State-selective all-optical detection of Rydberg atoms

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We present an all-optical protocol for detecting population in a selected Rydberg state of alkali atoms. The detection scheme is based on the interaction of an ensemble of ultracold atoms with two laser pulses: one weak probe pulse which is resonant with the transition between the ground state and the first excited state, and a pulse with high intensity which couples the first excited state to the selected Rydberg state. We show that by monitoring the absorption signal of the probe laser over time, one can deduce the initial population of the Rydberg state. Furthermore, it is shown that – for suitable experimental conditions – the dynamical absorption curve contains information on the initial coherence between the ground state and the selected Rydberg state. We present the results of a proof-of-principle measurement performed on a cold gas of 87Rb atoms. The method is expected to find application in quantum computing protocols based on Rydberg atoms.

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I. INTRODUCTION

Rydberg atoms coupled to electromagnetic fields form a promising system for the physical realization of quantum information protocols [1] and quantum simulations [2]. In these protocols qubits are realized by a set of atomic states, which includes one or potentially more Rydberg levels. One requirement of these schemes is the ability to measure the Rydberg states’ population in order to read out the results of the quantum operations. For accomplishing this task, most experiments with ultracold Rydberg gases use methods including field ionization and subsequent detection of electrons and/or ions on multi-channel plates or channeltrons [3]. These techniques offer high sensitivity and – for carefully chosen experimental conditions [4,5] – selectivity among the Rydberg levels [6–8].

Selective field ionization (SFI) techniques are based on the fact that the ionization threshold is different for each atomic state, increasing from higher to lower lying levels. Hence, by slowly ramping up the electric field and monitoring the electrons/ions over time it is possible to deduce the initial populations in each level. However, the population of a lower lying Rydberg level cannot be probed without destroying the population of any higher lying Rydberg state. Therefore this method is not applicable in protocols which require the independent probing of the different Rydberg states [3]. Another inherent property of the methods based on ionization is that the detected atoms are removed from the system and cannot be reused. Although this atomic loss is negligible in most cases [3], it might be a serious limitation in experiments working with only one or a few atoms [10].

One alternative to ionization detection methods is all-optical probing based on electromagnetically induced transparency (EIT) [11]. This approach has been successfully applied in order to non-destructively probe the Rydberg level structure in non-interacting [12,13] and weakly interacting [14] Rydberg gases, as well as in the presence of electric fields [13,15]. These experiments, however, did not access the population of the Rydberg state. On the other hand, an EIT-based scheme for the optical detection of Rydberg population [16,20] has been proposed and demonstrated in dense atomic clouds where the Rydberg blockade allows the spatially resolved detection of Rydberg atoms.

Here we propose an all-optical scheme for detecting the population in a selected Rydberg state in dilute gases. By using a series of laser pulses in EIT configuration this technique also allows for distinction between coherent superpositions and statistical mixtures of the ground and Rydberg states of the atoms. Since this scheme is based on the time-resolved observation of the optical response of individual atoms it may, in principle, be used down to the single atom level. Our method is state selective and applicable for testing the population not only in the highest Rydberg level of interest (cf. SFI) but any lower lying or intermediate Rydberg state.

We present our theoretical model along with numerical simulations and demonstrate the scheme in a proof-of-principle experiment with a dilute gas of 87Rb atoms showing the detection of the population in an initially prepared Rydberg state. Our analysis includes characteristic effects of Rydberg experiments such as blackbody-induced depopulation [21,22], superradiance [23] and dipole-dipole interaction [24].

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Let us consider a cold atomic gas interacting with two laser pulses in an EIT-like configuration \[1\]. One of the pulses, resonant with the atomic transition between the ground state \(|1⟩\) and the first excited state \(|2⟩\), is a weak probe pulse well below the saturation intensity, while the other one is a relatively strong coupling pulse which is resonant with the atomic transition between the first excited state \(|2⟩\) and a selected (arbitrary) Rydberg S state \(|3⟩\) (see Fig. 1). A single “reservoir” state \(|4⟩\) is used to model the neighboring Rydberg states \[13\].

We use a semiclassical approach for describing the dynamics of the system and the laser pulses are taken into account through their classical electric field. The atomic gas is modeled by a motionless ensemble of atoms. The state of the atoms is described by the density matrix \(\rho = \sum_{i,j=1}^{4} \rho_{ij} |i⟩⟨j|\), where the states \(|2⟩\) and \(|3⟩\) rotate with the atomic transition frequencies \(\omega_{21}\) and \(\omega_{23}\), respectively.

The time evolution of \(\rho(t)\) is described by the master equation
\[
\dot{\rho} = \left[\mathcal{H}, \rho\right] + \mathcal{U}[\rho].
\]

Here the Hamiltonian \(\mathcal{H}\) accounts for the interactions between the atoms and the laser pulses. The effects due to interatomic interactions are considered through dynamic effective rates in \(\mathcal{U}[\rho]\) along with radiative losses occurring in the system. The Hamiltonian \(\mathcal{H}\) is written as
\[
\mathcal{H} = -\frac{\hbar}{2} \left(\Omega_p |2⟩⟨1| + \Omega_c |3⟩⟨2| + \text{h.c.}\right) - \hbar \left(\delta_p |2⟩⟨2| + \delta_c |3⟩⟨3|\right),
\]
where \(\Omega_p = (E_p d_{12})/\hbar\) and \(\Omega_c = (E_c d_{23})/\hbar\) are the Rabi frequencies of the probe and coupling lasers, with \(E_p\) and \(E_c\) being the electric fields, and \(d_{12}\) and \(d_{23}\) the dipole matrix elements of the corresponding transitions, whereas \(\delta_p\) and \(\delta_c\) are the detunings of the probe and coupling laser from the corresponding transitions, respectively (see Fig. 1). Although we consider a situation where both the coupling and the probe laser are resonant with the atomic transitions they drive (i.e. \(\delta_p = \delta_c = 0\)), by including these detunings one can account for potentially uncompensated electric and/or magnetic fields in case of an experimental realization. The operator \(\mathcal{U}\) which governs the non-Hamiltonian part of the dynamics reads as
\[
\mathcal{U}[\rho] = \frac{\Gamma_{32}}{2} (2\sigma_{13}\rho_{31} - \sigma_{33}\rho_{33} - \rho_{33}\sigma_{33}) + \frac{\Gamma_{31}}{2} (2\sigma_{12}\rho_{21} - \sigma_{22}\rho_{22}) + \frac{\Gamma_{34}}{2} (2\sigma_{43}\rho_{34} - \sigma_{33}\rho_{33}) + \frac{\Gamma_{31}}{2} (2\sigma_{14}\rho_{14} - \sigma_{44}\rho_{44}) + \frac{\gamma_p}{2} (2\sigma_{11}\rho_{11} - \rho_{11}\sigma_{11}) + \frac{\gamma_c}{2} (2\sigma_{33}\rho_{33} - \rho_{33}\sigma_{33}),
\]
where \(\sigma_{kj} = |k⟩⟨j|\) are the atomic projection operators \((k, j \in \{1, 2, 3, 4\})\).

There are multiple sources of non-unitary dynamics in the system. One of them is the spontaneous emission from the first excited state \(|2⟩\) and the Rydberg state \(|3⟩\) which we take into account by introducing radiative decay rates \(\Gamma_{21}\) and \(\Gamma_{32}\). Another source, if present, is a depopulation of the Rydberg state \(|3⟩\) towards the neighboring Rydberg states. The depopulation may occur due to several phenomena depending on the actual realization of the system, such as amplified spontaneous emission and/or superradiance \[24\, 25\] as well as induced emission and absorption due to the blackbody radiation of the environment \[21\, 22\]. Following \[25\], we take these effects into account by the dynamic effective decay rate
\[
\Gamma_{34}(t) = \Gamma_{34,\text{sp}} \cdot [\rho_{44}(t) \cdot p_{\text{sup}} + 1] + \Gamma_{34,\text{bb}},
\]
with \(p_{\text{sup}}\) a superradiance parameter, and \(\Gamma_{34,\text{sp}}\) and \(\Gamma_{34,\text{bb}}\) the effective decay rates caused by spontaneous emission and blackbody radiation, respectively. In our model we assume that the entire population eventually ends up in the ground state \(|1⟩\). This assumption is valid...
provided the ionization from all involved states is negligible.

The above mentioned phenomena cause population-transfer between the atomic states. In contrast, there are a group of processes which do not result in a significant energy-decay in the system but leads to a relevant coherence decay rates \( \gamma_p \) and \( \gamma_c \). Due to the redistribution of population from \( |3\rangle \) to \( |4\rangle \), atoms in Rydberg P states are present in the cloud at various distances. As observed by [24], the dipole-dipole interaction with these P state atoms results in an inhomogeneous broadening of the Rydberg S state. We take this into account by adding an effective dephasing term to \( \gamma_c \),

\[
\dot{\gamma}_c(t) = \gamma_c + \gamma_{3,dd} \cdot \rho_{44}(t).
\]  

The optical response of the cloud under the effect of the two laser pulses is given by the macroscopic polarization \( \vec{P} = \mathcal{N} \text{Tr}[\rho \vec{d}] \) where \( \vec{d} = \sum_{i<j} (d_{ij} |i \rangle \langle j| + \text{h.c.}) \) is the atomic dipole operator. The absorption \( \alpha \) of the probe laser is then given by the imaginary part of the electric susceptibility \( \chi \),

\[
\alpha(t) = \Im(\chi(t)) = \frac{\mathcal{N} d^2_{12}}{\epsilon_0 \hbar \Omega_p} \Im(\rho_{21}),
\]

where \( \mathcal{N} \) is the atom density of the cloud. Note that here we make the approximation that the cloud is homogeneously irradiated and the propagation effects of the laser pulses can be neglected. The time dependent absorption signal is thus given by the master equation, which, using the operators given in Eqs. 4 and 5, reads as

\[
\begin{align*}
\dot{\rho}_{11} &= \frac{i}{2} \left( \Omega_p^* \rho_{21} - \Omega_p \rho_{12} \right) + \Gamma_{21} \rho_{22} + \Gamma_{41} \rho_{44} \\
\dot{\rho}_{22} &= \frac{i}{2} \left( \Omega_p \rho_{12} - \Omega_p^* \rho_{21} - \Omega_c \rho_{23} + \Omega_c^* \rho_{32} \right) - \Gamma_{21} \rho_{22} + \Gamma_{32} \rho_{33} \\
\dot{\rho}_{33} &= \frac{i}{2} \left( \Omega_c \rho_{23} - \Omega_c^* \rho_{32} \right) - (\Gamma_{32} + \Gamma_{34}) \rho_{33} \\
\dot{\rho}_{21} &= \frac{i}{2} [\Omega_c^* \rho_{31} - \Omega_p (\rho_{22} - \rho_{11}) + 2 \delta_p \rho_{21}] - \frac{1}{2} (\Gamma_{21} + \gamma_p) \rho_{21} \\
\dot{\rho}_{31} &= \frac{i}{2} [\Omega_c \rho_{21} - \Omega_p \rho_{32} + 2 (\delta_p + \delta_c) \rho_{31}] - \frac{1}{2} (\gamma_p + \gamma_c + \Gamma_{32} + \Gamma_{34}) \rho_{31} \\
\dot{\rho}_{32} &= \frac{i}{2} \left[ -\Omega_c^* \rho_{31} - \Omega_c (\rho_{33} - \rho_{22}) - 2 \delta_c \rho_{32} \right] - \frac{1}{2} (\gamma_c + \Gamma_{21} + \Gamma_{32}) \rho_{32} \\
\dot{\rho}_{44} &= \Gamma_{34} \rho_{33} - \Gamma_{41} \rho_{44} \\
\dot{\rho}_{41} &= \frac{i}{2} (-\Omega_p \rho_{42} + 2 \delta_p \rho_{44}) - \frac{1}{2} (\Gamma_{41} + \gamma_p) \rho_{41}
\end{align*}
\]

FIG. 2. (Color online) Pulse sequence for the coupling (blue) and probe (red) lasers. At \( t = 0 \) a fraction of the atomic population is prepared in the Rydberg state. In the simulations this fraction is an input value, while in the experiment it is determined by the length of the excitation pulse (Exc.). Before this, an optical pumping pulse (Pump) is used to pump the atoms to the correct polarization. The time evolution of the optical density is monitored after the excitation pulse with the probe laser (A, B, C). The coupling laser is added in time interval B (EIT pulse). The Rabi frequencies are taken from the experimental values in part IV.

\[
\begin{align*}
\dot{\rho}_{42} &= -\frac{i}{2} (\Omega_p^* \rho_{41} + \Omega_c \rho_{43}) - \frac{1}{2} (\Gamma_{21} + \Gamma_{41}) \rho_{42} \\
\dot{\rho}_{43} &= -\frac{i}{2} (\Omega_c^* \rho_{42} + 2 \delta_c \rho_{43}) \\
&\quad - \frac{1}{2} (\Gamma_{32} + \Gamma_{34} + \Gamma_{41} + \gamma_c).
\end{align*}
\]

When there is no coupling to the Rydberg state \( (\Omega_c = 0) \), an analytical steady state solution for \( \alpha \) can be obtained, which will be used for normalization:

\[
\alpha_0 = \frac{\mathcal{N} d^2_{12}}{\epsilon_0 \hbar} \cdot \Gamma_{21} (\gamma_p + \Gamma_{21}) \times \left[ \gamma_p^2 \Gamma_{21} + 2 \gamma_p (\Omega_p^2 + \Gamma_{21}^2) \right]^{-1}.
\]

III. DETECTION OF THE RYDBERG POPULATION: RESULTS OF THE NUMERICAL SIMULATION

We numerically solve the equation system (7) with the pulse sequence of the lasers given by \( \Omega_p(t) \) and \( \Omega_c(t) \) (see the sequence A, B, C in Fig. 2). The solution provides a description of the population dynamics while the atom is being probed by the weak laser on the \( |1\rangle \leftrightarrow |2\rangle \) transition along with a time-dependent coupling between states \( |2\rangle \) and \( |3\rangle \). Furthermore, through Eq. 6 it describes the absorption of the probe laser, which we give relative to
In Fig. 3 we show results for the cases where the atomic states (solid lines) and the relative absorption are different in the different parts of the time evolution. In part A, the absorption of the probe laser is close to zero, and slowly increases while a small fraction of the population decays from the Rydberg state \(|3\rangle\). Since the lifetime of the Rydberg states is much longer than 10\(\mu\)s, the amount of population transferred by spontaneous decay is small although not negligible on the \(\mu\)s timescale of the pulse sequence. At the beginning of part B the population in the selected Rydberg state is transferred to the ground state \(|1\rangle\) through a resonant transfer from state \(|3\rangle\) to \(|2\rangle\) induced by the coupling laser with Rabi frequency \(\Omega_c\) and the consecutive spontaneous emission from \(|2\rangle\) to \(|1\rangle\). For \(\Omega_c < \Gamma_{21}\), this process results in only a small increase in \(\rho_{22}\), because a half Rabi cycle induced by \(\Omega_c\) between states \(|2\rangle\) and \(|3\rangle\) would take longer than the lifetime of state \(|2\rangle\) (see Fig. 3(b)). Hence, assuming the initial population is either in the ground state \(|1\rangle\) or the selected Rydberg state \(|3\rangle\), we can determine the fractions by monitoring the absorption of the probe laser in part A of the time evolution. If there is a way to ensure that all the population missing from the ground state \(|1\rangle\) is in the selected Rydberg state \(|3\rangle\), then this is indeed sufficient. However, if the probability that a fraction of the population is in another state (for example, the interaction scheme to be realized contains more than one Rydberg state), the absorption level in part A of the time evolution is not enough in itself to give information about the population of the selected Rydberg state \(|3\rangle\).

As illustrated in Fig. 3(b) and (c), the absorption of the probe laser in part A is the same for different initial states of the atoms as long as the population in the ground state is the same. In contrast, the dynamics and the equilibrium become significantly different in part B. Since only the population in the selected Rydberg state is transferred back to the ground state by the coupling field, the absorption level in part C also changes with the initial population in \(|3\rangle\).

Another result of the simulations is the possibility to
obtain information on the initial coherence of the system. The time evolution of the relative absorption in the beginning of part B for three different initial preparations of the atoms is shown in Fig. 4. These initial preparations consist of the same fraction of population in the ground state |1⟩ and the selected Rydberg state |3⟩, but the coherence between these two states is different. One of the initial preparations is the mixed state \( \rho_{\text{mix}}(t = 0) = (|1⟩ ⟨1| + |3⟩ ⟨3|)/2 \) and the other two preparations are \( \rho_{\uparrow}(t = 0) = \frac{1}{2}(|1⟩ ± |3⟩)(⟨1| ± ⟨3|) \). According to the numerical calculations, the initial coherence changes the absorption of the probe pulse in the dynamic part of B significantly, but the absorption level during the rest of the pulse sequence is unaffected. If the Rabi frequency \( \Omega_c \) of the coupling laser is on the order of \( \Gamma_{23} \) or higher, oscillations of the absorption signal can be observed.

IV. EXPERIMENTAL SETUP

For demonstrating the detection of Rydberg population with time-resolved EIT, we conducted an experiment on a cloud of \( \approx 2 \times 10^7 \) \(^{87}\)Rb atoms at a temperature of \( \approx 150\mu\text{K} \). In this experiment the atoms are trapped in a magneto-optical trap (MOT), loaded to a magnetic quadrupole trap and then released. The time-resolved measurements are started after 3 ms of time of flight, in order to ensure that all magnetic fields have fully decayed, while effects of atomic motion are still negligible. The measurements are performed within 30 μs (excitation pulse and probe sequence, cf. Fig. 2). The density and optical density at the center of the cloud, measured by absorption imaging, are \( 5 \times 10^9 \) cm\(^{-3} \) and 1.7, respectively.

The transitions from the ground state \(|5S_{1/2}, F = 2⟩\) to the first excited state \(|5P_{3/2}, F = 3⟩\) and from there to the selected Rydberg state \(|3S_{1/2})\) are driven by two lasers with wavelengths of \( \approx 780 \text{ nm} \) (red, probe) and \( \approx 480 \text{ nm} \) (blue, coupling), respectively (see Fig. 4). Additionally, we use a repumper to pump atoms from \(|5S_{1/2}, F = 1⟩\) back to \(|5S_{1/2}, F = 2⟩\) via \(|5P_{3/2}, F = 2⟩\) during the whole pulse sequence. The frequencies of both lasers used in the experiment are referenced to a frequency comb and controlled with slow servo loops (< 100 Hz bandwidth). The linewidths of both lasers are narrowed with fast locks (> 1 MHz bandwidth) to scanning Fabry-Pérot interferometers to less than 20 kHz. As the Fabry-Pérot cavities are sensitive to acoustic noise, the effective linewidth for the experiment can be larger. The red and the blue laser are aligned in a counter-propagating configuration (see Fig. 5). We use an acousto-optic modulator (AOM) in each beam to create the intensity envelopes of the pulses. The switching time of the AOMs is 50 ns (20% to 80% light intensity). The time-dependent intensity of the red laser (probe sig-
nal) is measured with an avalanche photo detector (Thorlabs APD120A/M) and recorded with a digital oscilloscope. The time resolution of the setup is 20 ns.

As shown in Fig. 2 our measurement consists of three main parts. First we put all the atomic population into the Zeeman-sublevel of the ground state matching the polarization of the red laser by optically pumping for 14.5 µs. This pulse is long enough that any transient effects due to the switching of the AOM wear off before the next pulse. Next, we prepare the initial state of the atomic cloud with an excitation pulse. We always apply the red laser for 2 µs with a Rabi frequency of 3.3 MHz. If we want no population to be transferred to the Rydberg state, the blue laser remains switched off and due to the very short lifetime of the first excited state, the entire population practically remains in the ground state. Applying the blue laser with a Rabi frequency of 1.8 MHz for up to 2 µs causes a fraction of the population to be transferred to the Rydberg state, with the most atoms being excited in the case of the full 2 µs pulse. Due to the high Rabi frequency of the red laser there is no coherent excitation of Rydberg atoms. In the third part (probe sequence in Fig. 2), we use the red laser at a low intensity as the probe laser during time intervals A, B and C together with the blue laser as the coupling laser during time interval B (EIT pulse).

As a reference, we always add one experimental cycle without atoms in order to measure the intensity \( I_{\text{ref}}(t) \) of the red laser with the photodetector and another one with atoms but no excitation pulse and no coupling pulse to normalize the data later on. The parameters are then varied from cycle to cycle and the transmitted intensity \( I_T(t) \) of the probe light after passing through the cloud is measured. The experiment is repeated several times for each set of parameters to reduce photo diode noise. The optical density OD is calculated as follows:

\[
OD(t) = -\ln \left( \frac{I_T(t)}{I_{\text{ref}}(t)} \right). \tag{10}
\]

The resulting OD datasets are then normalized by dividing them by the OD dataset that had no excitation pulse and no EIT pulse (OD0):

\[
OD_{\text{rel}}(t) = \frac{OD(t)}{OD_0(t)} = \frac{\alpha(t)}{\alpha_0}. \tag{11}
\]

This relative optical density \( OD_{\text{rel}} \) is comparable to the relative absorption \( \alpha(t)/\alpha_0 \) that is calculated in the numerical simulation. To reduce the effect of the acoustic noise on our Fabry-Pérot cavities, we selected the 30 datasets where the mean transparency in the EIT pulse between 7 µs and 9 µs was maximal. We observe that in all measurements the relative optical density eventually returns to the level before the excitation pulse, ensuring that ionization effects are negligible.

![FIG. 6. (Color online) Measured optical density for the dynamics during the probe sequence. The shaded areas are 95% confidence intervals for the relative optical densities \( OD_{\text{rel}}(t) \) obtained from the measurements by applying Eqs. (10) and (11) for three durations of the excitation pulse \( \{0 \ \mu s \text{ for I, } 0.5 \ \mu s \text{ for II and } 2 \ \mu s \text{ for III} \} \). Solid lines represent fit results for the simulated relative absorption \( \alpha(t)/\alpha_0 \). The reversed order of the absorption signals in part B can be explained by dipole-dipole interactions.](image)

V. DETECTION OF THE INITIAL POPULATION: EXPERIMENTAL RESULTS

We demonstrate the optical detection of population for three different initial Rydberg excitation pulses. The experimental results for the optical density are shown in Fig. 3. In order to compare these results to the model we calculate values for the decay rates matching the chosen combination of states in our experiment (see Fig. 1). We calculate the spontaneous emission rates using the wavefunctions calculated in [18]. \( \Gamma_{32} \) is approximated by summing the spontaneous decay rates from \( |35S_{1/2} \rangle \) to all P states, which results in

\[
\Gamma_{32} = \sum_{n>5} \Gamma_{\text{sp,}35S_{1/2}\rightarrow nP} = 2 \pi \times 3.9 \text{ kHz}. \tag{12}
\]

The main contribution comes from \( |5P_{3/2} \rangle \) (\( \Gamma_{\text{sp,}35S_{1/2}\rightarrow 5P} = 1.2 \text{ kHz} \)) and other low-lying, fast-decaying states. The spontaneous decay rate \( \Gamma_{34,\text{sp}} \) is given by the rate \( \Gamma_{\text{sp,}35S_{1/2}\rightarrow 4P} = 2 \pi \times 16.8 \text{ kHz} \). Here we take only the strongest superradiant transition into account. The transition rate \( \Gamma_{34,\text{bb}} \) is approximated by a sum over all transition rates induced by blackbody radiation from \( |35S_{1/2} \rangle \) to all P states

\[
\Gamma_{34,\text{bb}} = \sum_{n>5} \Gamma_{\text{bb,}35S_{1/2}\rightarrow nP} = 2 \pi \times 2.7 \text{ kHz}. \tag{13}
\]

The rates \( \Gamma_{\text{bb,}35S_{1/2}\rightarrow nP} \) are calculated as in [21 22]. The main contribution to \( \Gamma_{34,\text{bb}} \) comes from neighboring Rydberg states. For the transitions induced by blackbody radiation a temperature of 300 K is assumed. The preceding approximations for the decay rates ensure that the
Similar to the calculation of $\Gamma_{sp}$ retrieved from the fit to the dataset with high excitation (see Fig. 6). The colors match the ones of the states in Fig. 3. The population of the Rydberg state ($35S_{1/2}$) starts to decay immediately after excitation at $t = 0$. The total decay out of the state ($35S_{1/2}$) is modeled correctly. Similar to the calculation of $\Gamma_{32}$ we obtain

$$\Gamma_{41} = \sum_{n>5} \Gamma_{sp,34P \rightarrow nS} = 2\pi \times 0.8 \text{ kHz}.$$  

The Rabi frequencies of the two lasers $\Omega_p = 0.83 \text{ MHz}$ and $\Omega_c = 2.10 \text{ MHz}$ and the coherence decay $\gamma_c = 112 \text{ kHz}$ are fitted to the dataset without Rydberg excitation (see Fig. 6). The Rabi frequencies are consistent with estimates based on beam power and geometry. The noise $\gamma_c$ is mainly caused by the acoustic noise on the Fabry-Pérot cavities. As the fit is only sensitive to $\gamma_p + \gamma_c$ and not to the single values, we choose $\gamma_p = 20 \text{ kHz}$.

An analysis of the datasets with the same conditions, but with Rydberg excitation, allows to fit the fraction of atoms excited to a Rydberg state ($0.494(8)$ and $0.284(10)$) within interval A, where only the red laser is on. The superradiance parameter $p_{\text{sup}} = 7.9(8) \times 10^3$ is fitted to part B and C and the dipole-dipole interaction parameter $\gamma_{3,dd}$ is adjusted in part B. The former scales with the absolute atom number while the latter scales with the atomic density. From the model one can now derive the time-resolved populations of the participating states as it can be seen in Fig. 7. The accuracy of these populations is within ±0.01, which was calculated using a variation method.

VI. CONCLUSION

We have demonstrated the all-optical detection of Rydberg population in a dilute gas, which is an alternative to the methods based on field ionization. Our results show that Rydberg population fractions can be measured with an accuracy of 0.01. By comparing the dynamics of the measured optical densities to our numerical simulations we quantified the decoherence effects occurring in the system, namely blackbody radiation induced transitions, superradiant decay and inhomogeneous broadening due to dipole-dipole interactions.

From our simulations we conclude that the detection scheme can also be used to obtain information on the coherence between the ground state and the Rydberg state. The numerical results predict that future studies using a coherent excitation method and experimental parameters similar to our experiment will be able to detect the initial and time-dependent coherence.

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