vec{R}, t)$ is given by a frequency difference of the detuning $\Delta\omega_L$ of the excitation laser from the Rydberg transition in an unperturbed atom and an additional mean field shift $\delta E(\vec{R}, t)$. Since the condensate density changes due to the appearance of successive Rydberg atoms, the detuning Δ and thus the excitation probability (3) depend also on time.

Following a coherent evolution at all possible grid points a localized Rydberg atom is potentially generated in our simulation on the time scale of the decoherence rate due to elastic scattering with a ground state atom. Therefore we choose the time step for our coherent evolution to be 200 ns , which we then interrogate for the presence of a localized Rydberg atom. We repeat checking the atoms every 200 ns until the end of the excitation pulse if no Rydberg atom was found in the previous iteration. Once a Rydberg atom is created we propagate the GPE with the V_{Ryd} term included to calculate the evolution of the perturbed BEC while the Rydberg atom is present in the BEC. The interaction of a single Rydberg electron with surrounding BEC atoms decays

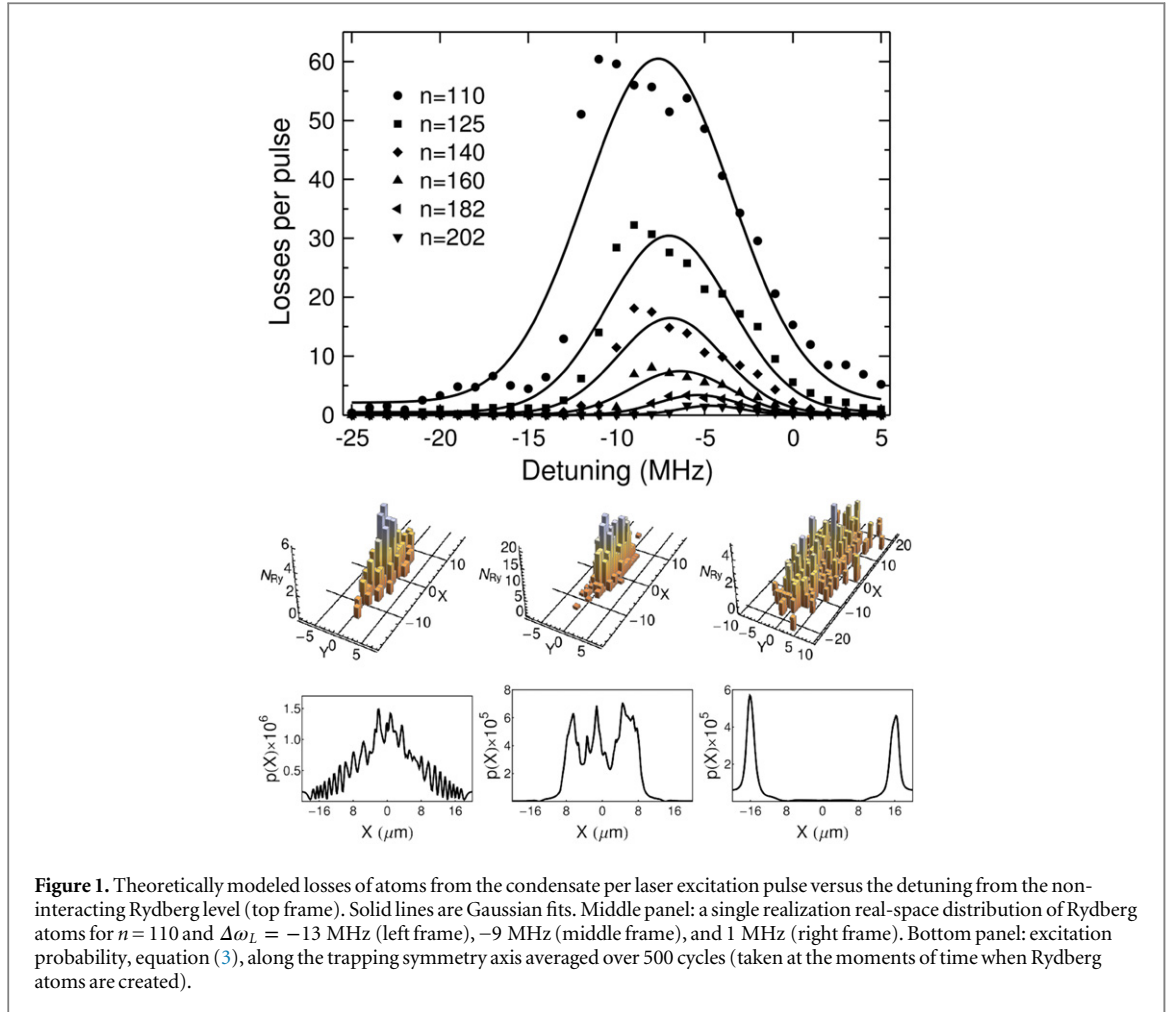


Figure 1. Theoretically modeled losses of atoms from the condensate per laser excitation pulse versus the detuning from the non-interacting Rydberg level (top frame). Solid lines are Gaussian fits. Middle panel: a single realization real-space distribution of Rydberg atoms for $n = 110$ and $\Delta\omega_L = -13$ MHz (left frame), -9 MHz (middle frame), and 1 MHz (right frame). Bottom panel: excitation probability, equation (3), along the trapping symmetry axis averaged over 500 cycles (taken at the moments of time when Rydberg atoms are created).

exponentially in time with a time constant of $\sim 10 \mu\text{s}$ ⁵. After $13 \mu\text{s}$ the whole procedure starts again, however, the density distribution of the BEC is changed by the previous cycle. A cycle consisting of the excitation of the Rydberg atom followed by a finite interaction time with the condensate atoms is repeated 300 or 500 times as it was done in the experiment.

The energy of the system increases by the time-dependent potential V_{Ryd} . Thus the condensate fraction is reduced. Some of the ground state atoms are promoted from the condensate to the thermal cloud. Within the CFA the two components of the bosonic gas—the condensate and the thermal cloud are identified by accounting for the coarse graining as an unavoidable element of the measurement process [19], [20]⁶. Here, we define a coarse grained one-particle density matrix as resulting from the column integration [21]

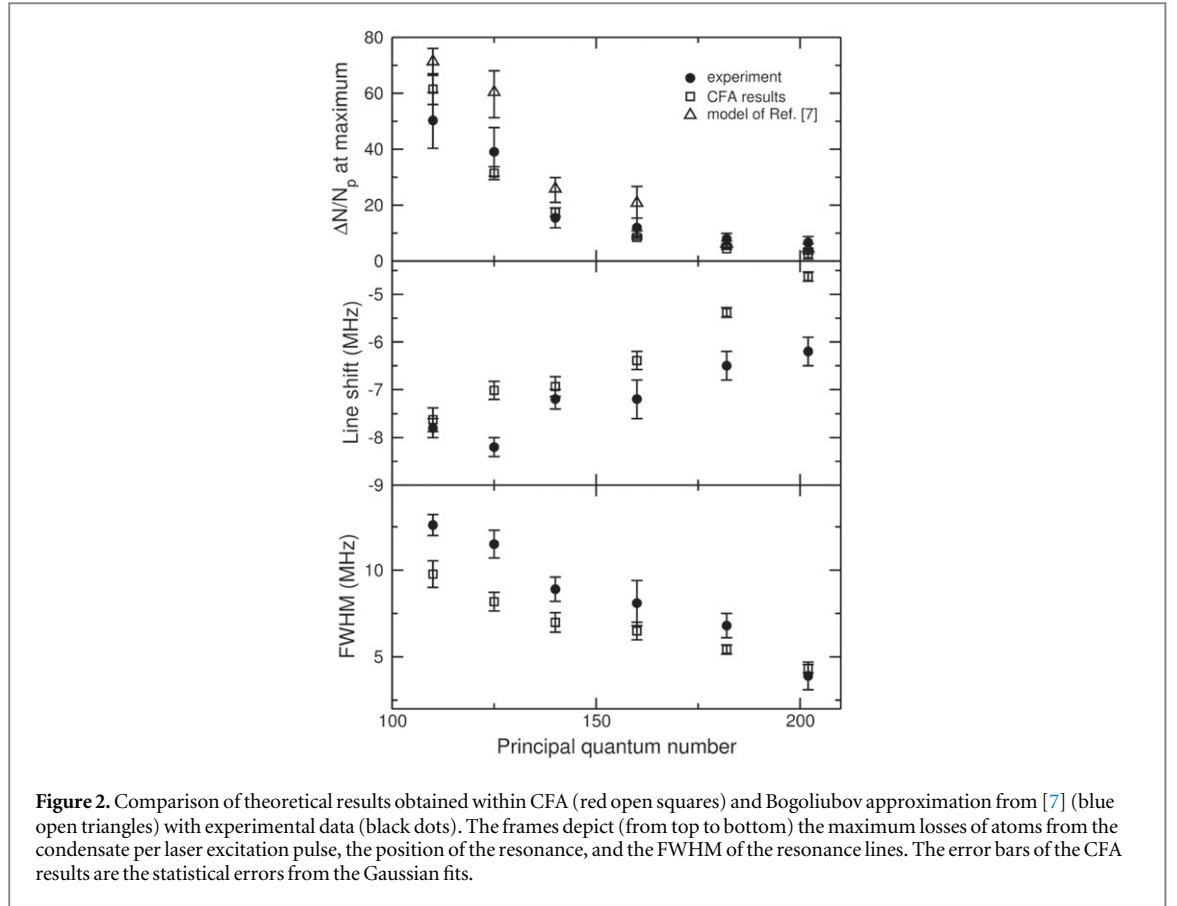
$$\bar{\rho}(x, y, x', y'; t) = \frac{1}{N} \int dz \Psi(x, y, z, t) \Psi^*(x', y', z, t), \quad (4)$$

where the x -axis is the condensate symmetry axis and the imaging is performed along the radial direction. The resulting density matrix, upon spectral decomposition [22], determines the fraction of the condensed atoms as a dominant eigenvalue.

We calculate the total condensate losses at the end of the excitation sequence, divide them by the number of excitation cycles and study the dependence of this quantity as a function of the laser detuning $\Delta\omega_L$ and the principal quantum number n of the Rydberg state (figure 1). On the blue side of the resonance the Rydberg atom is created almost in every shot but losses are small because Rydberg atoms are excited in regions of low density, far from the center (see middle panel in figure 1, right frame). Towards the center of the line more Rydberg atoms are excited around the center of the trap where the density of the condensate is high. This leads to the increase of losses reaching a maximum approximately at the point where $\Delta\omega_L$ is equal to $\delta E(\vec{R}, t)/\hbar$ calculated

⁵ See supplementary material available at stacks.iop.org/njp/17/053046/mmedia.

⁶ A close analogy exists with the classical, Maxwell electrodynamics. At a microscopic level at each point in space and time an electric field has a well defined value even for the most complicated field. While a product of electric fields at two points in space–time is a well defined but usually useless number, a coarse graining caused by the detectors makes such a quantity useful, enabling, for instance, a proper definition of coherence length/time.



at the center of the trap. On the red side of the resonance still many Rydberg atoms are excited in the center of the trap (compare left and central frames of the middle panel in figure 1), however, not in every excitation cycle and thus the overall losses decrease. In the case of $n = 110$ and $\Delta\omega_L = -12$ MHz \sim two third of the excitation pulses creates a Rydberg excitation while only every seventh trial is successful at $\Delta\omega_L = -16$ MHz. The asymmetry of the process with respect to the center of the line stems from the detuning which is a function of the local density. As in [7] our BEC exhibits quadrupole oscillations after the excitation sequence is finished. However, the losses do not continue.

The absolute values of maximal losses determined from the Gaussian fits to our numerical data (figure 1) are compared to experimental data and Bogoliubov calculations from [7] in figure 2 (top panel). We extract also the position of the resonance (middle panel) and the width of the line (bottom panel). The numerical results agree remarkably well with the experimental data considering the fact that only estimated values for the Rabi frequencies from the measurement and no additional free parameters were used.

While the overall atom loss was already quantitatively predicted within a Bogoliubov approach in [7], our method presented here provides additional insights and describes time evolution of the BEC during the experimental sequence in detail. This fact is of importance, since for every excitation but the first one, the condensate is already distorted due to the influence of the previous Rydberg atoms. Our method is nonperturbative and goes beyond first order approximation in phonon production. Moreover we predict the whole resonance line shape and include in the model the inhomogeneous density of the condensate caused by the trapping potential.

Having demonstrated that our model reproduces the experimental data very well, we now turn to the proposal of observing an electronic orbital by imaging the condensate density responding to the Rydberg potential (equation (1)). Our scheme relies on optical access with high numerical aperture as is readily available in many BEC experiments. Such high resolution optics enable the tight focusing of the excitation lasers into the center of the condensate, to define the position of the Rydberg atom(s) with high precision (figure 3). Moreover, this enables high resolution absorption images of the BEC to be taken. We consider Rydberg s- and d-states, which are accessible in typical two-photon excitation schemes [23, 24].

First, we study the case where the excitation lasers are kept on continuously (figure 4(e)) to re-excite the Rydberg state as soon as the previous one has decayed. Here, the localization of the Rydberg atom must be within an area smaller than the expected structure size. Otherwise, the combined impact of many Rydberg excitations will wash out the Rydberg electron orbital imprint on the BEC. To resolve the overall structure of the exemplary

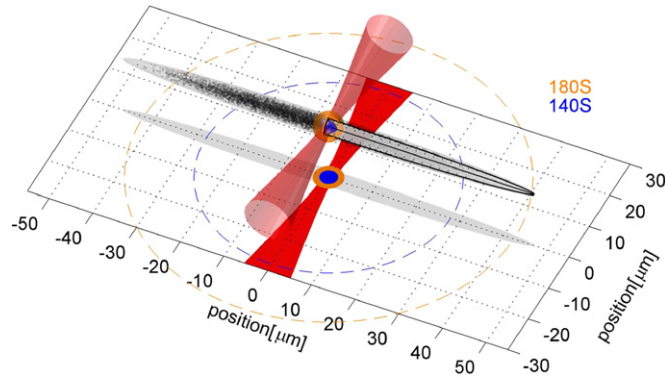


Figure 3. Rydberg atoms in 140S (blue) and 180D (orange) states are excited in the center of the condensate by a tightly focused laser beam (red). Dashed lines indicate the projection of the respective Rydberg blockade radii. All the sizes are to scale.

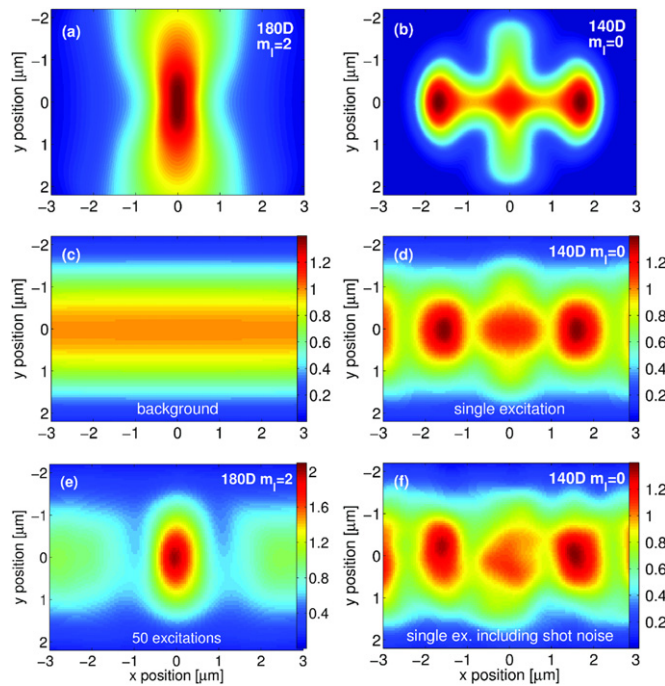


Figure 4. (a) and (b): calculated orbitals for different Rydberg electrons convolved with a finite imaging resolution of $1 \mu\text{m}$ ($1/e^2$ width of the point spread function). This is the maximum contrast which can be expected from the imprint on the condensate. (c): center part of the BEC density distribution. The condensate consists of 5×10^4 rubidium atoms and is confined in a harmonic trap with radial and axial frequencies $\omega_R = 2\pi \times 200 \text{ Hz}$ and $\omega_z = 2\pi \times 10 \text{ Hz}$, respectively. (d) and (f): simulated density change caused by a single Rydberg atom without (d) and with atom number shot noise (f). These density patterns form after a single Rydberg atom lasting for $30 \mu\text{s}$ (which will be the case for every twentieth shot for a lifetime of $10 \mu\text{s}$) and an additional evolution time of $180 \mu\text{s}$. (e): density distribution of the BEC after 50 Rydberg atoms have been consecutively excited in the center region ($1/e^2$ width: $1.2 \mu\text{m}$).

180D state (orbital radius $r = 3.3 \mu\text{m}$) it is sufficient to have the excitations within a diameter of $d = 1.5 \mu\text{m}$. Sufficient sharpness and a good contrast of the image requires about 50 excitations, since the scattering potential depth is lower than the chemical potential of the condensate.

To visualize the electron orbit of the Rydberg wavefunction with only one Rydberg excitation cycle (figure 4(d)) the parameters of the experiment have to be chosen more carefully. A detailed study of this situation can be found in the (see footnote 5) and is summarized in the following. The principal quantum number of the Rydberg state must not be chosen too large because although the orbital radius scales with n^{*2} , the effective potential drops with n^{*-6} . The calculations show that the effective potential should be at least one order of magnitude deeper than the chemical potential of the atoms so that they can react during the lifetime of the Rydberg atom. This situation is reached for a principal quantum number around 140. Additionally, the thickness of the condensate, which the imaging light is traveling through, should not be larger than the orbital

radius of the Rydberg electron. Otherwise the imaging light passes through an area that is not affected by the imprint of the electronic wavefunction, which results in a reduction of contrast. In such a single-shot experiment the atom number shot noise [25] is the main source of noise. For the proposed parameter set (figure 4(f)), a peak density $\rho \sim 10^{14} \text{ cm}^{-3}$ and a radial size of the condensate of $1.5 \mu\text{m}$ results in a $\sim 6\%$ background noise level (see supplementary material). This is well below the expected signal contrast of $\sim 24\%$. Therefore, the Rydberg orbital imprint on the BEC density should be observable. Note, that the impact of the shot noise may be reduced by averaging images from multiple runs.

To summarize, we have presented and verified a theoretical, microscopic model of a single Rydberg electron in a BEC. Our theoretical model has several simplifications: (1) we do not account for the real losses of the trapped gas due to three body recombination. (2) The electron in its motion is not slow in the vicinity of the ionic core. We assume the electron–atom scattering length to be velocity independent, which is a valid description beyond $n > 80$. (3) The impact of the ionic core on the heating process is neglected.

After verification of our theoretical model, we have proposed a novel scheme for mapping the electronic orbital onto the density of the condensate, thereby realizing a method to directly observe various electronic orbitals. Of course, with the available resolution we can image only the angular probability distribution of a Rydberg orbital. Its radial structure will be washed out and this is not only due to limited imaging resolution but also because tiny oscillating radial structure of higher Rydberg orbitals occurs on the space scale shorter than the healing length.

Also exotic shapes of single electron probability densities in electric and magnetic fields including circular Rydberg states [26], Stark states, Bohr-like wavepackets [27, 28] and one-dimensional atoms [29] could be investigated in a way we propose. Furthermore, this approach can also be extended to more complex systems like Rydberg atom macrodimers [30] and multi-electron systems. Phase-sensitive images could be obtained if a structureless reference state is used in a coherent superposition state. The technical requirements with respect to resolution, both for the local excitation of Rydberg atoms and the detection of the resulting structures, are met by state of the art experimental setups. Furthermore, various techniques like dark ground imaging [31], phase-contrast imaging [32], polarization contrast imaging [33, 34] and adapted forms of absorption imaging [35] are readily available to precisely determine the density distribution of a BEC in situ. The optical imaging of a single electron in a single shot experiment thus seems in direct reach.

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