

Transverse Photon-Photon Interaction in an Ultracold Atomic Gas

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Masterarbeit in Physik
angefertigt im Institut für Angewandte Physik

vorgelegt der
Mathematisch-Naturwissenschaftlichen Fakultät
der
Rheinischen Friedrich-Wilhelms-Universität
Bonn

September 2026

I hereby declare that the work presented here was formulated by myself and that no sources or tools other than those cited were used.

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Statement on Digital Tools and AI Assistance

In the preparation of this thesis, digital tools including AI-based assistance were used for supporting tasks such as language polishing, code writing for data plotting and analysis and code debugging. All results were verified independently by the author.

Task / Purpose	Tools Used	Role of the Tools
Writing and Language Polishing	ChatGPT	Grammar correction, spelling checks, sentence restructuring for clarity, sentence suggestions, synonym search.
Coding and Debugging	ChatGPT, Perplexity and Grok	Code writing, code writing assistance and code debugging.
Literature Search	ChatGPT and Gemini Notebook	Literature search and summarizing papers.
Figure Making	Matplotlib (Python), Canva and Inkscape	Plotting of data and creation of figures
Data Analysis	SciPy (Python)	Fitting of functions to data.

Contents

1	Introduction	1
2	Quantum Optics with Rydberg Polaritons	3
2.1	Electromagnetically Induced Transparency and Slow Light	3
2.2	Rydberg Polariton Interactions and Correlation Functions	5
2.3	Transverse Interactions	7
3	Rydberg-Atom Experiment with Integrated EMCCD Camera	11
3.1	Experimental Apparatus	11
3.2	Integration into Experimental Time Sequence	13
3.3	Comparing EMCCD to Single Photon Counting Modules	15
4	Characterization and Optimization of Probe Beam Quality	19
4.1	Probe Setup	19
4.2	Beam Collimation	20
4.3	Optical Aberrations	22
4.3.1	Mirror holders	22
4.3.2	Probe lens	23
4.3.3	Polarization control and filtering	27
4.4	Probe Signal Incoupling Efficiency	29
5	Precise Atomic Cloud Positioning and Control Beam Alignment	30
5.1	Light Propagation through a Two-Level System	31
5.2	Longitudinal Cloud Positioning	32
5.3	Methods of Transverse Cloud Positioning	34
5.3.1	SPCM-Assisted Alignment	34
5.3.2	Alignment with EMCCD Camera	35
5.3.3	Alignment via Cloud Splitting	36
5.4	EMCCD-Based Optical-Depth Measurements	37
5.4.1	Background subtraction	37
5.4.2	Comparing SPCMs and EMCCD	39
5.5	Transverse Control Beam Alignment	43
6	Nonlinear Transverse Response of an Interacting Rydberg Gas	46
6.1	Nonlinear Lensing at the Few-Photon Level	46
6.1.1	Pulse Shapes	46
6.1.2	Nonlinear Lensing for Different Rydberg States	47

6.1.3	Optically Controlled Lensing	50
6.2	Towards Photon Counting and Spatial Correlation Measurements	51
6.2.1	Noise Modelling and Thresholding	52
6.2.2	Short Pulsing	55
6.2.3	Readout Correlations	57
7	Conclusion and Outlook	62
	Bibliography	64
A	Appendix	68
A.1	Atom loss during pulse sequence	68

Introduction

Photons interact only very weakly with each other in free space and in most optical media. Strong effective photon-photon interactions therefore generally require a system that mediates the interaction. One platform for realizing such interactions is provided by Rydberg polaritons [1–4]. Rydberg polaritons are hybrid light-matter excitations consisting of a photonic component and a collective atomic excitation involving a highly excited Rydberg state. They are formed when photons propagate through an atomic medium under conditions of electromagnetically induced transparency (EIT) [5]. The photonic component allows the polariton to propagate through the medium, while the atomic component introduces interaction between the polaritons. The interaction between two Rydberg atoms shifts the energy of the pair state, with the shift increasing as their separation decreases [6]. Within a characteristic distance, commonly referred to as the Rydberg blockade radius, the interaction-induced level shift prevents a second atom from being resonantly excited to the same Rydberg state by the same optical fields. Under Rydberg-EIT conditions, this shifts nearby atoms out of the EIT resonance and thereby suppresses the propagation of a second polariton within the blockade radius. Consequently, the interaction between Rydberg excitations is transferred to the propagating photons, resulting in an effective photon-photon interaction [3]. The long-range interactions result not only in blockade but also in nonlinear refraction.

Previous studies of these interactions have primarily focused on regimes in which the blockade radius exceeds the waist of the probe beam, such that the system can effectively be treated as one-dimensional [1, 2]. In contrast, the influence of photon-photon interactions on the transverse spatial distribution of light in ultracold atomic media under Rydberg-EIT conditions has remained largely unexplored. Rydberg polariton interactions are predicted to modify the transverse output profile of the probe field [7]. The transverse output profile is highly influenced by refractive nonlinearities that can arise. The nonlinear refractive response is not restricted to transverse geometries and is also present in the one-dimensional limit. Probe beams wider than the blockade radius allow the study of transverse photon-photon correlations in addition to temporal correlations. In [8] the description of interacting Rydberg polaritons is extended beyond the conventional one-dimensional approximation to investigate correlations arising from the transverse component of the interaction. The theoretical analysis predicts that the Rydberg blockade gives rise to spatial photon anti-correlations in the transverse direction. In contrast to the temporal correlation length, which is affected by the finite EIT bandwidth, the transverse correlation length is primarily determined by the Rydberg blockade radius. Paraxial diffraction can further modify the transverse correlation profile during propagation through the medium.

The aim of this thesis is to investigate these transverse effects experimentally. For this purpose, an electron-multiplying charge-coupled device (EMCCD) camera is integrated into an existing rubidium cold-atom experiment. EMCCD cameras have proven to be useful tools in various areas of quantum optics, for example in measurements of SPDC photon pairs [9–11], Hong-Ou-Mandel interference [12] and the transverse momentum distribution of Rydberg polaritons under EIT conditions [13]. They have also found applications in other fields of physics, such as astronomy [14, 15] and biophysics [16, 17]. In contrast to the previously used single-photon counting modules, the EMCCD camera combines single-photon sensitivity with spatial resolution and therefore allows the transverse photon distribution to be measured. This makes it possible to study changes in the transverse beam profile caused by polariton propagation through the atomic medium and, ultimately, by interactions between Rydberg polaritons.

The thesis is structured as follows. First, an introduction to the concepts of EIT and interactions of Rydberg polaritons is given in Chapter 2. Next, the experimental sequence and the integration of the EMCCD camera into the cold-atom experiment as a single-photon-sensitive detector with spatial resolution are described in Chapter 3. Furthermore, a comparison between the EMCCD camera and single-photon counters for photon number estimation is presented. Chapter 4 summarizes observations with a test setup of the current probe beam. Here it is analysed how different optics affect the transverse photon distribution inside the atom cloud. In Chapter 5, the propagation through a two-level system is analysed. The analysis shows the increased requirement for precise alignment of the atomic cloud and the control beam due to refractions caused by cloud inhomogeneity and provides possible solutions. Chapter 6 presents observations of transverse photon-photon interactions in the mean-field regime. Furthermore, groundwork towards quantum correlation measurements is presented, and potential problems and solutions are discussed. Chapter 7 summarizes the thesis and gives an outlook on the next experimental steps.

Quantum Optics with Rydberg Polaritons

In this chapter, the concept of Rydberg polaritons is introduced. The central idea is to combine electromagnetically induced transparency (EIT) with strongly interacting Rydberg states to create hybrid light-matter excitations that inherit the propagation properties of photons and the strong interactions of Rydberg atoms. Rydberg polaritons provide a platform for effective photon-photon interactions and controlled modification of the temporal and spatial properties of light.

Section 2.1 introduces electromagnetically induced transparency and the associated reduction of the group velocity of light in an atomic medium. In Section 2.2, Rydberg polaritons and their interactions are discussed, together with correlation functions as a tool for characterizing the resulting photon-photon correlations. Finally, Section 2.3 introduces the transverse nature of these interactions and explains how Rydberg-mediated photon-photon interactions can modify the spatial distribution of the light field.

2.1 Electromagnetically Induced Transparency and Slow Light

The response of a medium to an electromagnetic field can be described by the linear susceptibility $\chi^{(1)}$, which is defined as [18]

$$\chi^{(1)} = \frac{P}{\epsilon_0 E} \quad (2.1)$$

with the polarization P and the electric field E . The susceptibility is a complex quantity $\chi^{(1)} = \text{Re}[\chi^{(1)}] + i \text{Im}[\chi^{(1)}]$ and provides a direct connection between the microscopic interaction of atoms with the electromagnetic field and macroscopic observables such as the refractive index and absorption. The amplitude transfer function is given by [19]

$$t(\omega_p, L) = \exp\left(\frac{ikL\chi^{(1)}}{2}\right) \quad (2.2)$$

$$= \exp\left(\frac{ikL \operatorname{Re}[\chi^{(1)}]}{2}\right) \exp\left(\frac{-kL \operatorname{Im}[\chi^{(1)}]}{2}\right) \quad (2.3)$$

with the wave number k , the angular frequency of the light field ω_p and the length of the medium L . The last expression, Equation 2.3, shows that the real part of the susceptibility determines the dispersive response of the medium whereas the imaginary part determines the absorption. The refractive index and transmission can finally be expressed by [19]

$$n(\omega_p) = \sqrt{1 + \operatorname{Re}[\chi^{(1)}]} \quad (2.4)$$

$$T(\omega_p) = \exp\left(-kL \operatorname{Im}[\chi^{(1)}]\right). \quad (2.5)$$

The susceptibility therefore provides a quantitative description of the optical response of an atomic medium and forms the basis for understanding both absorption and dispersion.

For a two-level system, the near-resonant susceptibility in the weak-probe limit is given by [20]

$$\chi_{2\text{-Level}}^{(1)} = -\frac{\sigma_0 \rho(x, y, z)}{k} \frac{2\frac{\Delta_p}{\gamma} - i}{1 + 4\left(\frac{\Delta_p}{\gamma}\right)^2} \quad (2.6)$$

with the detuning $\Delta_p = \omega_p - \omega_{\text{eg}}$ of the probe field from the two-level resonance, the resonant scattering cross-section σ_0 , the atom density $\rho(x, y, z)$ and the natural linewidth of the excited state γ . The real and imaginary parts of the two-level susceptibility are shown in Figure 2.1(a).

A strong control field extends the two-level system to a three-level ladder scheme by coupling the intermediate excited state to an upper state. The three-level susceptibility is then given by [20]

$$\chi_{3\text{-Level}}^{(1)} = \frac{\sigma_0 \rho(x, y, z)}{k} \frac{i\frac{\gamma}{2}}{\frac{\gamma}{2} - i\Delta_p + \frac{\Omega_c^2}{\frac{\Gamma}{2} - i\delta}} \quad (2.7)$$

with the control Rabi frequency Ω_c , the two-photon detuning $\delta = \Delta_p + \Delta_c$ with the control field detuning $\Delta_c = \omega_c - \omega_{\text{re}}$ and the upper-state decoherence rate Γ . The real and imaginary part of the three-level susceptibility for a resonant control beam are illustrated in Figure 2.1(b). In comparison to Figure 2.1(a) it is shown that while the absorption is maximized on probe resonance for the two-level system, the strong control field leads to an electromagnetically induced transparency window. For the three-level system, it can furthermore be seen that, at EIT resonance with $\Delta_p = \Delta_c = 0$, the real part is approximately linear and the group velocity can be written as [19]

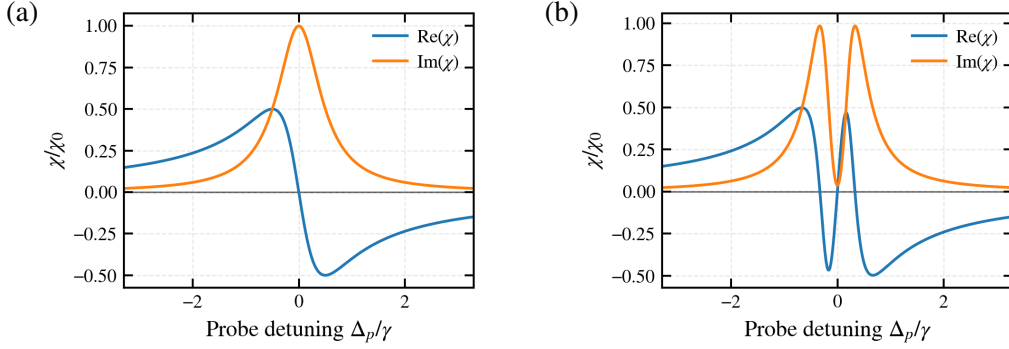


Figure 2.1: Real and imaginary parts of the linear susceptibility. (a) Two-level system. (b) Three-level system in a ladder scheme under EIT conditions.

$$v_{\text{gr}} = \frac{c}{n + \omega_p \frac{dn}{d\omega_p}}. \quad (2.8)$$

As shown in Equation 2.4, the real part of the susceptibility is directly connected to the refractive index of the medium, so a larger slope of the real part on resonance results in a larger dispersion $dn/d\omega_p$. Therefore, under EIT conditions, the propagation speed of light inside the atom cloud can be slowed down. Experimentally, velocities of only a few meters per second have been achieved [21].

2.2 Rydberg Polariton Interactions and Correlation Functions

Atoms with an electron excited to a state with a high principal quantum number n are known as Rydberg atoms. Rydberg atoms have long lifetimes and large orbital radii of their highly excited electrons [22]. A characteristic of Rydberg atoms is their strong interactions [6]. This section introduces the interactions between Rydberg atoms and describes the formation of Rydberg polaritons as coupled light-matter excitations under Rydberg-EIT conditions. Following this, photon-photon correlations are introduced and previous experimental observations of these correlations are discussed.

Since nS Rydberg states have no permanent electric dipole moment, there is no direct first-order dipole-dipole interaction between two atoms in these states. Instead, their interaction arises from second-order dipole-dipole coupling, resulting in a van der Waals interaction. The interaction potential for two Rydberg atoms separated by a distance R in the same nS state is described by [23]

$$U_{\text{vdW}} = \frac{C_6}{R^6} \quad (2.9)$$

where C_6 is the van der Waals coefficient, which scales with the principal quantum number as n^{11} .

An important concept of Rydberg interactions is the blockade mechanism. Because of the interaction-

induced level shift, the excitation of a second Rydberg atom can be shifted out of resonance depending on its distance R from the first Rydberg excitation. The corresponding blockade radius can be expressed as [24]

$$r_b = \left(\frac{|C_6|}{\hbar\Omega} \right)^{1/6} \quad (2.10)$$

with the single-atom coupling strength Ω . The combination of the Rydberg blockade mechanism with EIT enables effective interactions between photons, as a Rydberg excitation can modify the EIT condition experienced by nearby photons.

For few-photon pulses, propagation under EIT conditions can be described by a quasiparticle referred to as a dark-state polariton, which is a superposition of a photonic component and an atomic excitation [5]. A Rydberg polariton is created by choosing a Rydberg state as the upper state of the ladder scheme. Because photonic propagation is combined with the blockade effect, the EIT condition can be broken locally. The EIT condition is broken when the interaction-induced level shift exceeds the EIT linewidth. Here, the blockade radius can be expressed as [8]

$$r_b = \left(\frac{|C_6|}{2\hbar\Gamma_{\text{EIT}}} \right)^{1/6} \quad (2.11)$$

with the EIT linewidth Γ_{EIT} .

The resulting photon-photon interactions can be characterized by second-order correlation functions. Photon correlations can be measured in time and in space. For temporal correlations, the normalized second-order intensity correlation function of a light field can be expressed as [25]

$$g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle \langle I(t+\tau) \rangle}. \quad (2.12)$$

At the few-photon level, the intensity correlation is experimentally determined from photon-counting statistics, where coincidences between individual photon detection events are measured instead of continuous optical intensities. This gives the two-photon correlation function. For Poisson-distributed light, the correlation function yields $g^{(2)}(\tau) = 1$. If the blockade radius exceeds the probe-beam waist, the system can be treated as effectively one-dimensional, and Rydberg-mediated photon-photon interactions mainly result in temporal correlations between the transmitted photons.

Experimentally, different types of temporal correlations have been realized. At EIT resonance ($\Delta_p = \Delta_c = 0$) and high optical depth within a blockade volume, OD_b , the propagation of a second photon is strongly suppressed. Within the blockade radius, the Rydberg interaction destroys the EIT condition for the second photon, leading to enhanced absorption and temporal antibunching of the probe light [1]. This antibunching is characterized by $g^{(2)}(0) < 1$. The width over which $g^{(2)}(\tau)$ deviates

from unity defines the correlation time.

In contrast to this dissipative photon-photon interaction, primarily dispersive interactions have been realized with single-photon detunings exceeding the linewidth of the intermediate state ($|\Delta_p|, |\Delta_c| \gg \gamma$). The Rydberg-mediated interaction can then be described by an effective potential between the photons. For suitable detunings, this potential becomes attractive and can support a two-photon bound state. Experimentally, this bound state manifests itself as an enhanced probability of detecting two photons at small temporal separations, corresponding to photon bunching with $g^{(2)}(0) > 1$ [2].

While temporal correlations characterize photon separations along the longitudinal direction, spatially resolved detection also allows correlations between their transverse positions to be investigated. The corresponding spatial correlations are introduced in the following section.

2.3 Transverse Interactions

While longitudinal photon-photon interactions have been studied extensively [1–3], transverse interactions under Rydberg-EIT conditions remain comparatively unexplored. In the following, two approaches to studying transverse interactions are considered: dispersive changes of the mean optical field and correlations in the two-photon state. The polarizability P of the medium in Equation 2.1 linearly depends on the atom density [26]. Since the atomic cloud in typical ultracold-atom experiments follows a Gaussian density distribution, the cloud inhomogeneity together with the detuning-dependent refractive index (Equation 2.4) can lead to focusing or defocusing relative to free-space propagation without the atomic cloud.

Changes of the refractive index by polaritons inside the atom cloud were already used for creating an attractive potential for photons [2]. This Rydberg-mediated refractive nonlinearity can also be used to modify the mean-field transverse photon distribution of the probe beam. A second photon inside the blockade volume experiences the susceptibility of a three-level system whose upper state is shifted by more than the EIT linewidth. Under this condition, the optical response can be approximated by the two-level susceptibility. The susceptibilities for control-field detunings of ± 20 MHz are shown in Figure 2.2. In both cases, it can be observed that there is a small window in which the sign of the real part of the linear susceptibility is opposite from the two-level susceptibility. By choosing the probe detuning where this difference is maximized, a strong Rydberg-mediated refractive nonlinearity can be obtained, for which the effective refractive index experienced by the probe field depends on the photon rate.

An analytical theory of these refractive nonlinearities is derived in [7]. It is shown that the susceptibility can be expressed as

$$\chi(\mathbf{r}) = -\frac{12\pi\gamma\rho^2}{k^3\Delta_p\Omega_c^2} \int d\mathbf{r}' \left[\frac{|\Omega_p(\mathbf{r}'_\perp, z)|^2}{1 + \frac{|\mathbf{r}' - \mathbf{r}|^6}{r_b^6}} - i \frac{\gamma}{\Delta_p} \frac{|\Omega_p(\mathbf{r}'_\perp, z)|^2}{\left(1 + \frac{|\mathbf{r}' - \mathbf{r}|^6}{r_b^6}\right)^2} \right] \quad (2.13)$$

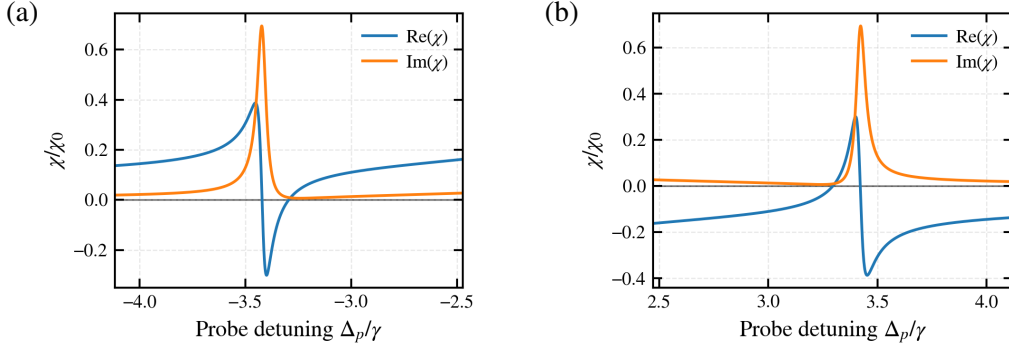


Figure 2.2: Real and imaginary parts of the linear susceptibility for a three-level ladder scheme under EIT conditions for the D2 line of ^{87}Rb . (a) For $\Delta_c/(2\pi) = 20$ MHz. (b) For $\Delta_c/(2\pi) = -20$ MHz.

with the atom density ρ , the transverse position $\mathbf{r}'_\perp$ and the probe Rabi frequency Ω_p . By substituting $\tau = \frac{y}{kr_b^2}$ and $\xi = \mathbf{r}'_\perp/r_b$ with y being the coordinate of beam propagation, they derive the two-dimensional Schrödinger equation

$$i\frac{\partial}{\partial\tau}\Omega(\xi, \tau) = \left[-\frac{\nabla_\xi^2}{2} + \alpha \int d\xi' |\Omega(\xi', \tau)|^2 U_1(\xi - \xi') - i\frac{\gamma}{\Delta_p} \alpha \int d\xi' |\Omega(\xi', \tau)|^2 U_2(\xi - \xi') \right] \Omega(\xi, \tau) \quad (2.14)$$

with the normalized probe amplitude Ω and the effective interaction potential $U_m(\xi)$. The parameter α describes the nonlinearity strength and can be expressed by

$$\alpha = \frac{36\pi^2 \rho^2 \gamma^2 C_6 P_p}{\hbar k^4 c r_b^3 \Omega_c^4} \quad (2.15)$$

with the probe power P_p . Experimentally, the nonlinear strength can be adjusted through the control-field Rabi frequency and, through the chosen Rydberg state and EIT conditions, the blockade radius.

Besides the mean-field effects, transverse quantum correlations can occur. A theoretical study of transverse correlations mediated by the Rydberg blockade mechanism is given in [8]. In the following, a brief introduction to this theory is given. The main concept is illustrated in Figure 2.3. Instead of choosing a Rydberg state such that the blockade radius exceeds the probe waist, a blockade radius is chosen that is smaller than the probe waist. This allows the formation of polaritons on top of each other, breaking the well-studied one-dimensional case. For longitudinal measurements, single-photon counters have been used, for example, in a Hanbury Brown-Twiss setup to measure correlations in the time domain. In the dissipative regime, the underlying interaction produces temporal antibunching. In contrast to that, a single-photon-sensitive device that offers spatial resolution is necessary for the

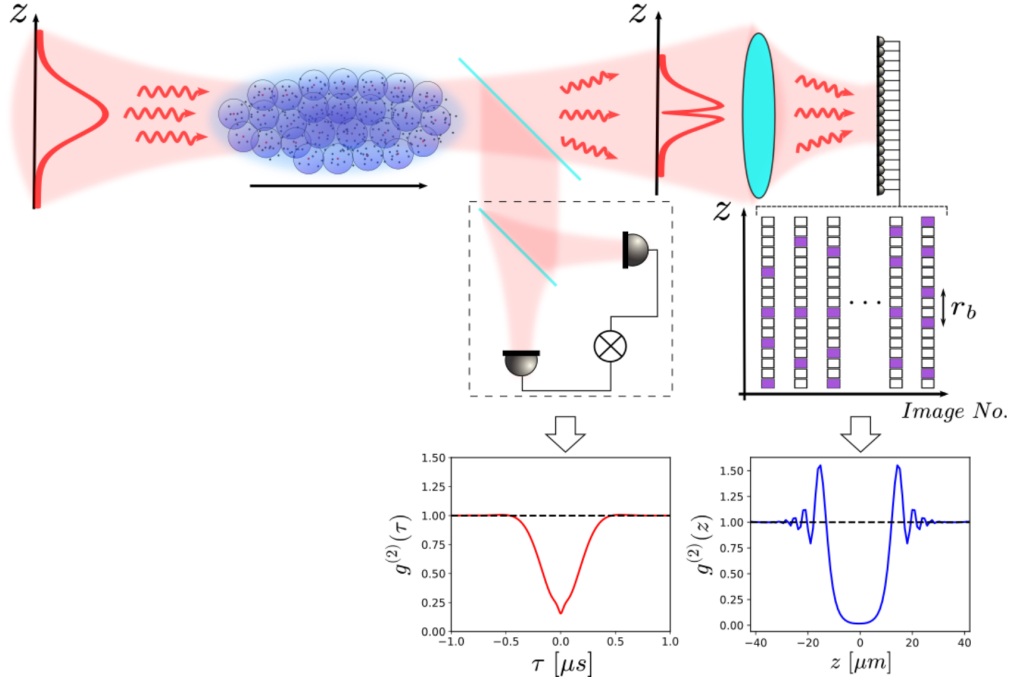


Figure 2.3: Probe photons propagating through an ultracold ensemble with a probing waist that is smaller than the blockade radius. The longitudinal interactions can be measured with a Hanbury Brown-Twiss (HBT) setup to extract a temporal correlation function. The transverse interactions can be measured with a single-photon-sensitive detector with spatial resolution. The recorded positions can be used to reconstruct the transverse second-order photon-correlation function. Adapted from [8].

measurement of transverse photon correlations.

A theoretical description of these transverse correlations starts from the full two-polariton system, which is represented by nine coupled propagation equations for the amplitudes EE , EP , ES , PE , PP , PS , SE , SP , and SS [8]. Here, E , P , and S denote the photonic component, the excitation of the intermediate state, and the excitation of the Rydberg state, respectively. To reduce this system, symmetric and antisymmetric combinations of the amplitudes are introduced

$$ES_{\pm} = \frac{ES \pm SE}{2}, \quad PS_{\pm} = \frac{PS \pm SP}{2}, \quad EP_{\pm} = \frac{EP \pm PE}{2}. \quad (2.16)$$

For a homogeneous medium and under the assumptions $\Gamma = 0$ and $\Delta_p = 0$, the amplitudes PP , $PS_{\pm}$, $EP_{\pm}$, and SS can be eliminated. This results in a coupled Dirac-like equation for ES_{+} and ES_{-} . By adiabatically eliminating the antisymmetric component ES_{-} , the system can be reduced to a single effective two-photon Schrödinger-like equation in the longitudinal and transverse relative coordinates [8]

$$\frac{\partial}{\partial Y}\psi = \frac{4L}{\text{OD}}\frac{\partial^2}{\partial y^2}\psi + \frac{i}{k}\frac{\partial^2}{\partial z^2}\psi - \frac{\text{OD}}{L}V^{(2)}(y, z)\psi, \quad (2.17)$$

where $Y = (y_1 + y_2)/2$ is the longitudinal center-of-mass coordinate, $y = y_1 - y_2$ and $z = z_1 - z_2$ are the longitudinal and transverse relative coordinates, respectively, OD denotes the optical depth and $\psi = ES_+$ is proportional to the two-photon amplitude EE . The effective Rydberg interaction potential is given by

$$\frac{1}{V^{(2)}(y, z)} = 1 - i \left[\left(\frac{y}{\tilde{r}_b} \right)^2 + \left(\frac{z}{\tilde{r}_b} \right)^2 \right]^3, \quad (2.18)$$

with $\tilde{r}_b = r_b/\sqrt[3]{2}$ [8]. The different coefficients of the two second-order derivatives reveal the different correlation mechanisms. In the longitudinal direction, the real coefficient leads to a diffusion-like broadening of the blockade-induced depletion and therefore to a correlation length that depends on the atomic response and the finite EIT bandwidth. In the transverse direction, the imaginary coefficient describes paraxial diffraction. Consequently, the transverse correlation length remains mainly determined by the Rydberg blockade radius, while diffraction only weakly modifies the resulting spatial correlation profile.

Rydberg-Atom Experiment with Integrated EMCCD Camera

Measuring the transverse photon distribution with spatial resolution at the few-photon level requires a single-photon-sensitive imaging detector. For example, EMCCD cameras provide a possibility to record a spatial photon distribution at very low light levels. This device combines spatially resolved photon detection with the high acquisition rate required to perform multiple measurements within a single experimental cycle. However, while the camera provides information about the transverse photon distribution, the signal recorded during each pulse is integrated over time, and any information about the temporal distribution of the detected photons is therefore lost. This chapter introduces the experimental setup and the integration of the EMCCD camera into the experiment. Finally, the performance of the EMCCD camera as a single-photon-sensitive detector is compared with that of the single-photon counting modules (SPCMs) previously used in the experiment.

3.1 Experimental Apparatus

This section provides a brief introduction to the steps of the experimental sequence that are relevant to this work. More detailed information is provided in the PhD theses of H. Gorniaczyk and L. Ahlheit [27, 28].

The different steps of the experimental sequence are illustrated in Figure 3.1. A rubidium dispenser creates a background gas pressure in the vacuum cell. The rubidium-87 atoms are then cooled and trapped using a magneto-optical trap (MOT). Six independent cooler and repumper beams are used in this process and a magnetic field introduces a position-dependent Zeeman splitting. The cooling transition is $F = 2 \rightarrow F' = 3$ and the repumper addresses the $F = 1 \rightarrow F' = 2$ transition of the D2-line ($5S_{1/2} \rightarrow 5P_{3/2}$) at 780 nm. This is followed by a MOT compression where the magnetic field gradient and the detuning of the cooling beams are increased. Next, the atoms are loaded into a far-detuned crossed optical dipole trap (ODT) operating at a wavelength of 1 064 nm, which further increases the atomic density [29]. The angle between the two arms is 31° , creating an elongated atomic cloud. After that, a combination of evaporative cooling and Raman sideband cooling is used to reduce the cloud temperature further. For the evaporative cooling, the power of the ODT beams, and hence the trapping potential, is reduced, resulting in the loss of the hottest atoms. More details about the Raman

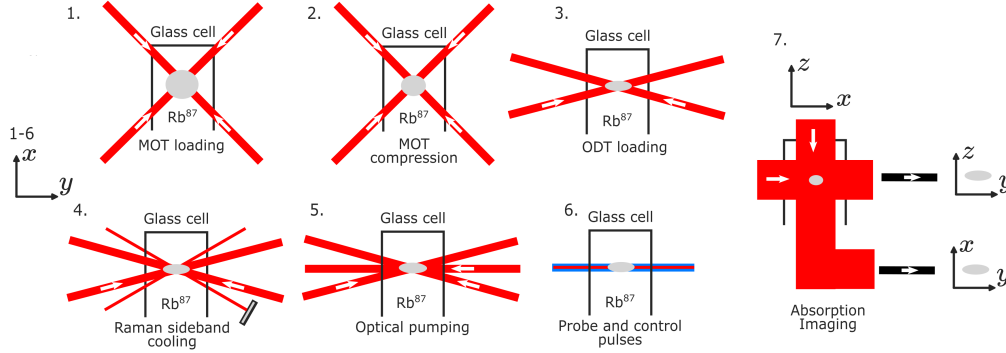


Figure 3.1: Schematic of the experimental cycle. 1. MOT loading using six cooling beams and six repumping beams. The beams along the z -axis are not shown in this figure. 2. MOT compression by ramping up the magnetic field gradient and increasing the detuning of the cooler beams. 3. The atoms are loaded into the crossed optical dipole trap, creating an elongated atomic cloud. 4. Raman sideband cooling with three laser beams. One beam along the z -axis is not shown in the figure. One horizontal beam is reflected. 5. Optical pumping with one laser beam. 6. Counter-propagating probe and control pulses. During the pulses, the optical dipole trap is turned off to avoid introducing a differential light shift. 7. Destructive absorption imaging. At the end of the sequence, the cloud is imaged with two large laser beams from different directions.

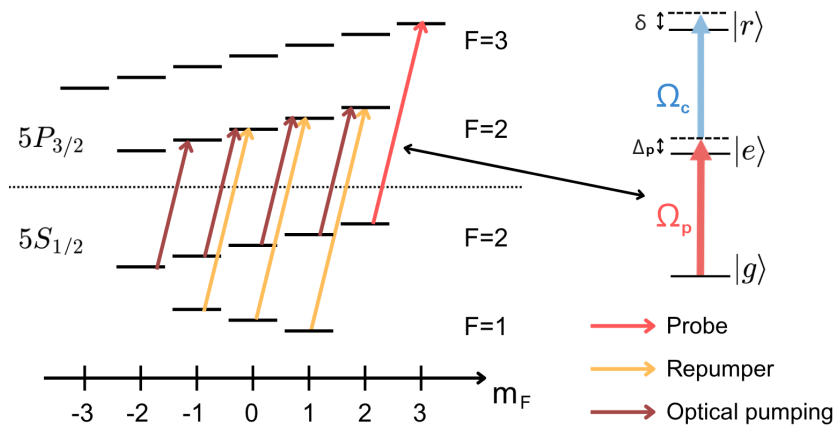


Figure 3.2: Level scheme for optical pumping and probe transition to the intermediate state. An optical pumping beam is used to drive the $|F = 2, m_F\rangle \rightarrow |F' = 2, m_F + 1\rangle$ transition. The repumper addresses the $F = 1 \rightarrow F' = 2$ transitions. The probe beam drives the closed transition $|F = 2, m_F = 2\rangle \rightarrow |F' = 3, m_F = 3\rangle$. The control field couples the intermediate state to the Rydberg state. Δ_p denotes the probe detuning and δ the two-photon detuning.

sideband cooling are given in [30]. The last step of the atom preparation is to pump the atoms into the $|F = 2, m_F = 2\rangle$ hyperfine ground state. The level scheme is shown in Figure 3.2. Here, a pump beam together with an overlapped repumper beam is used. With this step, the atom preparation is completed.

For the experiment, weak probe pulses at 780 nm are combined with a blue control laser at 480 nm. Each experimental cycle contains a sequence of short probe-control pulses. Typically, around 1000 pulses were applied in a time of 100 ms. The pulse duration is on the order of 10 μ s. During each pulse,

the ODT is turned off to avoid introducing a differential light shift. The ODT is turned on in between the pulses to prevent the atom cloud from falling and to preserve its shape and density. After each pulse, field ionization is used to remove any remaining Rydberg excitation. At the end of the sequence, the atomic cloud is imaged by resonant absorption imaging. Two orthogonal imaging beams are used to image the cloud in two different planes and therefore determine the cloud widths along all three spatial axes. For each experimental run, three images are acquired: an absorption image with atoms, a reference image of the imaging beam without atoms, and a dark image. With these images, the optical column density for each pixel is calculated as [28]

$$\text{OD} = -\ln \left(\frac{I_{\text{absorption}} - I_{\text{dark}}}{I_{\text{reference}} - I_{\text{dark}}} \right). \quad (3.1)$$

At the end, the pulse sequence is repeated without an atomic cloud to obtain a reference measurement for comparison with the atom-containing pulses.

3.2 Integration into Experimental Time Sequence

For the integration of an EMCCD camera into a cold-atom experiment, fast image acquisition is needed. The image acquisition rate can be increased using the frame transfer mode of the EMCCD camera. The working principle is illustrated in Figure 3.3(a). The sensor is divided into an imaging area and a storage area. The storage area is protected from light exposure. Once the exposure of the imaging area ends, the accumulated charges are shifted vertically into the storage area. After each vertical shift, a new line is shifted into the readout register. Then the charges are shifted horizontally through the readout register and the multiplication register until the readout register is empty again and the next vertical shift can follow. During the measurements presented in this work, only a 32×32 pixel region of the total 512×512 pixel sensor was used. This procedure allows the acquisition of the next image before the previous image has been read out and reduces the number of shifts, which can reduce the dead time to less than a millisecond. An OptoMask, consisting of adjustable masking blades that restrict the illuminated sensor area, was used to cover the remaining part of the imaging area.

The final frame rate is determined by the horizontal and vertical shift speed. The vertical shift speed (VSS) was set to $0.5 \mu\text{s}$ per line, which is the second-fastest vertical-shift setting available for the camera. Besides the VSS, the vertical shift amplitude (VSA) has to be chosen. The VSA determines the amplitude of the clocking voltage used to transfer charge vertically from one row of the EMCCD sensor to the next. The possible VSA settings are 'Normal', '+1', '+2' and '+3' from lowest to highest amplitude. A higher VSA increases the number of clock-induced charges and therefore the overall camera noise. Clock-induced charges are discussed in more detail in Subsection 6.2.1. However, if the VSA is set too low, charge smearing can occur and significantly degrade the image quality. This effect is shown in Figure 3.4. The VSA setting '+1', which was used for all measurements in this work, produces images without visible charge smearing, whereas clear smearing is observed for the 'Normal' setting. Higher VSAs did not increase the image quality. The highest vertical shift speed was not chosen since it demanded a VSA of '+3' instead of '+1' for eliminating charge smearing while only slightly increasing the possible frame rate. The horizontal shift speed (HSS) has to be chosen, which determines how fast the charge of each pixel is transferred through the readout register. A higher HSS increases the frame

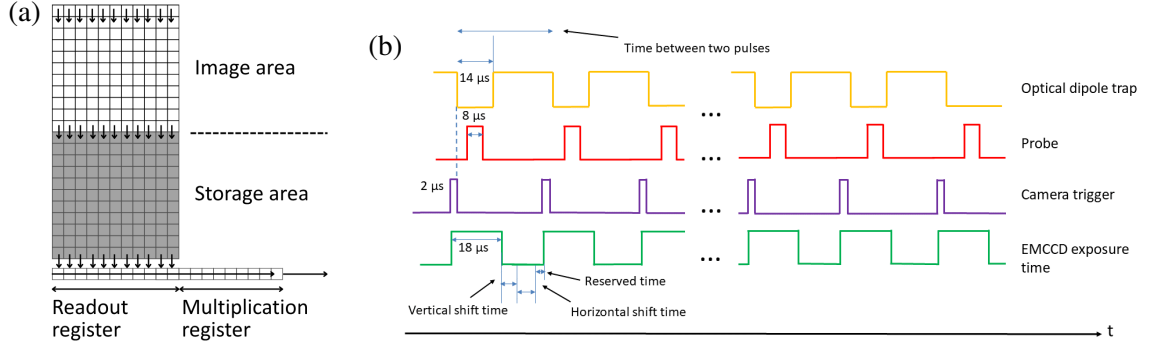


Figure 3.3: (a) Working principle of fast image acquisition. The charges are shifted vertically from the image area into the storage area. The last line in the storage area is shifted into the readout register, where the charges are shifted horizontally into the multiplication register. (b) Experimental time sequence showing ODT and probe power, camera trigger signal and the camera exposure time. The time scale is schematic and not linear as the time between two pulses is approximately $445\ \mu\text{s}$.

rate while also increasing the readout noise in a single image. The maximum HSS of $17\ \text{MHz}$ was selected.

The measurement window was extended to $200\ \text{ms}$ instead of the previously used $100\ \text{ms}$ [28]. The lifetime of the optical dipole trap was measured to be $21\ \text{s}$, which is much longer than the measurement window. The main source of atom loss is not the finite dipole trap lifetime but the repeated pulsing of the trap, because not all atoms are recaptured after each pulse. In this window, 450 pulses are sent and images are taken with the EMCCD camera. The timing sequence is illustrated in Figure 3.3(b). The probe pulse is centered in the $14\ \mu\text{s}$ time window where the dipole trap is turned off. The camera trigger starts $2\ \mu\text{s}$ before the ODT is turned off such that the probe is also centered within the camera exposure time. After that, vertical and horizontal shifts are performed. The time between two pulses cannot be shorter than the time required to transfer the charges from all pixels from the image area into the storage area. Once the shifts are completed, the next pulse can start.

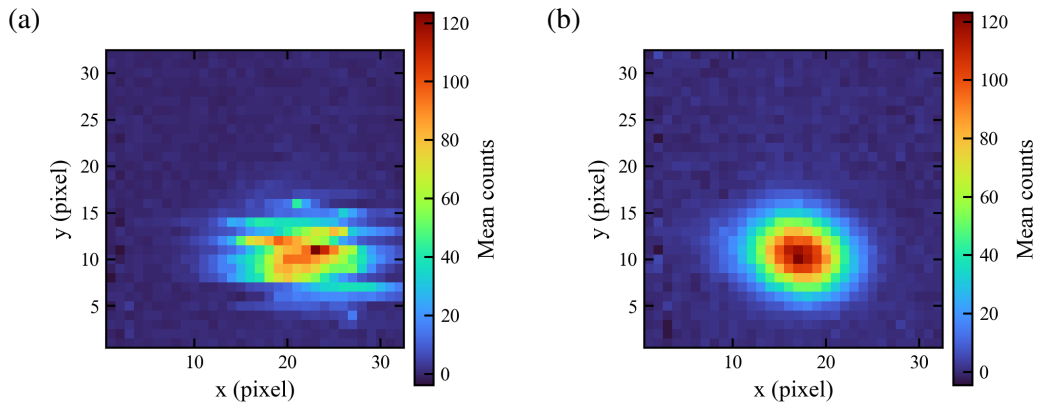


Figure 3.4: EMCCD image averaged over 13500 probe pulses. (a) with VSA setting 'Normal'. (b) with VSA setting '+1'.

3.3 Comparing EMCCD to Single Photon Counting Modules

In previous iterations of the experiment, single-photon counting modules¹ were used for experimental readout. Besides resolving the spatial photon distribution, the EMCCD camera also provides a second way of photon counting in the experiment. The approach to compare both devices is to reconstruct the mean photon number per pulse at the position of the atomic cloud. To perform such a measurement, both devices must be calibrated. The comparison was performed twice with different electron-multiplier (EM) gains.

To reconstruct the number of photons at the position of the atomic cloud, the fraction of photons lost between the cloud and the detector must be estimated. The few-photon pulses are generated with an acousto-optical modulator (AOM) followed by neutral density filters with a high optical density. Removing the neutral density filters from the setup allows the measurement of optical losses inside the probe path with classical light. The probe power is measured before (P_b) and after (P_a) the vacuum chamber as shown in Figure 3.5. Between the two measurement points there are losses due to the two probe lenses and the entrance and exit windows of the vacuum cell. Assuming equal transmissions T before and after the cell, the power inside the vacuum cell (P_i) can be calculated using

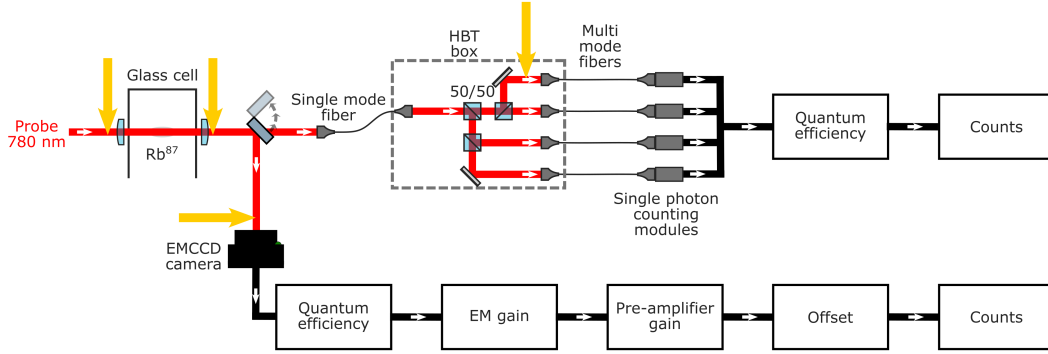


Figure 3.5: Setup for photon counting device comparison. The yellow arrows mark all positions where the probe power is measured. The probe beam power is measured before and after the chamber to calculate the power inside the chamber. A flippable mirror is used to control which device is used. For the EMCCD path, the power is measured before the EMCCD camera to calculate the optical losses. Furthermore, the quantum efficiency of the camera, the EM gain, the pre-amplifier gain and the offset have to be taken into account for the final counts. For the SPCM path, the light is collected by a single-mode fiber and guided into an HBT box. After optimizing the fiber coupling, the power is measured before each multimode fiber to take optical losses into account. A coupling efficiency of 90% was assumed for each multimode fiber. To reconstruct the photon number from the final counts, besides optical losses, the quantum efficiencies of the counting modules must be taken into account.

$$P_a = T^2 P_b, \quad P_i = T P_b = P_b \sqrt{\frac{P_a}{P_b}} = \sqrt{P_b P_a}. \quad (3.2)$$

¹ Excelitas SPCM-AQRH-13-FC

A flippable mirror is used to switch between the SPCMs and the EMCCD camera as a detection device. To determine the transmission of the EMCCD path, the power measured directly before the camera is divided by the calculated power inside the vacuum cell. In the SPCM path, the probe photons are collected by a single-mode fiber and guided into a Hanbury Brown-Twiss (HBT) box. Here, two beam splitters are used to equally distribute the light among the four detectors. The light is then guided to the individual SPCMs using four multimode fibers.

The full reconstruction of the average number of photons inside the vacuum cell per pulse is illustrated in Figure 3.5. Besides the optical losses, the quantum efficiencies of the single-photon counting modules must be taken into account. For the EMCCD path, the reconstruction requires additional steps. First, the quantum efficiency of the camera has to be taken into account which is approximately 80% [31]. The original electron signal is then multiplied by the EM gain and converted into digital counts by a pre-amplifier. Finally, an offset is added, yielding the recorded number of digital counts. Summing over all camera pixels gives the total number of counts, which is used to calculate the number of photons inside the vacuum cell.

First comparison

The measurements were performed in the vacuum cell without a loaded atomic cloud. Eleven different AOM settings with different voltages were used to vary the number of photons for an 8 μ s pulse. First, the average photon number per pulse was measured using the SPCMs over 90000 pulses for each AOM setting. The obtained values were background-corrected by an additional measurement in which a mechanical shutter prevented the transmission of probe light. The background-corrected values ranged from 0 to 68 photons per pulse. After that, the same procedure was repeated for all eleven settings with the EMCCD camera. The pre-amplifier gain was set to 4.92 electrons per count. It was discovered that there was a significant mismatch between the EM gain that was set in the software and the effective EM gain in the measurements. To determine the effective EM gain, dark images were used to fit the exponential tail in the noise distribution caused by clock-induced charges (CIC). The EMCCD noise sources are discussed in more detail in Subsection 6.2.1. The mean dark-count value of each pixel was subtracted from the corresponding pixel value resulting from the bias offset. The background subtraction procedure is further explained in Subsection 5.4.1.

The histogram of background-subtracted pixel values is shown in Figure 3.6(a). Only counts above a certain threshold were taken into account to strongly suppress the contribution of readout noise. The EM gain was extracted from the exponential fit using [32]

$$N(x) = A \exp\left(-\frac{x}{\tau}\right), \quad G_{\text{EM}} = 4.92 \tau \quad (3.3)$$

where x is the number of counts, A is the amplitude, which depends on the number of images that are included and τ depends on the EM gain and the camera conversion factor resulting from the pre-amplifier gain which was set to 4.92 electrons per count. An effective EM gain of 360 was obtained, whereas the software setting was 1000, showing the need for regular recalibration due to camera aging and separate extraction of the EM gain from the CIC-induced noise distribution. The measured photon numbers for all AOM settings and both detectors at the position of the atom cloud are shown in Figure 3.6(b). For

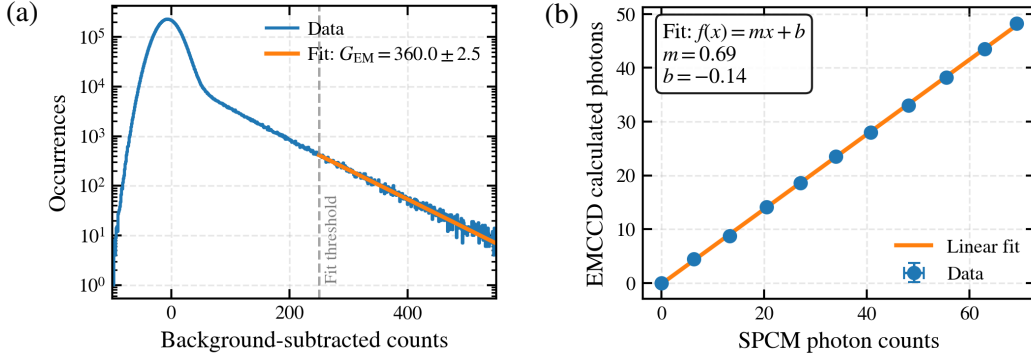


Figure 3.6: First Photon Counting Device Comparison. (a) Extraction of EM gain from amplifier noise distribution. (b) Comparison of EMCCD camera and SPCM result for 11 different AOM settings. The number of photons estimated with the EMCCD camera is approximately 31% lower than the estimated number with SPCMs.

direct comparison, a linear fit was performed showing that the EMCCD-based estimation of the photon number is 31% lower than the SPCM-based estimation. Since the true number of photons is unknown, only the relative difference between the two devices can be determined.

Second comparison

To validate this result, the procedure was repeated at a later time. During this period, the EM gain was recalibrated using the software provided by Andor for this purpose [33]. First, the photon number was measured for each AOM setting, the measurements covered a range from 0 to 69 photons estimated with the SPCMs. The results are shown in Figure 3.7. It can be seen that the EM gain is now more than twice as high as it was in the first measurement but that there is still a significant deviation from the software setting. The linear fit shows that the EMCCD-based estimation of the photon number is 27% lower than the SPCM-based estimation. So in both cases, the EMCCD-based photon-number estimates are approximately 30% lower than the SPCM-based estimates.

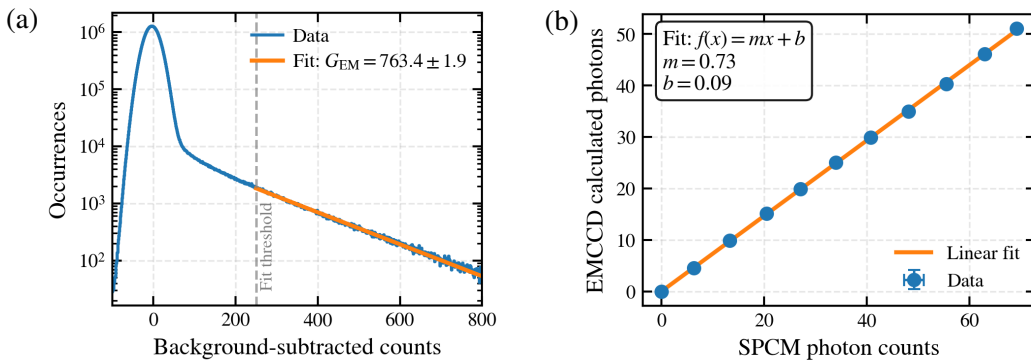


Figure 3.7: Second Photon Counting Device Comparison. (a) Extraction of EM gain from amplifier noise distribution. (b) Comparison of EMCCD camera and SPCM result for 11 different AOM settings. The number of photons estimated with the EMCCD camera is approximately 27% lower than the estimated number with SPCMs.

The systematic deviation may originate from one or more calibration factors shown in Figure 3.5. The losses and EM gain were measured separately for both measurements and there is no clear indication that these quantities caused the systematic deviation. The conversion factor is directly linked to the EM gain estimate. A systematic deviation of the pre-amplifier gain would therefore just be cancelled out by a systematic deviation of the extracted EM gain in opposite direction. The quantum efficiency of the counters was assumed to be 64% according to their specifications [34]. Since they were used over multiple years, degradation is possible. However, to explain the difference between the two photon counting devices, the efficiency would have to be substantially higher, exceeding 80%. The quantum efficiency of the EMCCD camera is another possibility. But here a real quantum efficiency of below 60% would be necessary to explain the deviation between the two counting devices. Additional electron losses due to the charge shifts in the registers are also a possible explanation. To test this, the VSA was increased, which raises the electrostatic potential barriers between adjacent charge-storage regions during vertical transfer and therefore reduces the probability of charge loss during the shifting process. However, there was no observable increase in the EMCCD counts. A systematic deviation of the assumed multimode fiber coupling is also possible but this can only explain a fraction of the total deviation.

In summary, the comparison between the EMCCD camera and the SPCMs reveals a reproducible relative discrepancy of approximately 30%. The quantum efficiency of both devices and the multimode fiber coupling to the SPCMs were not calibrated independently in either comparison, therefore, they are possible sources of this deviation. Possible losses during the charge transfer can also reduce the photon counts for the EMCCD camera but tests with different vertical shift amplitudes did not indicate loss. Currently, superconducting single-photon detectors are being prepared for integration into the experimental setup, providing an opportunity for a third photon counting device for future comparison.

Characterization and Optimization of Probe Beam Quality

The investigation of transverse photon-photon interactions relies on a well-defined probe-beam profile that is ideally close to that of a Gaussian beam. In particular, distortions or aberrations introduced during beam propagation can complicate the interpretation of interaction-induced changes in the transverse beam profile. A detailed understanding of the optical components and their influence on the beam propagation is therefore essential for distinguishing such effects from changes caused by the optical setup itself. In this chapter, the influence of the relevant optical elements on the probe-beam profile is investigated in detail. First, the probe setup is introduced in Section 4.1. Next, the collimation of the outcouplers is optimized in Section 4.2. The influence of different optical elements on the probe-beam focus quality is investigated in Section 4.3. Finally, a possible improvement to the probe-photon collection efficiency is discussed in Section 4.4.

4.1 Probe Setup

The probe setup prepares and guides the probe light through the atomic cloud and then to the different detection devices. An overview of the complete optical path is shown in Figure 4.1. After the laser pulses are prepared on the laser table, the probe light is guided to the experimental table with an optical fiber and collimated by an outcoupler.¹ A polarization beam splitter (PBS) is used for polarization filtering since the polarization after the outcoupler can slowly drift over time. The half-waveplate (HWP) and quarter-waveplate (QWP) prepare the light polarization to drive the closed transition shown in Figure 3.2. The probe beam then passes two dichroic mirrors² which are used to couple the 480 nm control light and the 1 020 nm light used in previous work for a magic lattice trap [28, 35], into the beam path. A lens³ with a focal length of $f = 50$ mm then focuses the probe beam onto the atomic cloud to a waist below $5\text{ }\mu\text{m}$. Then the beam is recollimated. After passing the same dichroic mirrors, waveplates, and PBS again the beam is guided to different measurement devices where the exact path is controlled by mirrors in flip mounts. The EMCCD path contains a $f = 750$ mm lens that, together with the EMCCD and the recollimating $f = 50$ mm lens, forms a telescope imaging a plane located shortly behind the

¹ Thorlabs TC25APC-780

² Thorlabs DMLP650 and DMSP950

³ Edmund Optics 12.5 mm dia. $\times$ 50 mm FL, VIS-NIR Coated, Achromatic Lens

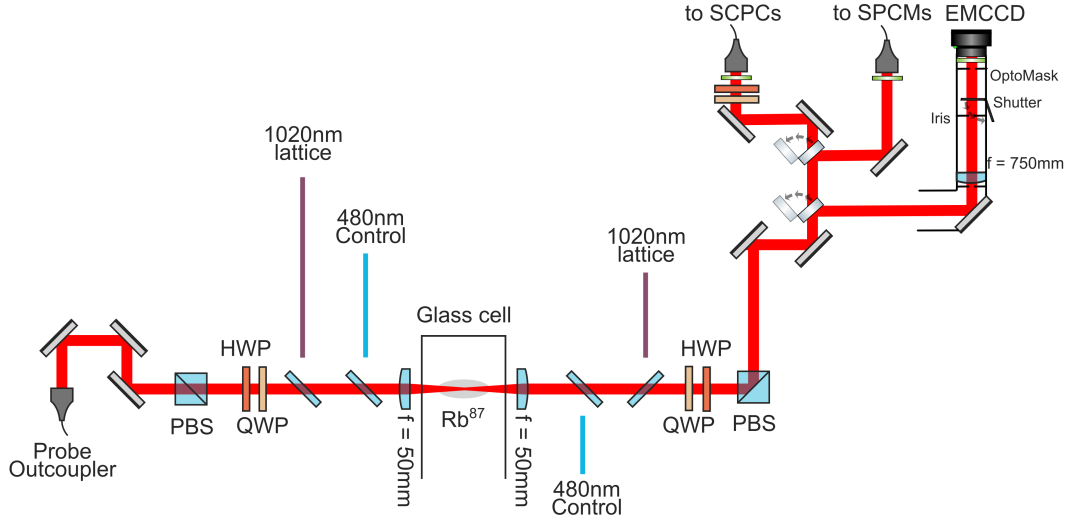


Figure 4.1: Probe beam setup. The probe light is prepared on a separate laser table. PBS and waveplates are used for polarization control while two dichroic mirrors are used to overlap the probe beam with 1 020 nm and 480 nm light. The light is focused onto the atom cloud using a $f = 50$ mm achromat lens. Mirrors in a flip mount are used to control which measurement device is used to detect the probe light.

atomic cloud. Stray light reaching the EMCCD sensor is reduced using an iris, a shutter, and an OptoMask.

To investigate the influence of each optical element on the Gaussian beam quality inside the vacuum chamber, the elements were added sequentially. Starting with only the collimator, the mirrors and lenses were added first, followed by the polarization optics and dichroic mirrors.

4.2 Beam Collimation

In the experiment, fixed triplet collimators are used because they produce an output beam that closely resembles an ideal Gaussian beam. A well-collimated beam is important to minimize changes in the beam radius during propagation and to provide well-defined input conditions for the following focusing lens. However, the lens positions inside these collimators are fixed and cannot be adjusted to compensate for variations between individual fibers and collimators. In the experiment, a finite beam divergence was already observed by measuring the beam size at different distances from the outcoupler. Additional camera and shear-interferometer measurements of several outcouplers of the same type revealed unit-to-unit variations ranging from converging to diverging output beams, as well as smaller variations between repeated insertions of the fiber into the same collimator. To retain the near-Gaussian beam profile provided by the triplet collimator while allowing the beam divergence to be adjusted, the distance between the fiber and the triplet lens system was made adjustable. The implementation and characterization of this adjustable collimator are described in the following.

The procedure for modifying the outcoupler is shown in Figure 4.2. First, all elements are removed from the outcoupler, as shown in Figure 4.2(a). A schematic of the optics inside the coupler is shown in

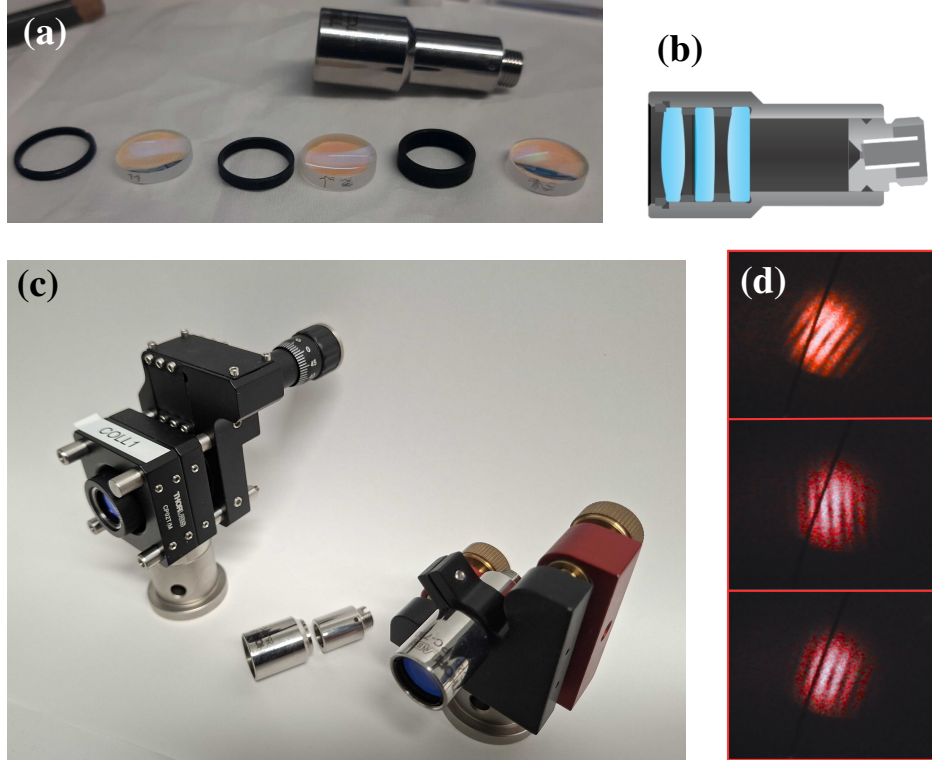


Figure 4.2: (a) Lenses and retainers taken out of the housing. (b) Schematic of the inside of the outcoupler. Taken from [36]. (c) Rebuilding of the collimator. Right: Original collimator. Center: Cut piece. Left: Rebuilt collimator with adjustable lens-fiber distance. (d) Shear interferometer interference patterns for different lens-fiber distances. The black line indicates the reference line of the shear interferometer for beam collimation. Top: Defocused. Center: Focused. Bottom: Collimated.

Figure 4.2(b). The outcoupler consists of two cylindrical sections with different diameters, with all three lenses located inside the section with the larger diameter. The innermost lens is fixed by being pressed against the transition to the narrower section of the housing. The spacing between the lenses is defined by spacers, while the outermost lens is held in place by a retaining ring. These six elements are shown in Figure 4.2(a) in the same left-to-right order as in the schematic in Figure 4.2(b).

The next step is to cut the collimator housing and clean it afterwards. The cut is made at the narrower section of the housing, as shown in Figure 4.2(c), which shows the original collimator, the collimator after cutting, and the final modified collimator. For the final configuration, all optical elements are reinserted into the remaining section containing the larger cylindrical housing in the same order and orientation as before. The narrower section is no longer required. The fiber is then mounted using a fiber-adaptor plate, and the distance between the fiber and the triplet lens system can be adjusted with a z-stage. Finally, the output beam is observed with a shear interferometer, as shown in Figure 4.2(d). By adjusting the distance between the fiber and the triplet lens system, the output beam can be collimated.

4.3 Optical Aberrations

After optimizing the outcoupler, different sources of optical aberrations affecting the probe focus, where the probe light interacts with the atomic cloud, are investigated. As a first step, a simplified setup containing only two mirrors and a long-focal-length lens was built. The long-focal-length lens was then replaced by the probe lens, followed by the successive addition of the polarization optics and dichroic mirrors to determine the influence of each optical element on the probe-beam focus.

4.3.1 Mirror holders

To investigate mirror-induced aberrations, two different mirror-mount designs were compared in a reduced test setup consisting only of the self-built outcoupler, two mirrors⁴, and a lens with a focal length of $f = 500$ mm. The standard mirror holder, which is also used in the current experimental setup, is shown in Figure 4.3(a). In this design, the mirror is fixed by a single screw that presses directly against its surface. This mounting method can introduce mechanical stress and deform the mirror, potentially leading to optical aberrations. An alternative holder design which can potentially be used in future designs of the experiment is shown in Figure 4.3(b). Here, the mirror is fixed by three clamps at the surface, providing a mounting method that is expected to reduce bending of the mirror. The aim of this section is therefore to determine whether an alternative mount design, in which the mirror is fixed more uniformly, can reduce these aberrations. In the following, the standard mirror holder is investigated first.

To test this, a camera⁵ and a translation stage were used to image the beam at different planes orthogonal to the direction of propagation around the focus to extract the beam radius. To determine the beam radius, a two-dimensional fit with a rotational degree of freedom was used

$$I(x, y) = I_0 + A \exp \left[- \left(\frac{x'^2}{2\sigma_x^2} + \frac{y'^2}{2\sigma_y^2} \right) \right], \quad (4.1)$$

$$x' = (x - x_0) \cos \Theta + (y - y_0) \sin \Theta, \quad (4.2)$$

$$y' = -(x - x_0) \sin \Theta + (y - y_0) \cos \Theta. \quad (4.3)$$

with the constant background intensity I_0 , the Gaussian amplitude A , the beam center (x_0, y_0) , the Gaussian widths along the principal axes σ_x and σ_y , and the rotation angle of the principal axes with respect to the laboratory coordinate system Θ . The rotational degree of freedom accounts for the possibility that the principal axes of the beam (x', y') do not coincide with the camera axes (x, y) . The angle Θ was fitted for one data point of high ellipticity and then fixed for the fits of all the other data points. The $1/e^2$ beam radii correspond to

$$r_1 = 2\sigma_x, \quad r_2 = 2\sigma_y. \quad (4.4)$$

⁴ BB1-E03

⁵ Coherent LaserCam-HR II 1/2-inch

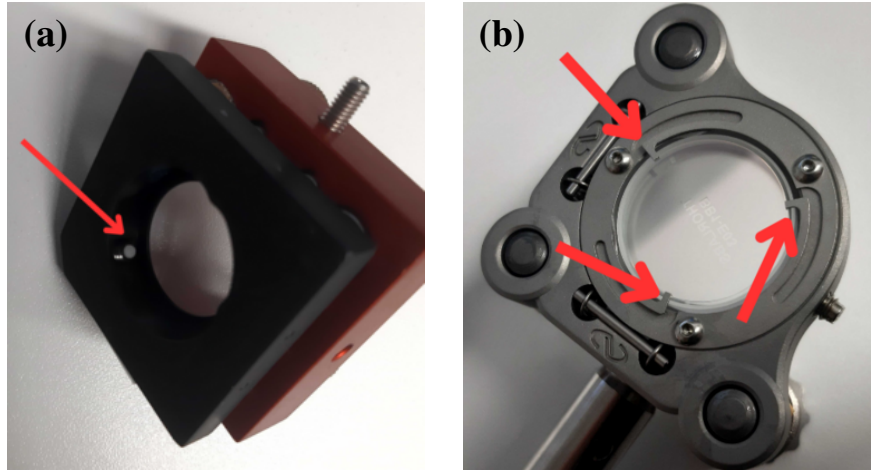


Figure 4.3: (a) Radiant Dyes mirror holders that are used in the experiment. The mirror is fixed by a single screw pressing against the mirror (marked with a red arrow). (b) Newport mirror holder which fixes the mirror with three clamps at the surface (marked with red arrows).

The mirror holder used in the main experiment was tested at two different torques to determine how strongly the applied mounting force affects the beam quality. The first torque is $16 \text{ oz} \cdot \text{in}$. The measured beam profiles as well as the extracted beam radii around the focus are shown in Figure 4.4(a) and 4.4(b). The fits show a small but measurable waist difference between the principal axes and a small astigmatism that may result from imperfect lens alignment. The individual images show a Gaussian profile without visible aberrations. For a higher torque of $32 \text{ oz} \cdot \text{in}$, the results with the same measurement procedure are shown in Figure 4.4(c) and 4.4(d). Here, a clear increase in astigmatism is observed, as shown by both the fit results and the individual images. Especially comparing the images at translation stage position $z = 0 \text{ mm}$ and $z = 13 \text{ mm}$ between Figure 4.4(b) and 4.4(d) shows the significant aberrations caused by excessive tightening torque. Therefore, it has to be checked that the mirrors are not fixed too tightly. A torque of $16 \text{ oz} \cdot \text{in}$ is enough to fix the mirror while simultaneously not introducing significant beam aberrations.

Finally, the alternative mirror holder introduced above was tested. Measurement results for this type of holder are shown in Figure 4.4(e) and 4.4(f). The clamped mirror holder produced results similar to those obtained with the single-screw holder at low torque. The remaining differences may originate from small run-to-run variations in lens alignment.

In summary, single-screw mirror holders can be used without substantially affecting the beam quality, provided that the retaining torque is sufficiently limited to avoid significant mirror deformation. A torque of $16 \text{ oz} \cdot \text{in}$ proved to be a reasonable choice in this test. Alternatively, comparable results were obtained using a mirror holder with multiple clamps.

4.3.2 Probe lens

After investigating the mirror mounts, the long-focal-length lens was replaced by the probe lens used in the experiment. Due to its shorter focal length, small misalignments can have a stronger influence on the focused beam. To minimize such alignment-induced effects, the probe lens was carefully aligned such

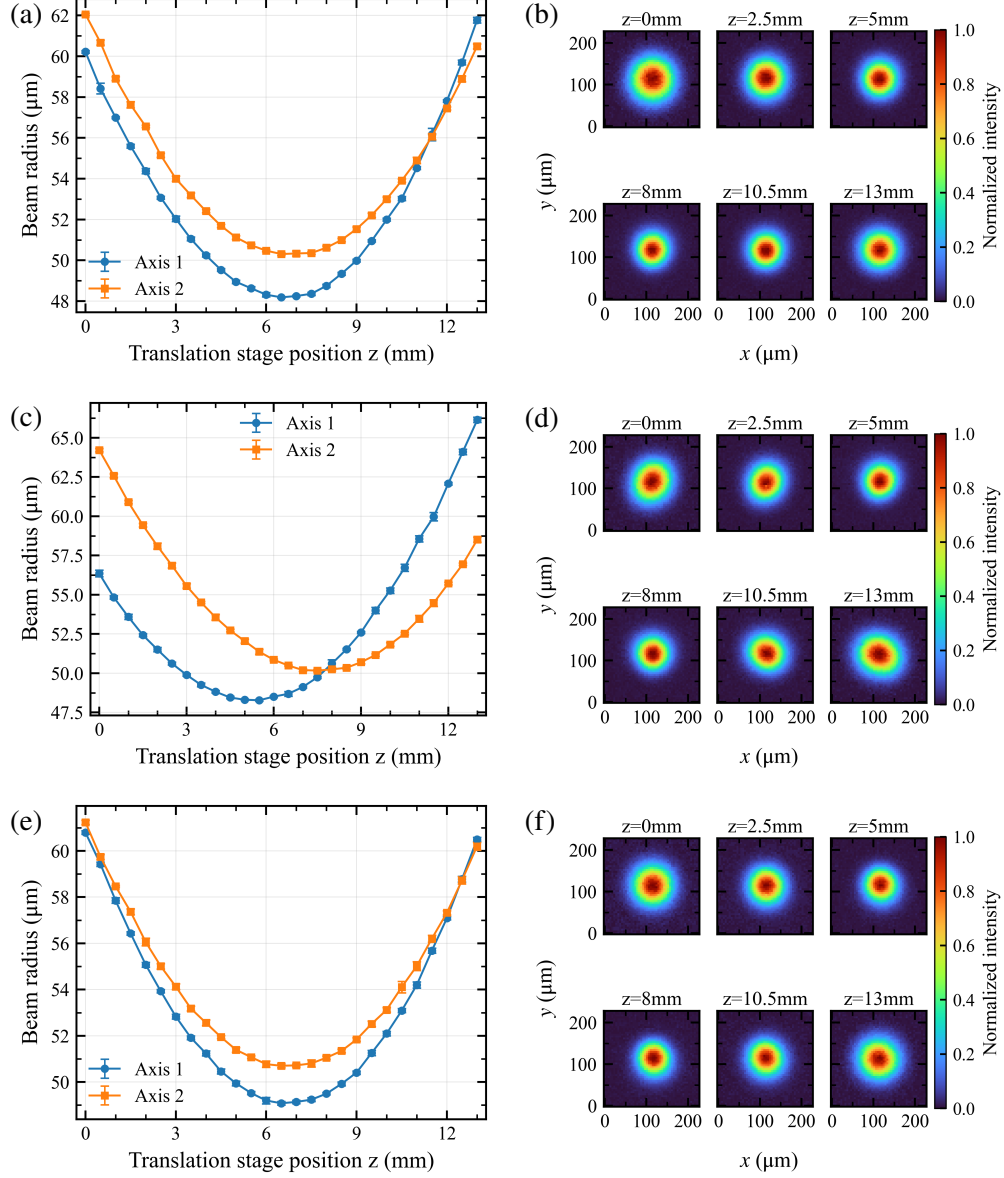


Figure 4.4: Focused-beam characterization for different mirror mounts. The left column shows the beam radii extracted from elliptical 2D Gaussian fits as a function of the camera position z along the translation stage, with an arbitrary origin at $z = 0$. Axis 1 and Axis 2 denote the two principal axes of the fitted profiles. Their assignment is consistent within each measurement series but not necessarily between different series. The right column shows the corresponding normalized transverse intensity profiles at selected positions, with x and y denoting transverse camera coordinates. (a,b) Mirror secured by a single screw tightened to a torque of 16 oz · in. (c,d) The same mounting configuration at 32 oz · in. (e,f) Clamped mirror mount.

that the beam passes through its center at normal incidence.

First, a long ruler was used as a reference to minimize the tilt of the lens relative to the beam propagation direction. Next, a camera was placed at a distance of $2f$ from the lens. The lens position was adjusted until the beam position on the camera was the same with and without the lens, ensuring that the beam passes through the center of the lens. A detailed description of this alignment procedure is provided in [37].

Again, a translation stage and a camera were used to analyse the beam in the focus region. A different camera with a smaller pixel size had to be used to resolve the beam. For this purpose, a camera with a pixel size of $1.4\text{ }\mu\text{m} \times 1.4\text{ }\mu\text{m}$ was used⁶. Interference fringes on the camera image, caused by the coherent nature of the laser light, were avoided by operating the laser below its lasing threshold. The comparison of the images below and above the laser threshold is shown in Figure 4.5 for the collimated beam.

The measurement procedure as well as the data analysis are analogous to Subsection 4.3.1. The step size was reduced to $10\text{ }\mu\text{m}$, which is the lowest step size possible with the used translation stage. The fit results are shown in Figure 4.6(a). It can be seen that using the alignment method described above, the astigmatism was reduced to a very low level. Furthermore, the fitted data clearly show for both axes that the beam widths do not evolve symmetrically before and after the focus. This can also be observed with the individual images in Figure 4.6(b). Close to the focal plane the beam converges more rapidly before the focus than it diverges after the focus. This behaviour was also analysed with larger step sizes and is only visible close to the focus. A possible explanation for the different behaviour before and after the focus is spherical aberration. No effect of spherical aberration on the transverse Gaussian profile could be resolved, likely due to the limited resolution of the camera. To test whether the observed beam distortions originate from spherical aberration, a second probe lens was used to recollimate the beam, which was then measured with a shear interferometer. The result is shown in Figure 4.6(c). It can be seen that indeed it shows an S-shaped fringe pattern which is characteristic of spherical aberrations [38, 39].

The measurements show that a strongly focused beam with only small astigmatism can be generated using the current probe lenses, although evidence of spherical aberration remains. Since aspheric lenses are specifically designed to reduce spherical aberration, an aspheric lens was tested as a possible alternative to the current probe lens. Minimizing such aberrations is particularly important for the investigation of transverse photon-photon interactions, as a well-defined Gaussian beam profile simplifies the description and simulation of the transverse beam propagation. The measurement result for the aspheric lens is shown in Figure 4.7. At the focus, the beam appears to have an approximately Gaussian intensity profile, although this is difficult to verify due to the limited camera resolution. Outside the focal plane, however, significant beam distortions are visible. Similar structures were reproduced with a second lens of the same type, indicating that they are not specific to a single lens. The aspheric lens was therefore considered unsuitable for the experiment. Since the current probe lens is achromatic, replacing it with the aspheric lens could additionally introduce chromatic aberrations. Therefore, it was decided to continue using the current probe lens. Based on these findings, a beam profiler that maps the intensity profile using a pinhole was designed to provide higher spatial resolution for future investigations of strongly focused beams. The beam profiler is described in detail in [40].

⁶ IDS U3-36P0XCP-M-NO Rev.1.2

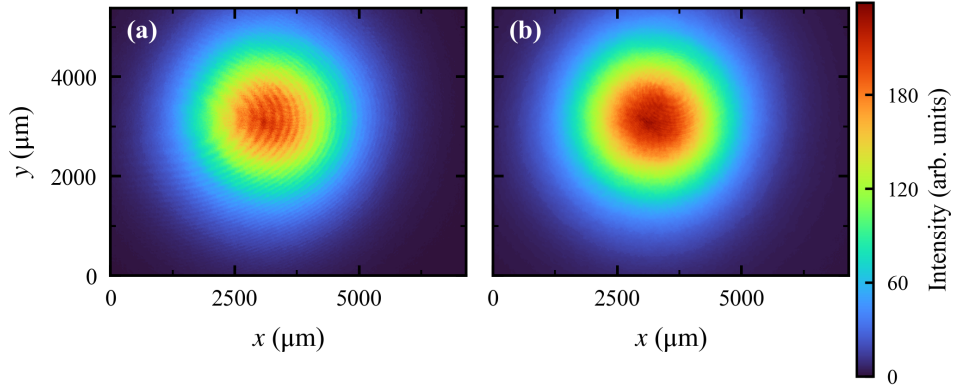


Figure 4.5: Camera image of collimated beam with IDS camera. (a) Above lasing threshold. The coherent light source creates additional interference patterns. (b) Below lasing threshold. Operating the laser diode below threshold removes the artificial interference pattern.

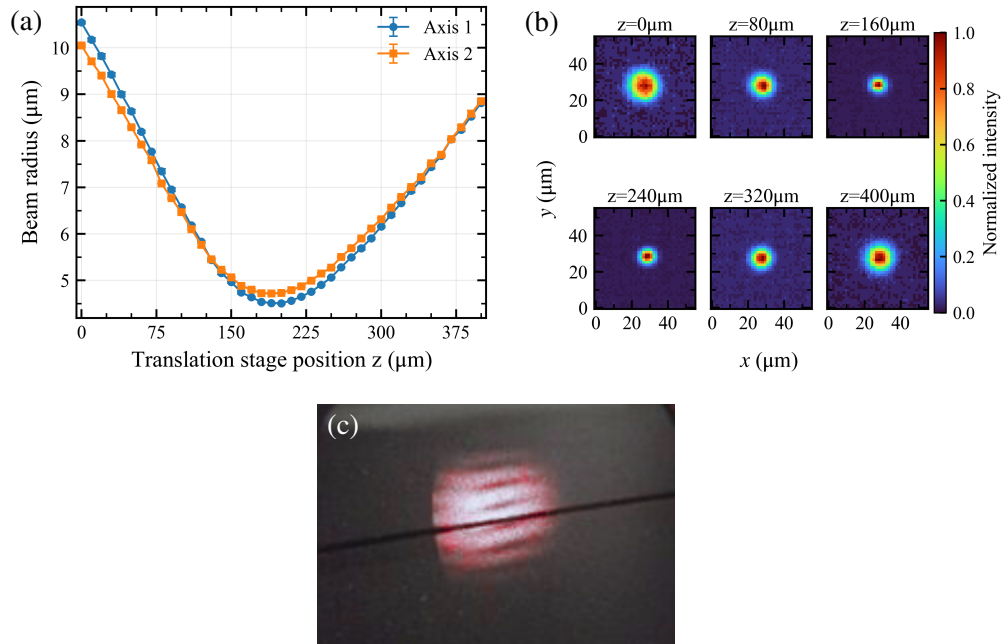


Figure 4.6: Probe lens characterization. (a) Radii from two-dimensional Gaussian fit along both principal axes. (b) Individual images close to the focal plane. (c) Shear interferometer result after focusing and recollimating the beam with two probe lenses. The S-shape is a characteristic of spherical aberration.

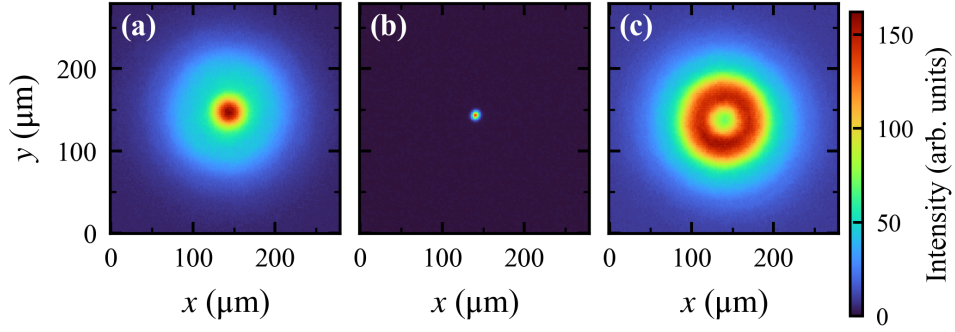


Figure 4.7: Aspheric lens around focus. (a) 2 mm before the focal plane. (b) focal plane (c) 2 mm after the focal plane.

4.3.3 Polarization control and filtering

After investigating the influence of the mirror holders and the focusing lens, the next step is to analyze how the polarization optics and the dichroic filters affect the beam quality inside the atomic cloud. For this comparison, the ellipticity ϵ was evaluated for each measured plane

$$\epsilon = \frac{\max(r_1, r_2)}{\min(r_1, r_2)}. \quad (4.5)$$

Representing the beam shape by its ellipticity allows different datasets to be compared more easily, as the beam dimensions along both principal axes are combined into a single quantity. Despite being a single quantity, the ellipticity and its evolution along the propagation direction provide information about both astigmatism and differences in the beam waists of the two principal axes. Astigmatism results in different focal positions for the two principal axes and therefore produces a characteristic variation of the ellipticity around the focus. Figure 4.8(a) illustrates this behaviour for different separations between the x and y foci, showing how increasing astigmatism changes the ellipticity curve. In addition, an elliptical input beam can result in different beam waists along the two principal axes even without a separation of their focal positions. The resulting influence on the ellipticity curve is shown in Figure 4.8(b). These simulations provide a basis for interpreting changes in the measured ellipticity after adding the polarization optics and dichroic filters.

First, the ellipticity curve of the data shown in Figure 4.6 in the previous subsection is analysed. It is shown by the orange curve in Figure 4.8(c). Here it can be seen that in contrast to the simulation, the curve is not symmetric due to the spherical aberration of the probe lens. Adding a glass plate representing the glass of the vacuum chamber shows no measurable beam distortions apart from a focus shift of approximately 1.3 mm. The black curve was shifted 1.3 mm in the plot to overlap it with the orange curve and simplify comparison.

The effects of the polarization optics and the dichroic mirrors are shown in Figure 4.8(d). It can be seen that the astigmatism is increased by the polarization optics (WPs and PBS). Additionally, an increase in ellipticity can also be observed after adding the waveplates since the curve becomes less

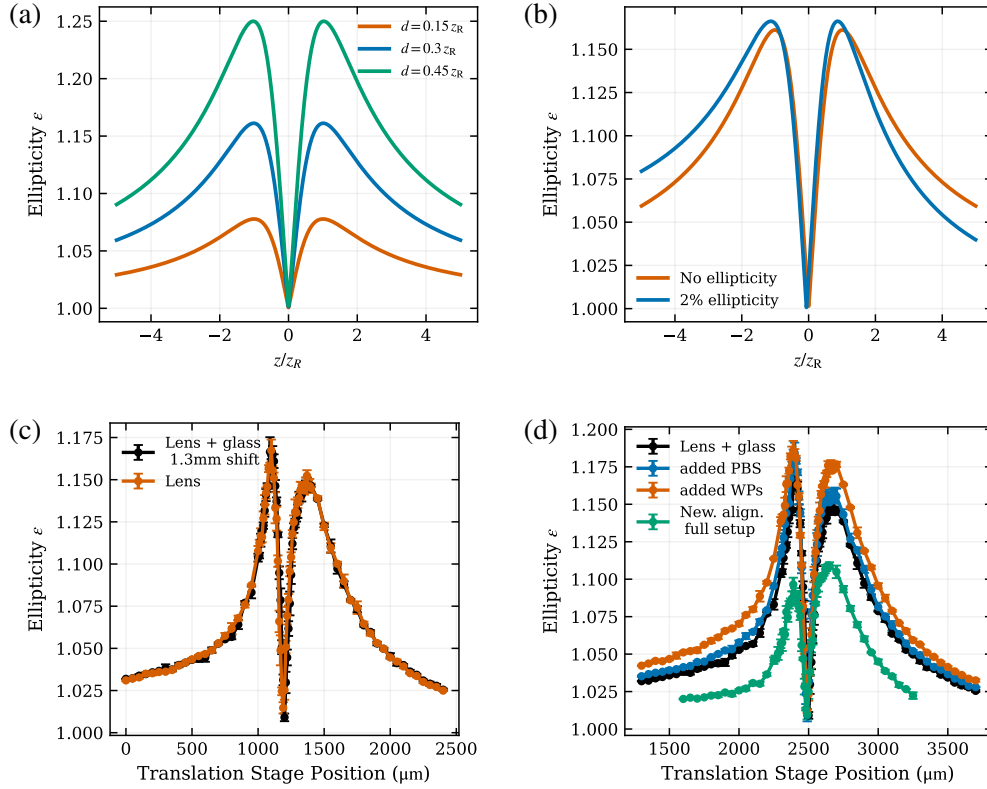


Figure 4.8: Gaussian beam simulation and experimental measurement results. (a) Simulation of the ellipticity along the beam propagation for different separations d between the foci of the main axes. (b) Simulation of the ellipticity along the beam propagation for a circular and an elliptical collimated input beam. (c) Measurements with and without the glass plate, showing a focus shift of 1.3 mm. The black curve is identical to that in (d) and is shifted by 1.3 mm for easier comparison with the measurement without the glass plate. (d) Measured ellipticity curves for different optical elements.

symmetric away from the focal region. While a small effect of the polarization optics on the beam quality is visible here, the effect of the dichroic mirrors is more difficult to isolate. The reason is that just adding the dichroic mirrors in the beam path leads to a significant increase in astigmatism. But this strongly increased astigmatism is caused by the dichroic filters shifting the beam and thereby degrading the lens alignment. It is therefore not possible to measure the direct influence of these optical elements without the uncertainty of the lens alignment.

The green curve in Figure 4.8(d) shows the best lens alignment that was achieved after multiple attempts using the alignment method described in Subsection 4.3.2. From that it can be concluded that the spread due to lens alignment quality is significantly larger than the influence of polarization optics and dichroic mirrors and the results do not justify replacing individual components to improve beam quality.

4.4 Probe Signal Incoupling Efficiency

The adjustable outcouplers that were discussed in Section 4.2 can also be used to collect the photons after the atom cloud to guide the light to the SPCMs. Since the measurements are performed at the few-photon level, a high coupling efficiency is essential to maximize the photon detection rate. This is particularly important for photon-correlation measurements, where the detection of both photons is required for a coincidence event and the coincidence rate therefore depends strongly on the overall detection efficiency. Here a two-lens, self-built collimator⁷ is currently in place and optimized coupling results in a coupling efficiency of approximately 80%.

Using the adjustable triplet lens system for coupling resulted in a coupling efficiency above 88%, which exceeds the specified back-coupling efficiency for the initial nonadjustable coupler [36]. Without polarization optics and dichroic mirrors, a coupling efficiency of more than 90% was achievable, confirming the higher back-coupling efficiency achieved with the adjustable coupler.

For a direct quantitative comparison with the fiber-coupler solution used in the experiment, both coupling measurements must be performed under identical conditions since the beam quality in the test setup is significantly better. However, this test shows that with optimized lens alignment, a probe photon collection efficiency close to 90% is achievable. Relative to an efficiency of 80%, this reduces the fiber-coupling loss from about 20% to about 10% and is a significant improvement of the experimental setup.

It can be concluded that the same optical elements can achieve improved beam quality when alignment and component stress are controlled. It was shown that excessive tightening of the mirror-retaining screw can introduce beam aberrations. Furthermore, accurate probe-lens alignment is crucial and should be performed as carefully and precisely as possible. The probe-lens alignment is the dominant practical factor affecting beam quality. The polarization control and dichroic filtering elements noticeably decrease the probe-beam quality. However, this effect is small compared to the aberrations caused by imperfect lens alignment and the lens itself and can therefore be neglected. The probe photon collection efficiency can be increased to almost 90% through beam-quality optimization and the use of an adjustable triplet fiber coupler.

As shown in Figure 4.1, the optical elements are used not only for the probe-beam but also for several other laser systems as well as the optical pumping beam which is not included in this figure. Realigning the lens system would require significant interventions into other parts of the experiment as well and was due to time constraints not done in the course of this thesis. However, the results should be especially considered for the design and alignment of the next generation of the experiment.

⁷ LA1509-B, $f = 100$ mm and A280TM-B, $f = 18.4$ mm

Precise Atomic Cloud Positioning and Control Beam Alignment

In a recent experimental upgrade, two mirror holders of the optical dipole trap were replaced by piezo-actuated mirror mounts. This upgrade enables more convenient and precise alignment of the optical dipole trap relative to the probe beam. The setup is shown in Figure 5.1. Instead of continuously controlling the mirror position with a piezo voltage, slip-stick actuators are used, for which the position is mechanically maintained after adjustment and is therefore less sensitive to voltage fluctuations and drift. An external controller generates a sawtooth signal. The slow expansion of the piezo rotates the mirror-holder screw, whereas the following rapid contraction leaves the screw at its new position. Further details on the operating principle of the mirror mounts and their use for three-dimensional positioning of the atomic cloud are provided in [41].

Before measuring Rydberg-polariton interactions, the two-level dynamics and the effects of atomic-cloud and control-beam misalignment must be understood because misalignment strongly changes

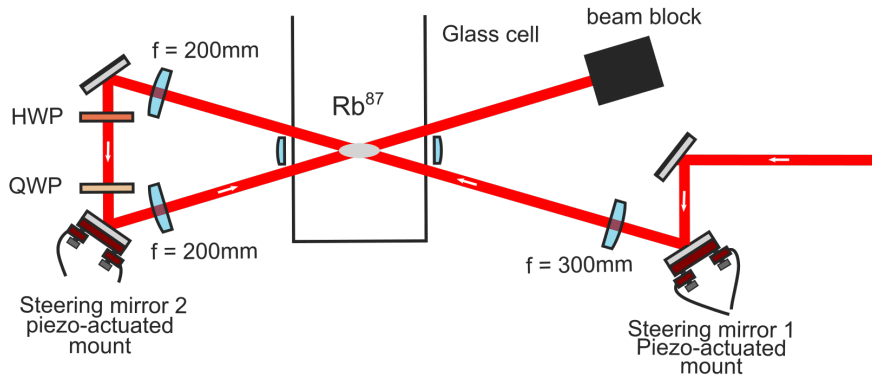


Figure 5.1: Crossed optical dipole trap setup with two piezo-actuated mirror mounts. The two arms have an angle of approximately 31° and form an elongated cloud shape. The waveplates are used to create an orthogonal polarization between the two arms to avoid lattice formation.

the profile measured with the EMCCD. Therefore, the propagation of the probe beam through an inhomogeneous atomic cloud created by the crossed dipole trap is simulated and shown in Section 5.1. This is followed by an investigation of the influence of the atomic cloud position and its precise alignment in Sections 5.2 and 5.3. The measurement results for the beam propagation close to two-level resonance are shown in Section 5.4. It furthermore investigates the OD estimation with the EMCCD camera. Finally, the control beam is added and its alignment procedure is discussed in Section 5.5.

5.1 Light Propagation through a Two-Level System

To understand the two-level system dynamics and to interpret measurement results with the two-level system, it is important to first understand how the atom cloud position affects the transverse beam profile. The propagation of the probe beam is complicated by several experimental factors, including probe-beam aberrations such as ellipticity and astigmatism, as well as cloud displacement and inhomogeneities in the atomic density distribution. The propagation of an inhomogeneous probe beam through an inhomogeneous atomic cloud near the two-level resonance is nontrivial and must be calculated numerically. For this purpose, the angular spectrum method is used which is an iterative technique for modelling light propagation through any medium described by a complex refractive index.

The derivation is based on [42]. A propagation step is calculated using

$$\psi_e(x, y + \Delta y, z) = \exp\left(\frac{ik_0(n^2 - 1)}{2}\Delta y\right) \mathcal{F}^{-1} \left\{ \exp\left(\frac{-i(k_x^2 + k_z^2)}{2k_0}\Delta y\right) \mathcal{F} \{ \psi_e(x, y, z) \} \right\} \quad (5.1)$$

where y is the propagation coordinate, ψ_e the complex electric field envelope and $\mathcal{F}\{\}$ the two-dimensional Fourier transformation. Since a detailed explanation of the angular spectrum method, the implementation into the simulation and validations are given in [43], this section focuses on the simulation results that are directly relevant for the measurements performed in this chapter. For the simulation, a probe astigmatism of $40\text{ }\mu\text{m}$ and probe waists of $\omega_x = 5.9\text{ }\mu\text{m}$ and $\omega_z = 4.8\text{ }\mu\text{m}$ were assumed. These values were measured experimentally by the telescope combined with the EMCCD for the free-space case without atom loading. These values may be subject to systematic deviations caused by aberrations of the telescope lenses. The cloud size was determined from the absorption imaging with the parameters $\sigma_x = 14\text{ }\mu\text{m}$, $\sigma_y = 25\text{ }\mu\text{m}$ and $\sigma_z = 11\text{ }\mu\text{m}$.

A simulation result for different probe detunings is shown in Figure 5.2(a). It can be seen that near resonance, the simulated output beam narrows at negative detuning and broadens at positive detuning because of the detuning-dependent refractive index, which was discussed in Section 2.1, combined with the inhomogeneous atom density distribution. For higher optical depths, a significant deviation from the Gaussian beam profile can be observed. This is illustrated in Figure 5.2(b). Here a ring-like structure can be observed close to resonance for positive probe detunings. This structure is not spherically symmetric due to the astigmatism, beam ellipticity and cloud ellipticity. For negative detunings, a stretching of the focused beam along one direction can be observed.

These simulation results illustrate substantial effects on the intensity distribution.

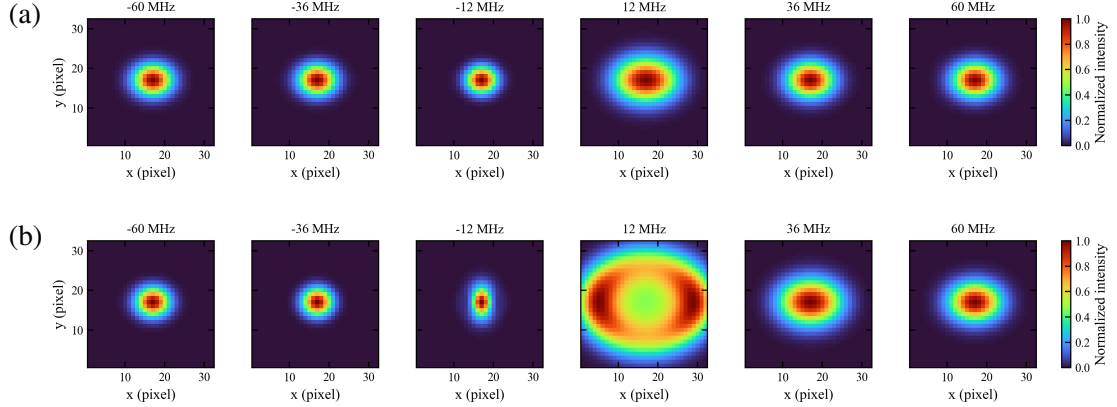


Figure 5.2: Simulation of probe beam propagation close to two-level resonance for different probe detunings $\Delta_p/(2\pi)$. The intensity is normalized to each individual image. (a) For an optical depth of 25. (b) For an optical depth of 65.

5.2 Longitudinal Cloud Positioning

The piezo-actuated mirror mounts provide an upgrade for precise cloud positioning. To determine the optimal alignment of the atomic cloud relative to the probe focus, the optical depth (OD) is simulated for different longitudinal cloud displacements. For large optical depths, the OD cannot be reliably determined directly on resonance in the experiment due to the large absorption. Therefore, the probe detuning is scanned and the OD is obtained from a Lorentzian fit to the measured transmission. This procedure is further described in Section 5.3. To be consistent with the experimental procedure, the OD is extracted the same way in the simulation. As measured in the experiment, the simulation takes 41 equally distributed data points from $\Delta_p/(2\pi) = -120$ MHz to 120 MHz into account.

It was shown in the previous section that the diffraction due to the atom cloud strongly influences the transverse shape of the outgoing beam. Since the photons are collected with a single-mode fiber as it was shown in Figure 3.5, this leads to detuning-dependent fiber coupling changes and therefore to an overestimation of the optical depth. Therefore, the transmission of the simulated beam is calculated in two ways. The first is just the pure absorption which is

$$T_{\text{absorption}} = \frac{\sum_i |E_{\text{atoms}, i}|^2}{\sum_i |E_{\text{free space}, i}|^2} \quad (5.2)$$

with the complex electric field for the probe propagating through the atom cloud $E_{\text{atoms}, i}$ and the complex electric field for the probe beam propagating in free space $E_{\text{free space}, i}$ at each pixel i . The second method takes the diffraction effects into account as well by calculating the mode overlap with the free-space beam

$$T_{\text{overlap}} = \frac{|\sum_i E_{\text{atoms}, i} E_{\text{free space}, i}^*|^2}{\sum_i |E_{\text{free space}, i}|^4}. \quad (5.3)$$

This is not the real transmission of the probe but it illustrates what is really measured with the SPCMs due to the photon collection via a single-mode fiber. However, this simulation is only qualitative. For an accurate simulation of the experimental conditions with the SPCMs, the long propagation of the probe beam including the recollimation by the second probe lens as well as the fiber mode would have to be taken into account.

The simulated OD for both cases for different longitudinal displacements of the atom cloud with respect to the probe beam is shown in Figure 5.3(a). Here, the extracted OD remains approximately constant for cloud positions within about one Rayleigh length of the focus. The experimental data show the OD extracted from the SPCM measurements. The experimental results show a plateau over a displacement range comparable to that predicted by the simulation. Small fluctuations within the plateau may result from slight differences in the quality of the transverse cloud alignment between individual measurements. Furthermore, a strong decrease in OD is observed beyond a certain cloud displacement. The width of the measured curve appears to be slightly larger than theoretically expected. A possible explanation is a small systematic error in the imaging calibration, since the longitudinal cloud position of each data point was determined from the absorption images. The zero point of the experimental longitudinal cloud displacement is set to the point that was chosen experimentally as the atomic cloud position. It was chosen such that it is centered between the two outermost data points where the OD dropped by approximately a factor of two compared to the plateau.

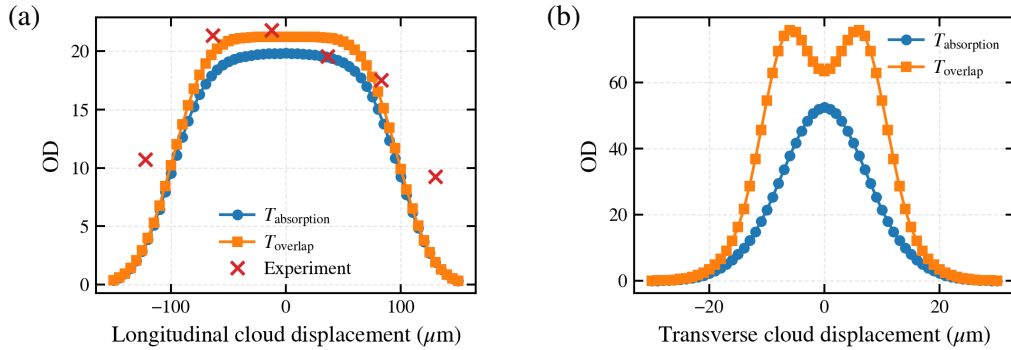


Figure 5.3: Simulated OD results for the probe propagation through a displaced cloud. The results are shown for the pure absorption $T_{\text{absorption}}$ and the mode overlap T_{overlap} as defined in Equation 5.2 and 5.3. (a) Longitudinal cloud displacement including experimental data. (b) Transverse cloud displacement.

While the longitudinal cloud displacement showed no qualitative difference between the behaviour of the absorption alone and the mode overlap, this is not necessarily the case for other configurations. An example is shown in Figure 5.3(b). Here, the same simulation was performed without longitudinal cloud displacement while varying the transverse cloud position for a higher OD exceeding 40. While the

absorption continuously decreases with increasing transverse displacement, there is a small range in which the OD extracted from the mode overlap increases. In this regime, the effect of the diffraction-induced mode change is larger than the decrease in absorption.

5.3 Methods of Transverse Cloud Positioning

While the experimental apparatus is not that sensitive to the longitudinal cloud position, it is extremely sensitive to the transverse cloud position. This section shows how transverse cloud alignment was previously performed using the SPCMs in Subsection 5.3.1. After that, two new approaches to more accurate transverse cloud alignment are presented using the EMCCD camera in Subsection 5.3.2 or classical probe light combined with absorption imaging in Subsection 5.3.3.

5.3.1 SPCM-Assisted Alignment

Before the EMCCD camera was integrated into the experimental setup, the SPCMs were used for cloud alignment. The SPCM-based alignment procedure relies on measurements of the OD. Since the achievable optical depths are sufficiently high that almost no photons are transmitted on resonance, the OD can no longer be determined directly from the resonant absorption. Instead a scan of the probe detuning is performed. The number of photons is measured with and without the atom cloud to determine the transmission at each detuning. The theoretical probe transmission can then be followed from the two-level susceptibility and Equation 2.5 and is given by

$$T(\Delta_p) = \exp\left(-\frac{\text{OD}}{1 + \frac{4\Delta_p^2}{\gamma^2}}\right). \quad (5.4)$$

The Lorentzian fit is used to extract the OD from the probe detuning scan. In the following, this measurement procedure is referred to as OD scan and the result is referred to as OD curve.

In previous years, the dipole trap was aligned relative to the probe beam by minimizing the asymmetry of the measured probe detuning scan [44]. An example of an asymmetric OD curve is shown in Figure 5.4(a). This asymmetry arises from detuning-dependent changes in the fiber-coupling efficiency caused by diffraction of the probe beam as it propagates through the atomic cloud. When the cloud is transversely misaligned relative to the probe beam, the fiber-coupling efficiency differs significantly between the two sides of the OD curve. After realigning the cloud position, the OD curve looks symmetric as shown in Figure 5.4(b). It can also be seen that the OD obtained from the fit is higher for a misaligned cloud. This is qualitatively in agreement with simulation results shown in Figure 5.3(b). It shows that there is a certain regime where the decrease of fiber coupling efficiency is more dominant than the decrease of absorption due to cloud misalignment. In general, the OD scans with the SPCMs suffer from a systematic overestimation of the OD due to reduced fiber coupling efficiency caused by diffraction. This will be further discussed in Section 5.4.

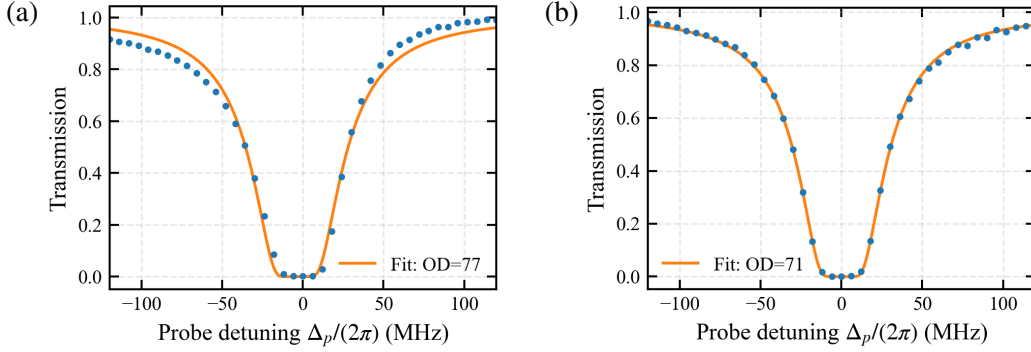


Figure 5.4: Probe detuning scan with Lorentzian fit for OD estimation given in Equation 5.4. (a) The misaligned dipole trap position results in an asymmetric OD curve that can no longer be described accurately by the Lorentzian fit. (b) Same scan after cloud position alignment. The transmission is similar for both signs of the detuning.

The SPCM-based alignment procedure has several limitations. First, this procedure is not sufficiently sensitive at low ODs. Second, a misaligned fiber coupling can create the same effect. As a consequence, a reduced fiber coupling efficiency combined with a moderate cloud misalignment can result in a symmetric OD curve. Furthermore, at ODs exceeding 100 with this measurement and fitting procedure, strong diffraction effects can create fiber coupling efficiency changes for positive and negative detunings, so that a slightly asymmetric OD curve may still be present even at the optimum transverse cloud position.

To overcome these limitations and improve the positioning of the atomic cloud, two alternative alignment methods are presented in the following.

5.3.2 Alignment with EMCCD Camera

The EMCCD camera provides a new tool for precise transverse cloud alignment as diffraction effects can be directly imaged. It was shown in Section 5.1 that a focusing of the beam close to resonance for negative detunings and a defocusing of the beam for positive detunings is expected. The same observation was made experimentally. Corresponding measurement results for different detunings are shown in Section 5.4. For small cloud displacements, this mainly leads to a small shift of the probe beam position close to resonance in comparison to the reference measurement without atoms. The shift occurs in opposite directions for positive and negative probe detunings. This effect was confirmed both with the simulation and experimentally. Furthermore, significant beam distortions were observed for larger cloud displacements on the order of $5\ \mu\text{m}$. These distortions were primarily observed for positive detunings, while they were less apparent for negative detunings due to the combined effects of focusing and the limited spatial resolution. An example is given in Figure 5.5. It shows the beam distortions for the simulated beam propagation as well as in the experiment for a transversely displaced cloud and a probe detuning of $\Delta_p/(2\pi) = 12\ \text{MHz}$. In both cases, substantial deviations from the initial Gaussian beam shape as well as a beam position change in comparison to the reference beam can be observed. These images were created by averaging the counts in each pixel over all images acquired during multiple experimental cycles.

Based on these observations, the following alignment procedure was developed. A detuning close to resonance is chosen and possible beam-shape distortions and position changes are observed. Then

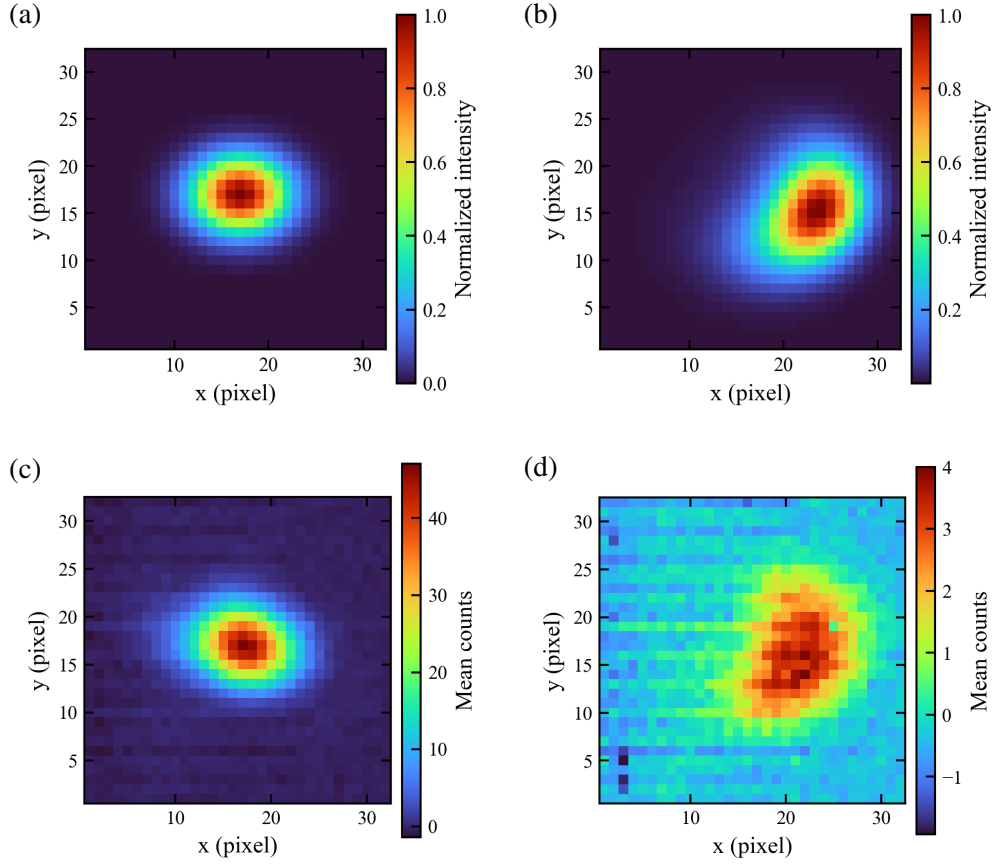


Figure 5.5: Simulation and experimental results for transverse cloud alignment with EMCCD. (a) Simulation of the reference beam (b) Simulation with $\Delta_p/(2\pi) = 12$ MHz with $5\ \mu\text{m}$ horizontal displacement and $1\ \mu\text{m}$ vertical displacement. (c) Experimentally measured reference beam. (d) Experimentally measured beam distortion for imperfect cloud alignment at $\Delta_p/(2\pi) = 12$ MHz.

the dipole trap is moved and a new image is taken. Averaging over 10 to 20 experimental cycles, corresponding to less than one minute of acquisition time, is usually sufficient to achieve good image quality. Once the diffracted beam recovers a Gaussian profile and approaches the position of the free-space beam, two-dimensional Gaussian fits can be used to determine the beam centers in both images and further improve the alignment. The achievable alignment precision is ultimately limited by the finite step size of the piezo-actuated mirror mounts.

5.3.3 Alignment via Cloud Splitting

A third option for transverse cloud positioning with respect to the probe beam is cloud splitting. Here, the neutral density filters, which normally strongly attenuate the probe beam to reach the few-photon level, are removed to use the probe beam as a classical light source. The classical probe beam is shot through the atom cloud at atomic resonance which pushes the atoms away and creates a hole in the center of the atomic cloud. This hole can then be observed using resonant absorption imaging. The absorption imaging effectively images the sum of atoms along the propagation direction of the imaging

beam. The central hole produces an absorption image that looks like a split cloud for both horizontal (x,y) and vertical (z,y) imaging. An example of an equally split cloud using this alignment procedure is shown in Figure 5.6 for the vertical imaging. Misalignment of the cloud results in a splitting of the cloud in unequal parts. By using the absorption images to equalize the cloud splitting, the cloud can be precisely positioned such that the probe beam hits the center of the cloud.

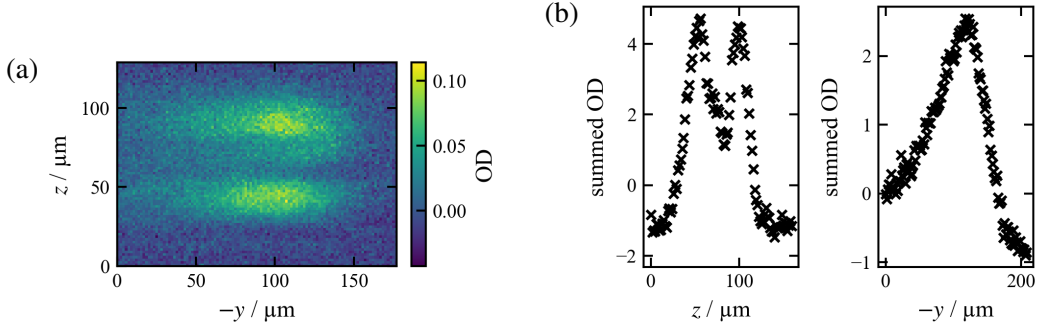


Figure 5.6: Cloud splitting with classical probe light. (a) Absorption image obtained by calculating the optical depth for each pixel according to Equation 3.1. (b) Corresponding OD profiles summed along the horizontal and vertical axes.

While alignment based on cloud splitting provides a simple and fast method at low optical depths, its accuracy becomes limited at higher optical depths. At an OD of approximately 20, this approach works reliably and gives results consistent with the EMCCD-based alignment procedure. However, at larger ODs of around fifty, diffraction effects in the imaging of the atomic cloud influence the observed cloud splitting, resulting in deviations from the alignment obtained with the EMCCD method. Therefore, cloud-splitting-based alignment should primarily be used at lower optical depths, whereas the EMCCD-based approach is preferable for ODs significantly exceeding 20.

5.4 EMCCD-Based Optical-Depth Measurements

Previous measurements of the optical depth in the experiment were performed using the SPCMs. However, as shown in Section 5.1, the interaction of the light with the two-level system leads to significant changes of the beam shape close to the resonance. Therefore, using the SPCMs to estimate the OD systematically overestimates the optical depth as it misinterprets reduced fiber coupling efficiency as absorption. By detecting the probe light directly on the camera rather than only the fraction coupled into a single spatial mode, the EMCCD camera can be used to avoid this systematic overestimation.

5.4.1 Background subtraction

To determine the transmission at a given probe detuning, four quantities are required: the integrated signal counts, the integrated reference counts, and the corresponding signal and reference background counts. The signal and reference measurements are acquired for each detuning, whereas the two background measurements are recorded with the probe shutter closed and are therefore independent of the probe detuning. All four datasets are averaged over many acquired images before the counts

are summed over all pixels. The integrated signal background is then subtracted from the integrated signal counts, while the integrated reference background is subtracted from the integrated reference counts. Finally, the transmission is calculated as the ratio of the two background-corrected integrated counts. Accurate background subtraction is therefore essential for optical-depth measurements with the EMCCD camera and for maintaining a high image quality, since an incorrect background estimate can introduce artificial spatial patterns. Initial dark-frame measurements showed that the background varies between pixels and also changes systematically during the pulse sequence, which appears to originate from internal camera dynamics. Therefore, if only part of the pulse sequence is analyzed, for example to compare the first and last parts of the sequence, the background must be determined only from the corresponding pulse interval of the background images rather than from the full sequence. In addition, the integrated background counts exhibit shot-to-shot fluctuations whose magnitude exceeds the mean number of counts generated by a single detected photon. Consequently, the background measurements must be averaged over many experimental cycles before the integrated counts are calculated.

During the first iterations of the EMCCD-based OD scans, significant deviations from the expected measurement results were observed since the counts on resonance were non-zero, as shown in Figure 5.7(a). It was found that the background taking procedure of averaging the background over multiple cycles and taking the background before or after a measurement was not sufficient. Besides shot-to-shot background fluctuations, long-term drift of the background has to be taken into account. A long-term measurement demonstrating the background drift is shown in Figure 5.7(b). This problem is known and Andor provides a baseline clamp technique intended to stabilize the electronic offset [33]. However, performing measurements at such a low photon level and summing the counts over all pixels requires a background level that is more stable than the baseline clamp technique can provide.

The solution is not to acquire the background only before or after the measurement, but to acquire background frames continuously throughout the measurement. Each loop of the OD scan consists of 41 probe-detuning measurements ranging from -120 MHz to 120 MHz. After the last detuning point, an additional experimental cycle is performed with the probe shutter closed to acquire background frames. The next loop then starts again at -120 MHz. Since the measurement is averaged over multiple loops, this dynamic background-acquisition scheme accounts for long-term drift because the background frames are distributed over the measurement period in the same manner as the data frames.

Using this method, the background-subtracted counts measured on resonance are zero within the statistical uncertainty, which was obtained by error propagation of the mean errors of the background-corrected integrated signal and reference counts. This was measured for optical depths above 20 where the transmission on resonance is negligibly small. To demonstrate the effect on the image quality, the image at 12 MHz probe detuning is shown with background taken before the measurement in Figure 5.7(c) and with dynamic background taking in Figure 5.7(d). It is shown that taking the background separately from the data leads to additional patterns on top of the image due to pixel-dependent background drift. This is especially relevant for cases where the mean photon number per pixel is significantly below one, as is the case at a probe detuning of 12 MHz due to large absorption and defocusing of the beam. The dynamic background taking increases the image quality significantly as it removes artificial patterns caused by background drift variations for different pixels.

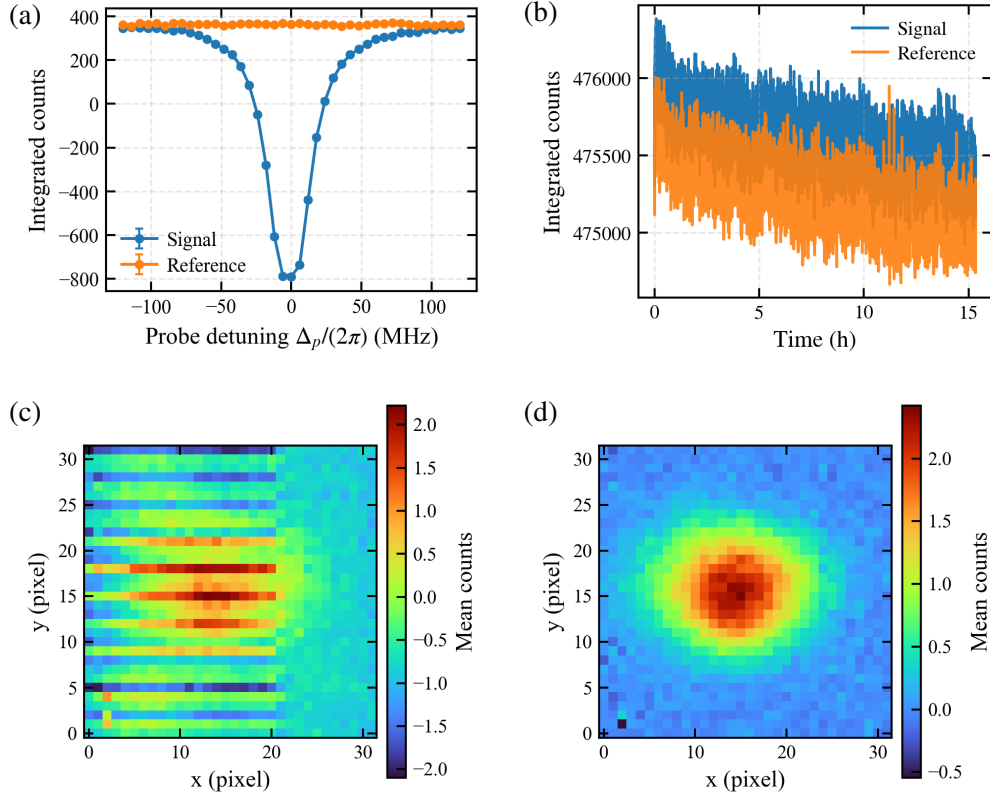


Figure 5.7: Impact of long-term background drift. (a) OD scan with background taken before the measurement. (b) Long-term drift of dark image integrated counts for signal (with atoms) and reference (free space) pulses. (c) Image at 12 MHz probe detuning and $OD \approx 25$ with background taken before the measurement. (d) Same data as in (c) with background taken during the measurement.

5.4.2 Comparing SPCMs and EMCCD

After ensuring correct background subtraction with the EMCCD camera, the OD scans obtained with the two devices can be compared directly. The result is shown in Figure 5.8(a). It can be seen that the OD extracted from the SPCM data, $OD \approx 71$, is significantly higher than the value extracted from the EMCCD data, $OD \approx 49$. This is in agreement with the initial idea that the diffraction near resonance changes the fiber-coupling efficiency and thereby leads to an overestimate of the absorption with the SPCMs. Some selected images at different detunings are shown in Figure 5.8(b). The EMCCD images show focusing at negative detuning and defocusing at positive detuning. This opposite behaviour is in agreement with the two-level susceptibility that is shown in Figure 2.1(a) and the simulation results in Figure 5.2(a).

When comparing the two detection methods, changes in the optical depth during the pulse sequence must also be considered. Atom loss during dipole-trap pulsing reduces the OD, allowing the dependence of the discrepancy between the two methods on OD to be examined within the same measurement. The extracted OD for different pulse windows is shown in the appendix in Figure A.1(a) for the SPCM measurement and in Figure A.1(b) for the measurement with EMCCD. In both cases there is a significant

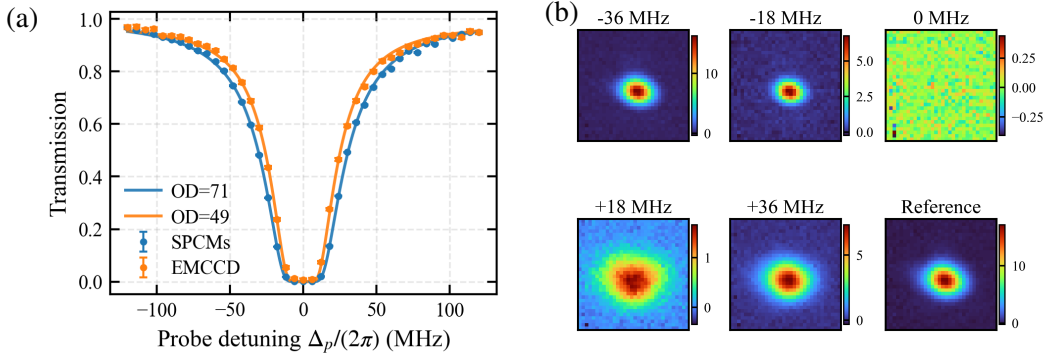


Figure 5.8: (a) Probe-detuning scans for a two-level system with SPCMs (blue) and EMCCD (orange) with Lorentzian fit from Equation 5.4 to extract the OD. It can be seen that the extracted ODs differ significantly. (b) Selected EMCCD images for different detunings close to atomic resonance showing the mean EMCCD counts for each pixel. The reference corresponds to the free-space case in the absence of atoms. The colorbar represents the mean counts per pixel after background subtraction.

loss of OD caused by atom loss during the dipole trap pulsing. It can be seen that for the first fifty pulses, the ratio of the ODs obtained with the two methods is approximately 1.6, whereas it is only approximately 1.28 for the last fifty pulses, for which the OD is lower. This also indicates that, at higher OD, the systematic overestimation of the SPCMs becomes more relevant while for low ODs the SPCM measurement is a reasonable approximation.

To repeat the detector comparison at an approximately constant OD throughout the pulse sequence, the power of the optical dipole trap is increased linearly. This compensates for the reduction in OD caused by atom loss by compressing the remaining cloud. The atom density of the cloud follows a Maxwell-Boltzmann distribution. Using the calculated potential $U(\vec{r})$, the atom density [29]

$$n(\vec{r}) = n_0 e^{-\frac{U(\vec{r}) - U_{\min}}{k_B T}} \quad (5.5)$$

can be calculated with the peak atom density n_0 . The aim is to increase the laser power in order to increase the potential depth $U(\vec{r})$ at the center of the atom cloud since the dipole trap potential scales linearly with the intensity [29]

$$U(\vec{r}) = -\frac{1}{2\epsilon_0 c} \text{Re}[\alpha] I(\vec{r}). \quad (5.6)$$

Here, α denotes the complex polarizability. The increased intensity increases the potential depth and therefore compresses the atomic cloud and increases the density in the probe region, thereby maintaining the density sampled by the probe despite atom loss. The combined effect of the ODT-power ramp and a higher rubidium-dispenser current is shown in Figure 5.9. Figure 5.9(a) again compares the OD measurement with SPCMs and EMCCD. It can be seen that the OD is now even higher and that the ratio between the two approaches increased to approximately 1.78. This also indicates that the systematic

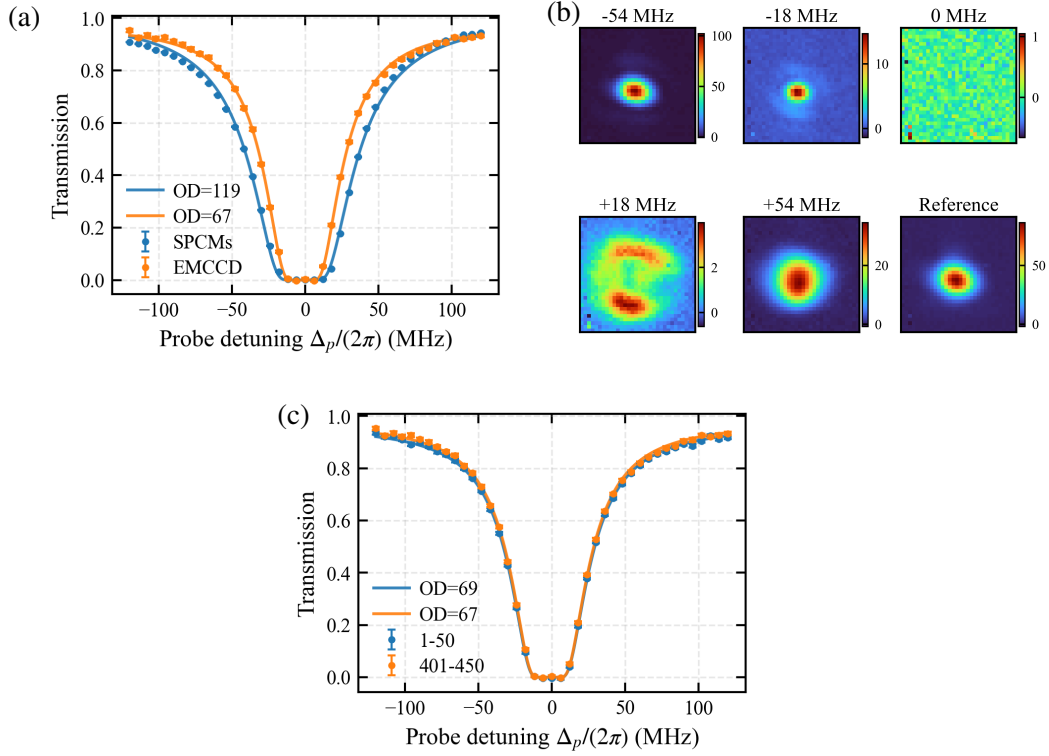


Figure 5.9: Two-level system with linearly increasing ODT power. (a) Probe-detuning scans for a two-level system with SPCMs (blue) and EMCCD (orange) with Lorentzian fit from Equation 5.4 to extract the OD. It can be seen that the extracted ODs differ significantly. (b) Selected EMCCD images for different detunings close to atomic resonance showing the mean EMCCD counts for each pixel. The reference corresponds to the free-space case in the absence of atoms. The colorbar represents the mean counts per pixel after background subtraction. (c) OD fits with EMCCD data for the first and last 50 pulses. The optical depth differs less than 5% between the two pulse windows.

overestimate obtained with the SPCMs increases with OD. The reason for this increased overestimation is shown in Figure 5.9(b). While for low ODs the diffraction effects are limited to a simple focusing and defocusing of the Gaussian profile, more significant beam profile changes appear for larger ODs as is particularly observable in the image recorded at a probe detuning of 18 MHz. This beam shape change is qualitatively in agreement with the simulation result shown in Figure 5.2(b). The simulated output profile depends strongly on the assumed cloud dimensions and input-beam parameters. Due to the limited resolution in the cloud size measurement and the uncertainty of whether aberrations were caused by the focusing probe lens or the telescope, only a qualitative comparison was possible. Furthermore, the simulation assumes no atom loss, cloud size change or temperature change throughout the pulses. Even at ± 54 MHz where the transmission exceeds 80% significant diffraction was observed. Figure 5.9(c) shows the comparison of the first and last 50 pulses of the pulse sequence with the EMCCD. Here the OD only differs by a small percentage due to the ramping of the ODT power.

The linear increase of the ODT power keeps the OD stable but may introduce other significant side effects on the beam profile. It can affect the cloud size and temperature throughout the pulse sequence.

Both were measured after the pulse sequence with and without linear ramping of the dipole trap. The cloud size was measured using absorption imaging and showed a noticeable change of the cloud size. The width was extracted from a two-dimensional Gaussian fit. The temperature was measured by observing the cloud size after different times of flight t . The cloud temperature T can then be extracted from the expansion of the atomic cloud [45]

$$\sigma(t) = \sqrt{\sigma_0^2 + \frac{k_B T}{m_a} t^2} \quad (5.7)$$

where m_a is the mass of a single ^{87}Rb atom, σ_0 is the initial cloud size and k_B is the Boltzmann constant. Using this relation, the temperature was determined to be approximately $9\text{ }\mu\text{K}$ without the ODT-power ramp and approximately $18\text{ }\mu\text{K}$ with the ramp. Thus, the measured cloud temperature approximately doubled. The temperature measurements are shown in Figure A.2 in the appendix. They also show a reduction of σ_y of approximately $3\text{ }\mu\text{m}$.

To check the influence of cloud size and temperature change on the beam profile, the beam profile was compared for different pulse windows. This is shown in Figure A.4 in the appendix. It can be seen that the increase of ODT power does not noticeably change the camera image while there are drastic changes observable without this ramping due to atom loss. In both cases, a small shift of the cloud center is also observed throughout the pulse sequence which may be introduced from thermal effects during the ODT pulsing.

To validate that the SPCMs are systematically overestimating the OD due to additional fiber coupling losses caused by diffraction near resonance rather than the camera underestimating the OD, a third method for OD estimation is tested. This method uses the dependence of the on-resonance EIT pulse delay τ on the optical depth and the control Rabi frequency Ω_C [46]

$$\text{OD} = \frac{\tau \cdot \Omega_C^2}{\gamma}. \quad (5.8)$$

The control Rabi frequency extraction is shown in Figure 5.10(a). A scan under EIT conditions at Rydberg state 54S was performed using the fit function

$$T(\Delta_p) = \exp \left(-\text{OD} \frac{\Gamma \left(\Gamma \gamma^2 + 4\Gamma(\Delta_p - \Delta_c)^2 + \gamma \Omega_c^2 \right)}{4\Gamma^2(\Delta_p - \Delta_c)^2 + \gamma^2 \left(\Gamma^2 + 4\Delta_p^2 \right) + 2\gamma\Gamma\Omega_c^2 + \left(\Omega_c^2 - 4\Delta_p(\Delta_p - \Delta_c) \right)^2} \right) \quad (5.9)$$

which can be followed from Equation 2.5 and the corresponding susceptibility. A control Rabi frequency of $\Omega_C = 2\pi \times (8.82 \pm 0.49)\text{ MHz}$ was extracted. Because the EIT spectrum was recorded at a different rubidium-dispenser-current setting than the measurement shown in Figure 5.9, the corresponding OD measurements were repeated using both the SPCMs and the EMCCD camera. The dispenser current affects the background-gas pressure and thereby the MOT loading rate, which can change the optical

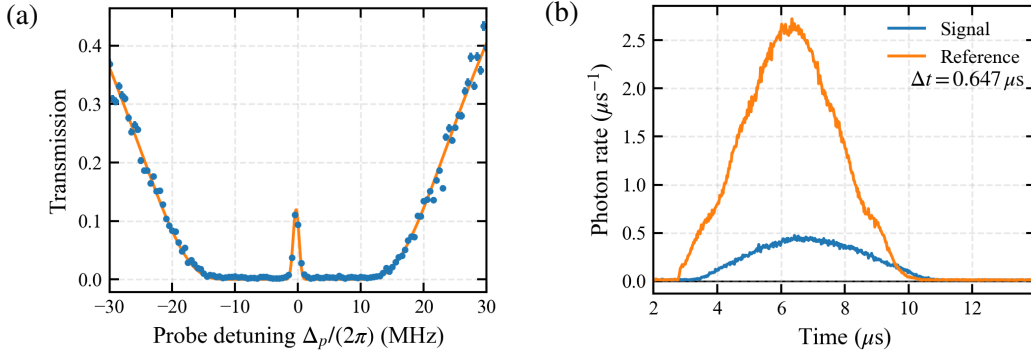


Figure 5.10: (a) Transmission for different probe detunings under EIT conditions at 54S with fit from Equation 5.9. The extracted control Rabi frequency is $\Omega_C = 2\pi \times 8.8$ MHz. (b) Photon rate measured with and without atoms on EIT resonance. The extracted pulse delay is calculated with Equation 5.10.

depth. The repeated OD scans are shown in Figure A.3 in the appendix. The scans yielded an OD of 87.14 ± 0.47 for the counting modules and 53.99 ± 0.52 for the camera. The errors were obtained from the fit. Next, the on-resonance EIT pulse delay was measured, as shown in Figure 5.10(b). The pulse delay was extracted by calculating the time centroids for both pulses

$$\tau = \langle t \rangle_{\text{signal}} - \langle t \rangle_{\text{reference}} = \frac{\int t \cdot I_{\text{signal}}(t) dt}{\int I_{\text{signal}}(t) dt} - \frac{\int t \cdot I_{\text{reference}}(t) dt}{\int I_{\text{reference}}(t) dt} \quad (5.10)$$

and resulted in a pulse delay of (647.2 ± 6.7) ns. The error was estimated by calculating the standard mean error over multiple scans. Using Equation 5.8, the OD was estimated to be 52.1 ± 5.9 , which is in agreement with the EMCCD-based value of 53.99 ± 0.52 within the respective uncertainties. The agreement between the pulse-delay estimate and the EMCCD-based OD, together with the diffraction-based explanation of the SPCM-based OD scans, indicates that the OD scans with the SPCMs should not be used for accurate OD estimation. Instead the EMCCD measurement and the pulse-delay measurement obtained with the SPCMs produced consistent results. However, the EMCCD requires significantly more measurement time than the SPCMs to achieve the same statistical precision. This is caused by large short-term fluctuations in the background signal, which strongly affect the counts integrated over the EMCCD image, as shown in Figure 5.7(b). Therefore, for applications in which the systematic deviation of the SPCM approach is not relevant, the use of SPCMs may still be preferable. One such application is the longitudinal positioning of the atomic cloud, since it was shown in Section 5.2 that changes in the mode overlap do not qualitatively affect the measurement results.

5.5 Transverse Control Beam Alignment

After studying the two-level system behaviour close to resonance and investigating different methods for dipole trap positioning, the control beam has to be aligned with respect to the atom cloud. The control beam waist is $14 \mu\text{m}$. To maximize the precision of the alignment, one mirror mount in its

beam path was replaced by a piezo-actuated mirror mount. In principle, the same three concepts that were used for the atom cloud positioning are applicable again. Before the EMCCD was integrated, the control beam was aligned by maximizing the on-resonance EIT transmission measured with the SPCMs. However, images with the EMCCD camera show that maximizing the EIT transmission with the SPCMs did not eliminate the detuning-dependent beam shifts visible in the EMCCD images. Close to the center of the atom cloud, the EIT transmission measured with the SPCMs is insensitive to small control beam movements. Therefore, two alternative control beam alignment methods are presented.

One approach again uses the EMCCD camera. Here, the diffraction effects that follow from the real part of the susceptibility in Figure 2.1(b) can be used. Under EIT conditions, a small probe detuning should result in opposite diffraction effects for opposite signs of the detuning. The result is shown in Figure 5.11 for $\Delta_p/(2\pi) = \pm 1$ MHz. Here it can be seen that the control beam alignment is optimized in the vertical direction but not in the horizontal direction, as the horizontal beam position differs visibly between the two images. For two reasons, it is important to optimize the position of the atomic cloud before aligning the control beam. First, any subsequent change in the cloud position also changes the relative alignment between the cloud and the control beam, requiring the control beam to be realigned. Second, the combination of misaligned cloud with respect to the probe and misaligned control may introduce more complex diffraction behaviour, making the control alignment procedure inaccurate.

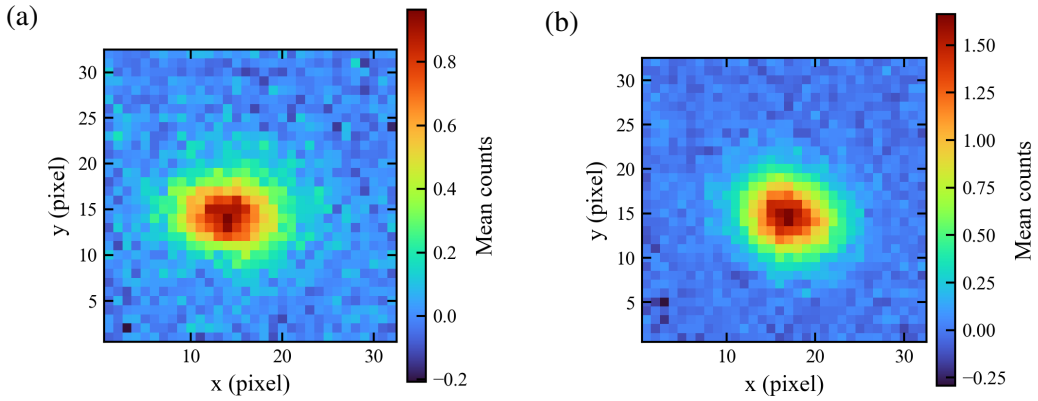


Figure 5.11: EMCCD images with diffraction under EIT conditions with small probe detuning. (a) $\Delta_p/(2\pi) = 1$ MHz. (b) $\Delta_p/(2\pi) = -1$ MHz.

Besides using the EMCCD camera, cloud splitting combined with resonant absorption imaging can be used for aligning the control beam. Here, resonant pushing of the central atoms, as used for the probe beam, is not possible since the 480 nm control laser is far-off resonance. Instead the approach is to use a high control power to overlap the attractive ODT potential with a repulsive potential created by the control laser. For this purpose, the control light is no longer pulsed, but is left on continuously after the loading of the atoms into the ODT. The measurement result for an imperfect vertical alignment of the control beam is shown in Figure 5.12. By equalizing the cloud splitting, the control beam can be aligned precisely with respect to the atom cloud.

In summary, the two-level response of the transverse photon distribution was analyzed in this chapter.

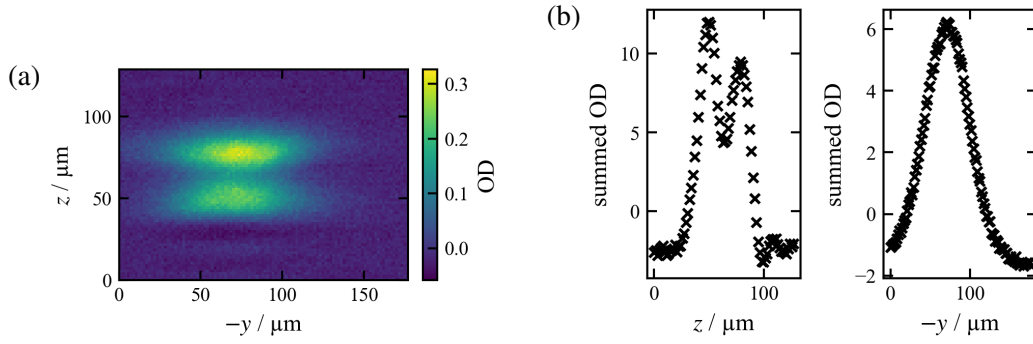


Figure 5.12: Cloud splitting with the control beam. A constant high-power control beam creates a repulsive potential which overlaps with the attractive ODT potential and creates a cloud splitting. In this image, the control beam is not centered with respect to the atom cloud along the z -direction which leads to an unequal splitting. (a) Absorption image obtained by calculating the optical depth for each pixel according to Equation 3.1. (b) Corresponding OD profiles summed along the horizontal and vertical axes

Detuning-dependent focusing and defocusing of the beam as well as significant deviations from the Gaussian profile were observed. Different alignment methods were presented and discussed. Combined with the piezo-actuated mirror mounts, these methods improve the alignment precision. Lastly, two approaches for accurate control beam alignment were presented. With the alignment carefully optimized and the two-level dynamics characterized, the basis is established for the following investigation of photon-photon interactions.

Nonlinear Transverse Response of an Interacting Rydberg Gas

In the previous chapters, the integration of the EMCCD camera into the experiment, sources of probe beam aberrations, accurate dipole trap and control beam positioning and their influence on the photon distribution were discussed. Furthermore, it was shown how the EMCCD can be used to estimate the OD and how the propagation close to the two-level resonance influences the outgoing intensity profile. After characterizing the relevant experimental influences on the photon distribution, the interaction-dependent transverse response can be investigated. The effect of Rydberg blockade on the transverse photon distribution can be studied for the mean-field distribution and for photon correlations. Experimental results on the mean-field transverse photon distribution are presented in Section 6.1. Section 6.2 presents an investigation of thresholding at the few-photon level and discusses its application to spatial correlation measurements and the remaining challenges.

6.1 Nonlinear Lensing at the Few-Photon Level

To investigate transverse photon–photon interactions in the mean-field regime, the power of the incident laser pulse is varied. Increasing the pulse power increases the average number of photons per pulse and therefore increases the probability that multiple photons interact within the atomic medium. These interactions can modify the spatial propagation of the light and therefore lead to changes in the transverse photon distribution that is imaged with the EMCCD camera.

6.1.1 Pulse Shapes

During the mean-field measurements, two different temporal pulse shapes for the probe pulses were used. The first pulse shape was a Gaussian pulse described by

$$I(t) = A \exp\left(-\frac{(t - \mu)^2}{2\sigma^2}\right). \quad (6.1)$$

In the measurements in this chapter with a Gaussian pulse shape, the center position μ and the width σ of the pulse are not changed. The photon rate is changed by changing the amplitude A . For each

amplitude setting, the mean number of photons per pulse $\bar{N}$ was measured with the SPCMs and the peak photon rate of the Gaussian pulse was calculated using

$$R_{\text{peak}} = \frac{\bar{N}}{\sqrt{2\pi} \sigma} \quad (6.2)$$

During this work, the pulse shape was changed to a Tukey pulse. This pulse shape provides a long flat-top interval with a constant photon rate, while finite rise and fall times limit spectral broadening. This pulse shape is defined by

$$I(t) = \begin{cases} \frac{A}{2} + \frac{A}{2} \cos\left(\pi \frac{t + t_{\text{rise}} + \frac{t_{\text{up}}}{2}}{t_{\text{rise}}} - \pi\right), & \text{if } -t_{\text{rise}} - \frac{t_{\text{up}}}{2} \leq t < -\frac{t_{\text{up}}}{2}, \\ A, & \text{if } -\frac{t_{\text{up}}}{2} \leq t < \frac{t_{\text{up}}}{2}, \\ \frac{A}{2} + \frac{A}{2} \cos\left(\pi \frac{t - \frac{t_{\text{up}}}{2}}{t_{\text{rise}}}\right), & \text{if } \frac{t_{\text{up}}}{2} \leq t < t_{\text{rise}} + \frac{t_{\text{up}}}{2}, \\ 0, & \text{else.} \end{cases} \quad (6.3)$$

If not stated otherwise, the Gaussian pulse is used with a width σ of 1.5 μs and the Tukey pulse is used with a rise time of $t_{\text{rise}} = 800$ ns and a flat-top duration of $t_{\text{up}} = 6.5$ μs .

6.1.2 Nonlinear Lensing for Different Rydberg States

Strong nonlinearities in the transverse photon distribution can be created through photon-photon interactions by generating large refractive-index gradients [7]. To create a refractive gradient between a photon propagating under EIT conditions and a photon for which the Rydberg level is shifted out of resonance, a single-photon detuning must be introduced. Here, a control beam detuning of $\Delta_c/(2\pi) = -20$ MHz was used. As shown in Figure 2.2(b), a probe detuning can be chosen such that the real part of the susceptibility is positive while the susceptibility of the two-level system is negative. This produces a large contrast between the refractive responses of the blockaded and unblockaded regions [47]. Experimentally, the point of maximum diffraction gradient was determined by scanning the probe transmission on the steep slope of the EIT peak. The maximum diffraction occurs at the point where the transmission lies midway between its maximum and minimum values.

To extract the beam size, the data are fitted with the two-dimensional Gaussian model defined in Equations 4.1, 4.2 and 4.3. Due to the complex dynamics of the elliptical, astigmatic beam inside the atom cloud, the angle of the main axis changes throughout the measurement series. Therefore an effective beam radius is extracted from the fitted waists r_1 and r_2

$$r_{\text{eff}} = \sqrt{r_1 r_2}. \quad (6.4)$$

The photon-photon interaction can then be observed by a changing beam size depending on the photon

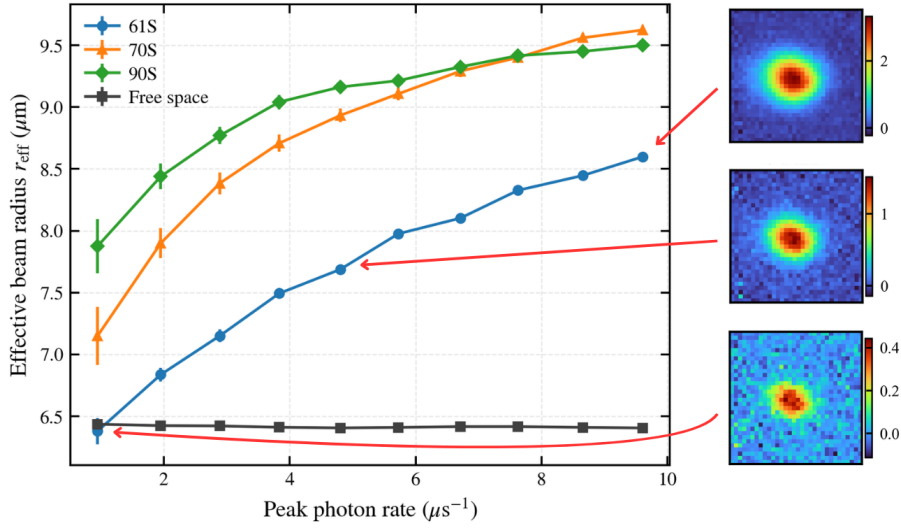


Figure 6.1: Effective beam radius for different peak photon rates of the Gaussian pulse. The curves show the result for three different Rydberg states as well as the free-space case without any atoms. The three images correspond to the marked data points for 61S and show the mean number of counts for each pixel. Error bars represent the standard errors obtained from the Gaussian fit.

density. In Chapter 5 it was shown that for the two-level system, the beam size increases compared to the free-space beam at the imaging plane. Since the sign of the susceptibility differs at the chosen probe detuning for the two- and three-level systems, a transition from a probe beam smaller than the free-space beam to a larger beam is expected.

The results for Gaussian pulses and the Rydberg states 61S, 70S and 90S as well as the reference beam are shown in Figure 6.1. The average photon number per pulse is varied from approximately 3 to 30 photons. The experimental parameters for all measurements in this section are $\text{OD} \approx 30$ and $\Omega_c/(2\pi) \approx 9 \text{ MHz}$. As given in Equation 2.15, the interaction strength highly depends on the blockade radius. By choosing different Rydberg states, the blockade radius can be varied. Since the C_6 coefficient is proportional to r_b^6 , the interaction strength increases for higher Rydberg states. Especially for low photon rates, the different interaction strengths can be observed as the beam size differs significantly for the three Rydberg states. At high photon rates, the beam radii measured for 70S and 90S approach the value associated with the two-level response. The crossing between the 70S and 90S curve may have arisen from small OD drifts between the measurements. For 61S, significantly higher photon rates are needed as the one-dimensional case is broken and polaritons can propagate on top of each other, meaning that significantly more photons are needed to saturate the cloud.

While the beam size approaches the free-space beam size for 61S for the lowest photon rate, a clear transition from a focused to a defocused beam was not observed. A further decrease in the photon rate would bring the detected signal close to the noise level of the camera, making reliable measurements increasingly difficult. Therefore the Rydberg state was switched to 54S to further lower the interaction strength. The pulse shape was changed to Tukey pulses to have a long constant photon rate and to

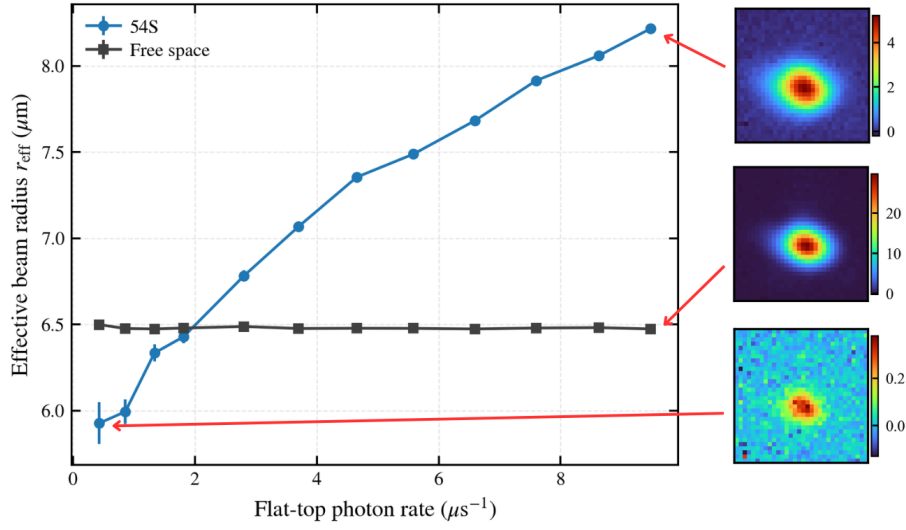


Figure 6.2: Effective beam radius for different flat-top photon rates of the Tukey pulse for 54S. The three images correspond to the marked data points and show the mean number of counts for each pixel. Error bars represent the standard errors obtained from the Gaussian fit.

optimize the spectral width of the pulse. The measurement result is shown in Figure 6.2. Here, a clear transition from a beam size smaller than the free-space value to one larger than the free-space value is observed within the investigated photon-rate range.

By changing the sign of the probe and control detunings, a corresponding reversal of the dispersive nonlinearity would be expected in the simplified model. However, this behavior was not observed experimentally. This is consistent with observations in [2], where a pronounced asymmetry with respect to the detuning sign was attributed to an additional Raman resonance involving the interaction-shifted Rydberg state at a certain interatomic separation. This resonance introduces enhanced dissipative dynamics for interacting photon pairs and therefore modifies the otherwise predominantly refractive interaction.

To verify that the observed nonlinear lensing originates from Rydberg-mediated photon-photon interactions rather than power-dependent changes in the atomic cloud, such as radiation-pressure-induced atomic motion, the measurement was repeated without the control field. The result is shown in Figure 6.3(a). It is clearly shown that the two-level system does not follow the same behaviour. Small fluctuations at low photon rates for the two-level result may arise from fit uncertainty caused by the combination of a defocused beam and a small number of photons per image, leading to an extremely low photon density per pixel. Additionally, deviations from a Gaussian mode have been observed for the two-level system, although they remain relatively small for the detuning and OD considered here.

For the Tukey pulse, the beam-radius dependence on pulse duration was measured at two probe photon rates, as shown in Figure 6.3(b). In both cases, the beam size shows a slight decrease with increasing pulse duration. However, this dependence becomes weaker for longer pulses. Since the lensing

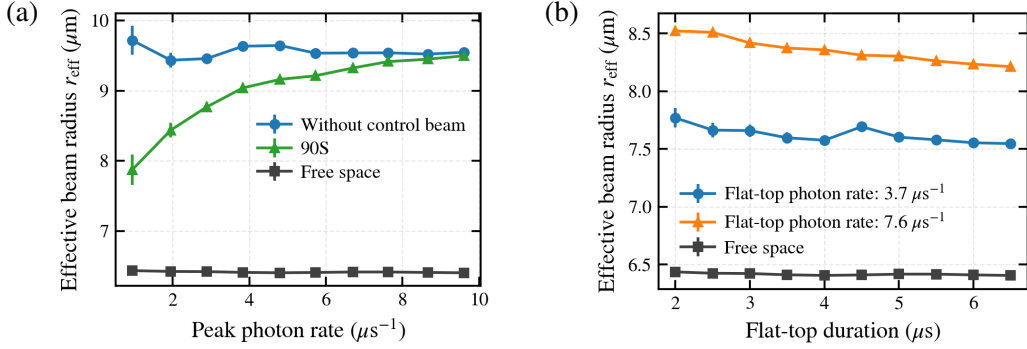


Figure 6.3: (a) Effective beam radii for the two-level response, the three-level response at 90S, and free-space propagation for different peak photon rates of a Gaussian pulse. (b) Effective beam radii for different pulse durations at 61S. The rise time of the Tukey pulse is $0.8 \mu\text{s}$ for a flat-top duration of $6.5 \mu\text{s}$. For the other data points the rise time was varied such that the ratio of rise time and flat-top duration is constant for all data points. Error bars represent the standard errors obtained from the Gaussian fit.

measurements were performed in this long-pulse regime, the continuous-wave (CW) approximation can be considered appropriate for these measurements.

6.1.3 Optically Controlled Lensing

Another parameter that can be used to tune the interaction strength between the Rydberg polaritons is the control Rabi frequency Ω_c . As shown in Equation 2.15, stronger nonlinearities are expected for smaller control Rabi frequencies. Here it has to be considered that the control Rabi frequency affects not only the interaction strength but also the initial three-level susceptibility that is given in Equation 2.7. So the variation of the control Rabi frequency is not a pure variation of the interaction strength. The measurement was performed for $\text{OD} \approx 50$ and a Rydberg state of 45S. Due to the larger OD, higher effective beam radii were achievable compared to the previous measurements. The control detuning was again chosen to be $\Delta_c/(2\pi) = -20 \text{ MHz}$. The probe detuning was again determined experimentally by identifying the point at which the transmission reaches half of the EIT peak transmission. Since the control Rabi frequency affects the EIT linewidth, slightly different probe detunings were required to operate at the point of maximum diffraction gradient. Consequently, changing the control Rabi frequency affects not only the three-level susceptibility but also, indirectly, the two-level susceptibility due to the corresponding change in probe detuning.

The result is shown in Figure 6.4. The increased beam size due to larger interaction strength for smaller control Rabi frequencies can be clearly observed for all three probe detunings. Section 5.5 demonstrated that the repulsive potential generated by a continuously applied high-power control beam can significantly alter the atomic distribution. As a control measurement, the experiment was therefore repeated using the same control powers as those corresponding to the control Rabi frequencies in Figure 6.4, but with the control field detuned by several gigahertz. Under these conditions, no measurable change in the beam shape was observed, indicating that control-beam-induced modifications of the atomic distribution are negligible in the power regime used for the present measurements.

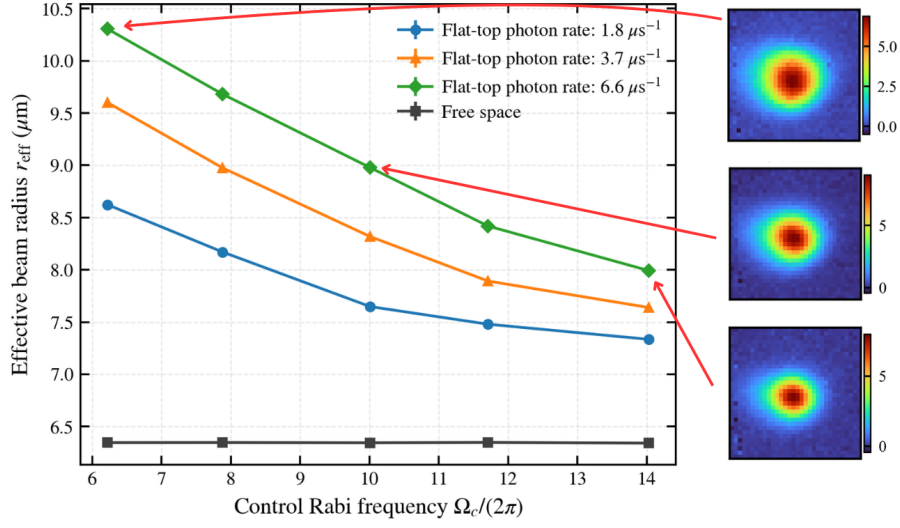


Figure 6.4: Effective beam radii in dependence of the control Rabi frequency for different probe photon rates. The three images correspond to the marked data points and show the mean number of counts for each pixel. Error bars represent the standard errors obtained from the Gaussian fit.

In summary, this section showed the observation of power-dependent changes in the transverse probe-beam profile at the few-photon level, which were attributed to Rydberg-mediated photon–photon interactions. By varying the principal quantum number, the interaction strength could be modified, resulting in distinctly different dependencies of the effective beam radius on the photon rate. For the 54S state, a clear transition from a beam size below to one above the free-space reference value is observed with increasing photon rate. Control measurements without a control field showed no comparable behavior, supporting that the observed lensing originates from the Rydberg-EIT nonlinearity rather than from linear two-level effects or power-dependent changes of the atomic density distribution. Measurements with different pulse durations further indicated that the pulse duration used in the experiment is adequately described by the CW approximation. Finally, the transverse nonlinear response could also be controlled through the control Rabi frequency, demonstrating optically tunable lensing at the few-photon level. It should be noted that changing the control Rabi frequency not only modifies the interaction strength but also affects the three-level susceptibility. Furthermore, by shifting the maximum of the real part of the susceptibility, it changes the choice of probe detuning and, consequently, the two-level susceptibility.

6.2 Towards Photon Counting and Spatial Correlation Measurements

Averaging the EMCCD counts for each pixel over multiple pulses and experimental cycles and subtracting the background counts with dark images revealed strong nonlinearities of the transverse photon distribution caused by photon-photon interaction. However, this method cannot be used to measure spatial correlations of interacting photons based on transverse Rydberg blockade. To move from averaged images to single-event measurements, the counts of the EMCCD camera have to be converted into photon events for each image. This can be realized by setting a threshold for the EMCCD

counts. For each image, all pixels above the threshold are counted as photon events. The correlation measurements are performed at a very low illumination level, such that the probability of multiple photons being detected on the same pixel is negligible. This section discusses the noise sources of the EMCCD and possible thresholding choices as well as experimental requirements for spatial correlation measurements.

6.2.1 Noise Modelling and Thresholding

To investigate correlations between individual photons instead of mean-field effects, a different data-processing approach is required. To extract photon events from each individual frame, a binary threshold is applied: pixels below the threshold are assigned zero photon events, whereas pixels above the threshold are assigned one photon event. To determine an appropriate threshold, the EMCCD noise distribution must be examined in more detail.

The process of EMCCD imaging can be divided into three stages, with each stage introducing a different type of noise [48]. The first stage is the illumination of the camera chip where the photoelectrons are accumulated. The number of generated photoelectrons follows a Poisson distribution. In the second stage, stochastic multiplication in the EM register produces a gamma-distributed output for a fixed number of input electrons. The last stage is the readout process. The readout noise follows a normal distribution. Overall, the EMCCD noise can be modelled with a Poisson-gamma-normal distribution [49].

The camera was operated at a temperature of -93°C to ensure that the dark-current contribution was negligible on the acquisition timescale. The remaining background noise is dominated by the readout noise and the spurious charge generation. Spurious charges are generated by holes undergoing impact ionization during the charge transfer process, which is known as clock-induced charge (CIC) [32]. This process can occur in the imaging or storage area during the vertical charge shift and is known as parallel-CIC. With the formulas given in [50], the parallel-CIC probability distribution is

$$p(x_p) = p_p \frac{\exp\left(-\frac{x_p}{\phi G_{EM}}\right)}{G_{EM}} \quad (6.5)$$

with p_p being the probability for a parallel-CIC event, x_p being the number of counts for a parallel-CIC event, G_{EM} being the EM gain and ϕ being the conversion factor of 4.92 electrons per count. Additional spurious electrons can be generated during charge transfer through the multiplication register. These events are referred to as serial-CIC. In this case, the spurious charge passes through only a fraction of the amplification register rather than its entire length. The probability distribution for a spurious charge generated inside the multiplication register is

$$p(x_s) = p_s \sum_{i=1}^m \frac{\exp\left(-\frac{x_s}{\phi p_c^{m-i}}\right)}{p_c^{m-i}} \quad (6.6)$$

and is adapted from [51], with m being the number of cells in the multiplication register, x_s being the number of counts and p_s being the probability of a serial-CIC event. The probability p_c that an electron

generates another electron in one cell of the multiplication register can be determined from the mean EM gain

$$G_{\text{EM}} = (1 + p_c)^m. \quad (6.7)$$

The camera has 536 cells in its multiplication register.

The full noise model is obtained by the convolution of all noise sources. By the convolution theorem, its characteristic function is

$$\begin{aligned} \Phi_{\text{tot}}(q) &= \Phi_{\text{PG}}(q) \cdot \Phi_{\text{N}}(q) \cdot \Phi_{\text{sCIC}}(q) \\ &= \exp\left(p_p \left(\frac{1}{1 - iqG} - 1\right)\right) \exp\left(-\frac{\sigma^2 q^2}{2} + iqS_0\right) \\ &\quad \cdot \left(1 - mp_s + p_s \sum_{j=0}^{m-1} \frac{1}{1 - iq(1 + p_c)^j / \phi}\right). \end{aligned} \quad (6.8)$$

The total probability density is recovered by the inverse transform,

$$p_{\text{noise}}(x) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \Phi_{\text{tot}}(q) e^{-iqx} dq. \quad (6.9)$$

The noise distribution and the corresponding fit are shown in Figure 6.5. The obtained noise parameters are $\sigma = 20.6$ counts, $p_s = 1.3 \cdot 10^{-4}$ and $p_p = 1.5 \cdot 10^{-2}$.

The threshold should maximize the ratio of true to false counts while minimizing the loss of real photon events. The threshold changes the retained contribution of the readout noise. However, it cannot discriminate between photoelectrons and parallel-CIC electrons because both move through the complete EM register and therefore follow the same probability distribution. Typically, a threshold in the range of 3σ to 5σ of the readout noise is chosen [52, 53]. This range is illustrated by the grey area in Figure 6.5.

A different approach can be used to confirm that the threshold range is reasonable. Therefore, the mean number of counts above threshold was calculated for a series of dark images and images with an input rate of approximately two photons per pulse reaching the EMCCD chip. To reduce the number of false counts, each image was cropped during post-processing to an 8×8 pixel ROI centred on the beam. The counts above threshold for different thresholds T are shown in Figure 6.6(a). Using

$$R(T) = \frac{N_l(T) - N_d(T)}{N_d(T)} \quad (6.10)$$

with N_l and N_d being the counts above threshold with and without illumination, the true-to-false count ratio $R(T)$ can be determined for each threshold, as shown in Figure 6.6(b). The true-to-false count ratio

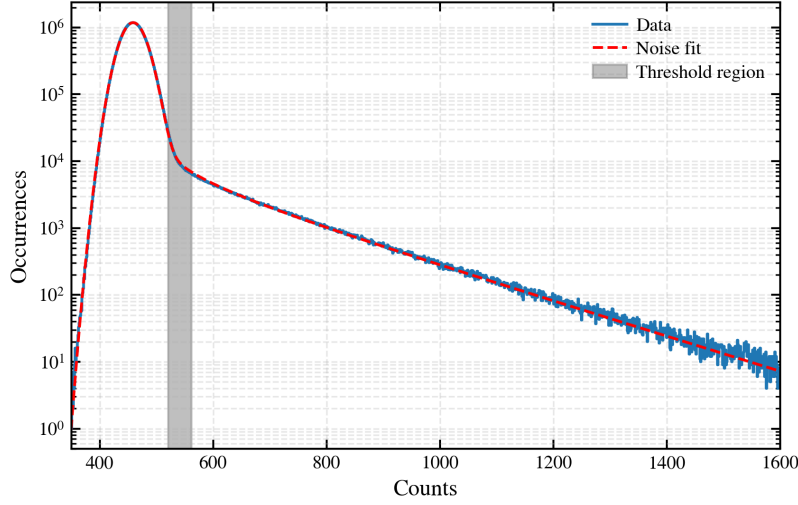


Figure 6.5: Histogram of EMCCD counts for a series of dark images in logarithmic scale with EMCCD noise fit based on a Poisson-gamma-normal distribution with parallel- and serial-CIC. The obtained fit parameters are $\sigma = 20.6$ counts, $p_s = 1.3 \cdot 10^{-4}$ and $p_p = 1.5 \cdot 10^{-2}$ and $S_0 = 458.7$. The grey area shows the region between $S_0 + 3\sigma$ and $S_0 + 5\sigma$ illustrating the range of commonly used thresholds.

approaches a plateau for background-subtracted thresholds between 60 and 100 counts, corresponding to the range of 3σ to 5σ . A higher threshold would therefore increase the loss of true photon events without noticeably improving the ratio.

In comparison to previous work with the same camera and the same shift speeds, the number of CIC events increased by approximately one order of magnitude [54]. This is a major limitation for photon-correlation measurements, since the number of false counts is approximately ten times higher than it used to be, ultimately exceeding the number of true counts for few-photon pulses, as illustrated in Figure 6.6. In [54], the VSA setting 'Normal' instead of '+1' was used. However, measurements performed with the 'Normal' setting exhibited charge smearing, making this setting unsuitable as it was shown in Figure 3.4. A possible explanation is ageing of the EMCCD camera. Before recalibration at the beginning of this work, the measured EM gain differed from the software setting by almost a factor of three, indicating substantial calibration drift due to ageing effects of the EM register. The recalibration led to a higher multiplication-register voltage, which may increase the serial-CIC rate. The increase in parallel-CIC and the charge smearing observed with the 'Normal' clock amplitude cannot be directly attributed to ageing of the EM register. However, the pronounced ageing effects observed in the EM register suggest that other parts of the camera may have been affected by ageing as well. Nevertheless, the origin of the increased CIC cannot be determined reliably without further investigation.

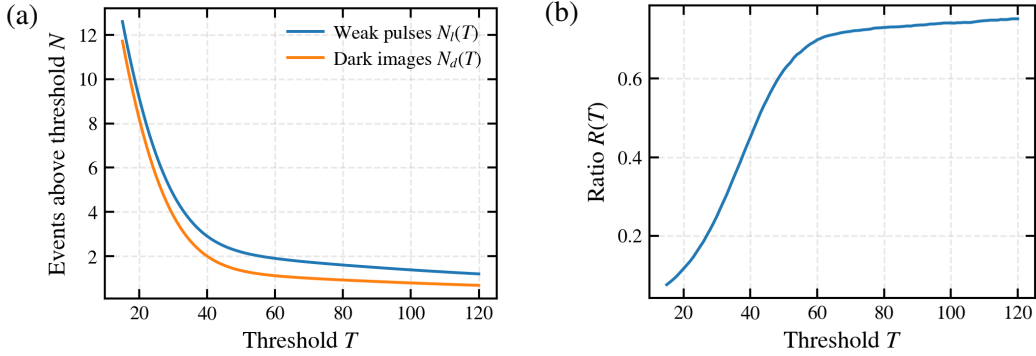


Figure 6.6: (a) Events above threshold for different thresholds for dark images and images with weak probe pulses with an 8×8 ROI. (b) True-to-false count ratio calculated using Equation 6.10.

6.2.2 Short Pulsing

In order to measure spatial photon correlations, the polaritons should only propagate on top of each other rather than in a longitudinal chain since only one image can be taken during a single pulse. Having multiple polaritons behind each other in a single pulse will mix interacting and non-interacting photon pairs and thereby wash out the measured correlation.

To prevent the polaritons from forming a longitudinal chain, the pulse duration must be shorter than the width of the temporal correlation function. The temporal correlation functions for the Rydberg states 45S, 54S and 70S are shown in Figure 6.7. The data show that the width of the $g^{(2)}(\tau)$ function increases for Rydberg states with a higher principal quantum number due to a larger blockade radius. Consequently, a high principal quantum number is preferable for increasing the interaction strength. At the same time, the Rydberg state influences the depth of the $g^{(2)}(\tau)$ function. For 45S, only weak antibunching is observed, since the smaller blockade radius allows multiple polaritons to propagate alongside one another. For 70S, $g^{(2)}(0)$ approaches zero, which means that the result is consistent with strong transverse blockade approaching a one-dimensional case. Therefore, the choice of principal quantum number is constrained by the requirement that the cloud is not fully transversely blocked. For our experimental conditions, Rydberg states between 54S and 70S represent a reasonable compromise between maximizing the interaction strength and avoiding the one-dimensional regime.

The temporal envelope of a light pulse and its frequency spectrum are related by a Fourier transform. The spectral amplitude is given by

$$\tilde{E}(\omega) = \int_{-\infty}^{\infty} E(t) e^{-i\omega t} dt \quad (6.11)$$

where $E(t)$ is the complex electric-field envelope of the pulse. Therefore, a strong temporal confinement of the optical field leads to a broader frequency range. This relation can be expressed by the time-bandwidth product

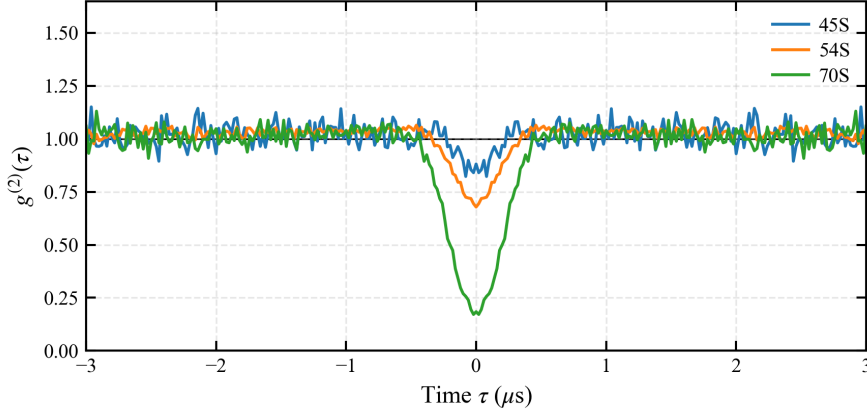


Figure 6.7: Temporal second-order correlation functions under EIT conditions for different Rydberg states.

$$\Delta t \Delta \nu \geq K \quad (6.12)$$

where Δt and $\Delta \nu$ denote the temporal and spectral widths, respectively, and K depends on the pulse shape. For a Gaussian pulse, using the full width at half maximum (FWHM) of the intensity distributions, the relation becomes

$$\Delta t \Delta \nu = \frac{2 \ln 2}{\pi} \approx 0.441. \quad (6.13)$$

The dependence of the time-bandwidth product on the pulse shape is discussed in [55].

Assuming a constant temporal phase, the field envelope was reconstructed by taking the square root of the measured photon rate and then Fourier transformed according to Equation 6.11. The squared magnitude of the resulting spectral amplitude was normalized to its maximum for each pulse. The calculated frequency spectra for two different input-pulse durations under EIT conditions are shown in Figure 6.8. It directly compares the frequency spectra for the incoming and outgoing photons. For the longer Tukey pulse, with a rise time of 250 ns and a flat-top duration of 800 ns, most of the spectral distribution lies within the EIT window, while only the secondary maxima of the spectral intensity extend beyond it. However, this pulse duration would still result in a chain of polaritons since it can be seen in Figure 6.8(a) that the signal pulse begins well before the reference pulse has ended, meaning that the leading part of the pulse leaves the cloud before the trailing part enters. Consequently, not all polaritons coexist in the medium. For the shorter pulse shown in Figure 6.8(c), the first photon leaves the cloud after the last photon enters the cloud. This pulse duration lies within the measured correlation time, ensuring interaction between the outgoing photons. However, as shown in Figure 6.8(d), the spectral width increases substantially, resulting in only a small fraction of the pulse spectrum lying within the EIT window.

Several approaches can be used to obtain a pulse whose duration is shorter than the correlation time

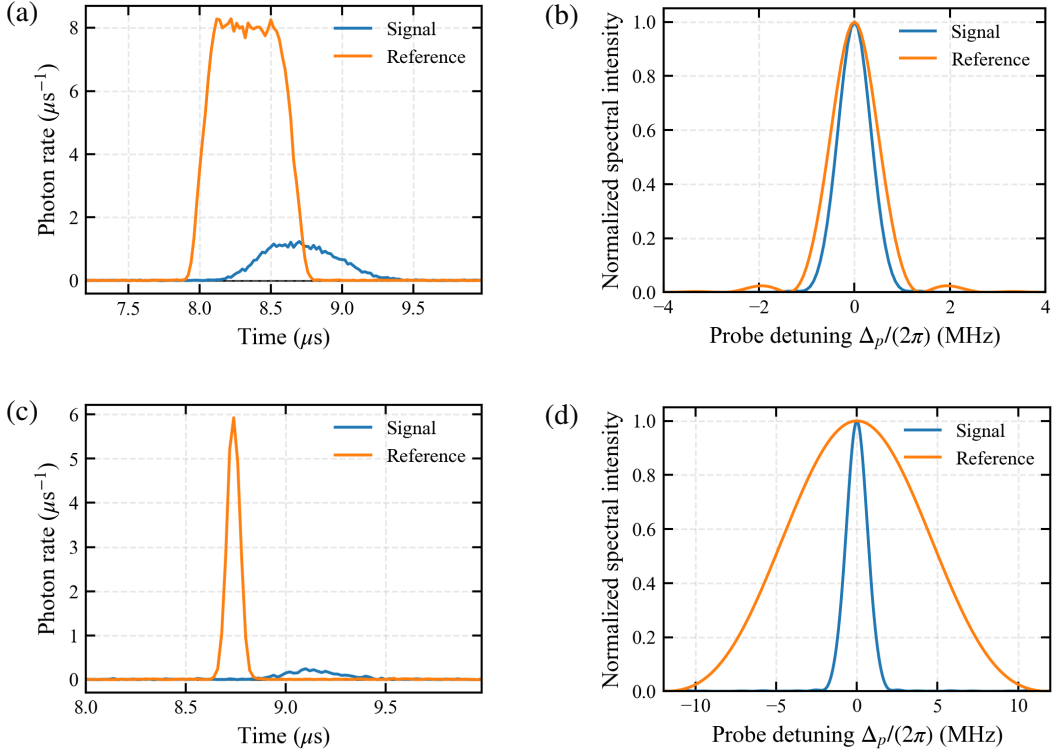


Figure 6.8: Short Pulses under EIT conditions on 61S. The signal trace denotes the pulse recorded with atoms, whereas the reference trace denotes the pulse recorded without atoms. (a) Tukey pulse with 800 ns flat-top duration and 250 ns rise time. (b) Normalized magnitude of the discrete Fourier transform of the photon-rate profiles shown in (a). (c) Tukey pulse with a 60 ns flat-top duration and a 30 ns rise time. (d) Normalized magnitude of the discrete Fourier transform of the photon-rate profiles shown in (c).

while its spectral width remains smaller than the EIT bandwidth. The EIT width scales with Ω^2 [19]. However, Equation 5.8 shows that for a constant OD, the pulse delay τ scales with Ω^{-2} . Therefore, the control Rabi frequency is not a useful parameter as it changes both the interaction time and the EIT-width. A more promising approach is increasing the OD. This reduces the polariton propagation speed allowing pulses with a larger temporal and shorter spectral width. Different methods can be used to increase the OD. The MOT loading time can be increased, however, this would lengthen the atom-preparation cycle and reduce the data acquisition rate. The rubidium-dispenser current can be increased to obtain a higher background pressure. The downside is that this reduces the lifetime of the ODT. A 2D MOT can be used to increase the loading rate of the 3D MOT by providing an intense source of rubidium atoms [56].

6.2.3 Readout Correlations

In Subsection 6.2.1, it was discussed that the fraction of background counts for the spatial correlation measurement will be significantly higher than in previous temporal correlation measurements. In principle, the true correlation function can be estimated after correcting for an uncorrelated background using [57, 58]

$$g_{\text{BC}}^{(2)}(\tau) = \frac{g^{(2)}(\tau) - (1 - \rho^2)}{\rho^2} \quad (6.14)$$

where $g_{\text{BC}}^{(2)}(\tau)$ is the background corrected correlation function, $g^{(2)}(\tau)$ is the measured correlation function including false background counts and ρ is the fraction of true counts. The same principle can be applied to spatial rather than temporal correlation measurements. However, the approach in Equation 6.14 is only valid under the assumption that the background events are uncorrelated with the signal and with one another. Since the background count rate will exceed the signal count rate for the spatial correlation measurement, it becomes particularly important that background events above threshold are uncorrelated.

To determine whether the background events are uncorrelated, the spatial correlation function is calculated for a long series of dark images. Therefore, the mean number of above-threshold events per pixel is calculated

$$\bar{I}(a) = \bar{I}(x_a, y_a) = \frac{1}{N} \sum_f I_f(a) \quad (6.15)$$

where $I_f(a)$ is the binary event value at pixel a in frame f , f is the frame index, and N is the total number of frames. For the calculation of the spatial correlation function, the data are binned as illustrated in Figure 6.9(a). The black pixel is the reference pixel, and each coloured pixel illustrates the corresponding radial bin in the correlation function if there is an event in the colored pixel and in the reference pixel. The binning size is one pixel. So for the bin centered at $d = 2$ px all pixel pairs with separations from $d = 1.5$ px to less than $d = 2.5$ px are included. This method prevents having an increased number of data points with very small distances relative to each other due to different diagonals which would give the misleading impression of a higher resolution than is actually achieved

$$g^{(2)}(r) = \frac{\sum_f \sum_{|a-b| \in [r-0.5, r+0.5)} I_f(a) I_f(b)}{N \sum_{|a-b| \in [r-0.5, r+0.5)} \bar{I}(a) \bar{I}(b)}. \quad (6.16)$$

The calculated spatial correlation function is given in Equation 6.16. Using this, the spatial correlation function is calculated for a series of 2.88×10^6 dark images for two different thresholds as shown in Figure 6.9(b) and 6.9(c). The data exhibit two systematic features. First, it seems like overall the baseline is systematically slightly above one, where one corresponds to uncorrelated events. This global bunching becomes smaller for higher thresholds. The second observation is that there is a significant bunching at the first data point and a significant antibunching for the second data point. The antibunching for the second data point becomes weaker for higher thresholds whereas the bunching at the first data point does not change significantly for both thresholds.

To investigate the origin of the behaviour observed in the first two data points, the covariance

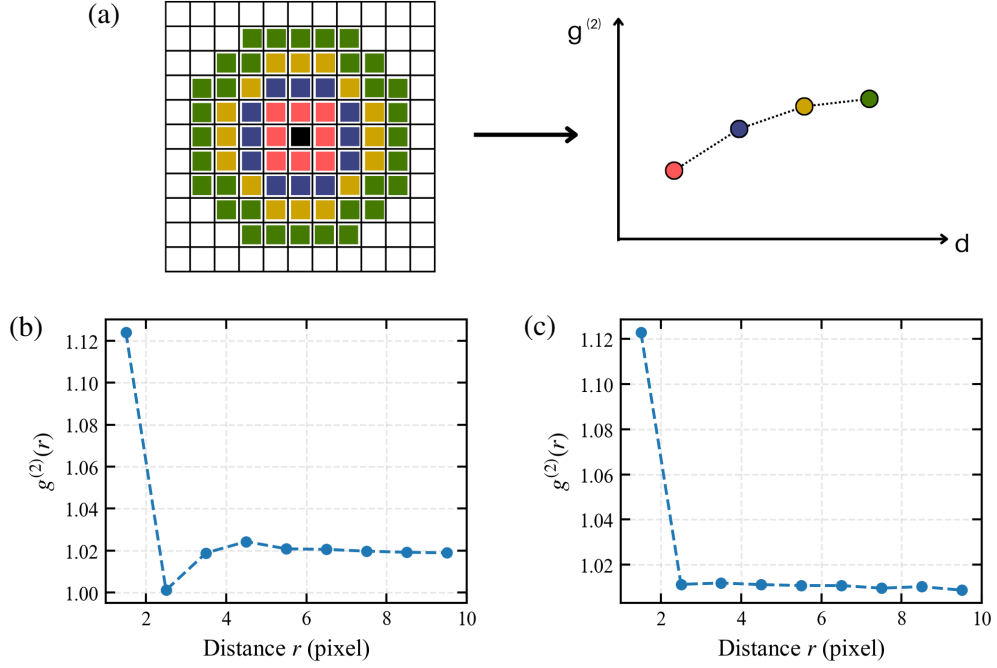


Figure 6.9: Spatial correlations with EMCCD. (a) Illustration of the binning. Each data point represents the width of one pixel. The colored pixels illustrate the corresponding data point for a coincidence with an event in a black pixel and in a colored pixel. (b) Correlation measurement with dark images and threshold 40. (c) Correlation measurement with dark images and threshold 80.

$$\text{Cov}(a, b) = \langle ab \rangle - \langle a \rangle \langle b \rangle \quad (6.17)$$

is calculated between different pixels. A covariance of zero indicates no correlation between the two pixels, whereas positive and negative covariances indicate positive and negative correlations, respectively. Figure 6.10 shows covariance maps relative to two different reference pixels. It can be seen in both cases that there are strong correlations with nearby pixels in the same sensor column. The same pattern was observed for many reference pixels, indicating that the short-range vertical correlation is present across the whole image area. A similar behaviour of correlated pixels in the vertical rows was observed in [9] where this effect was explained by charge smearing during the readout process. To reduce this artificial correlation between pixels in the same column, same-column coincidences are excluded from the spatial correlation function, as shown in Equation 6.18.

$$g^{(2)}(r) = \frac{\sum_f \sum_{|a-b| \in [r-0.5, r+0.5], x_a \neq x_b} I_f(a) I_f(b)}{N \sum_{|a-b| \in [r-0.5, r+0.5], x_a \neq x_b} \bar{I}(a) \bar{I}(b)}. \quad (6.18)$$

A small global bunching remains for dark images. A first explanation was the background drift shown in Figure 5.7(b). The background is constantly taken during the measurement and an averaged background is subtracted for all data. This approach was valid for the mean-field case. However,

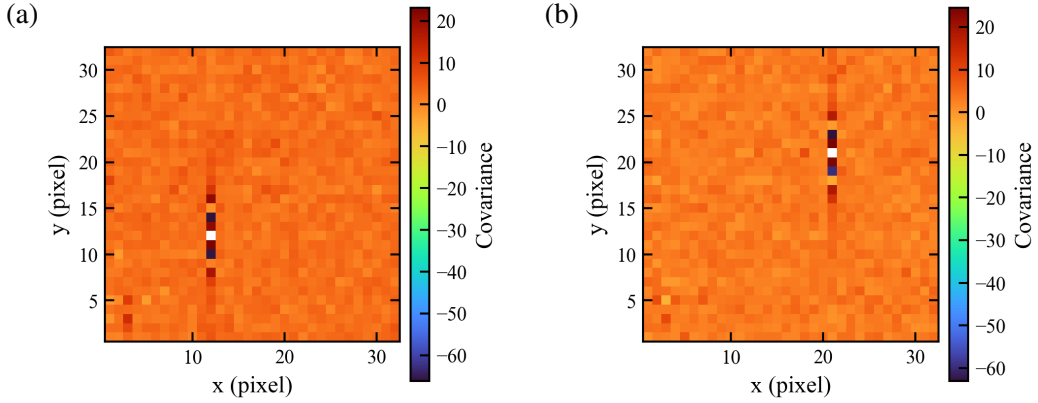


Figure 6.10: Covariance maps showing the covariance calculated using Equation 6.17. The white pixel is the reference pixel. The colour of each remaining pixel represents its covariance with the reference pixel. (a) Reference pixel (12,12). (b) Reference pixel (21,21).

for thresholding, the background drift may introduce artificial bunching. Therefore, the data were divided into shorter time windows, and a separate background estimate was subtracted from each subset. Although this approach is important for accounting for long-term background drift, it did not explain the weak global bunching.

Another possible explanation was that the number of CIC-induced coincidences differed systematically between pulse windows. Figure 6.11 shows the average number of events above threshold for a threshold of sixty along the pulse series. Each image was cropped to a 16×16 pixel ROI during post-processing. The advantage of cropping the image during data processing is that dark counts from regions without probe light can be excluded from the analysis. Here it can be clearly seen that the CIC event probability depends on pulse index. The number of dark events is elevated during the first pulses and then approaches a baseline after approximately fifty to one hundred pulses. A possible explanation for this behaviour is residual charge accumulation during atom preparation, during which no images are acquired. Additional dummy frames during atom preparation may reduce the initial excess of dark events, thereby reducing both the global background correlation and the total number of dark counts and coincidences.

The proposed measures are intended to suppress the observed readout correlations in dark images, which is a necessity for measuring transverse photon-photon correlations. In summary, in this section, necessary steps towards spatial correlation measurements were discussed. It was shown that the dark count rate due to CIC events is a major limitation for measurements of photon-photon correlations and that the number of CIC events increased in comparison with previous studies with the same camera [54]. It was shown that, for the measurement of transverse correlations, the pulse duration should be shorter than the width of the temporal correlation function. At the same time, a sufficiently high OD is required to ensure that the spectral width remains within the EIT window. Finally, correlations of dark counts were measured, and methods for suppressing these readout correlations were discussed.

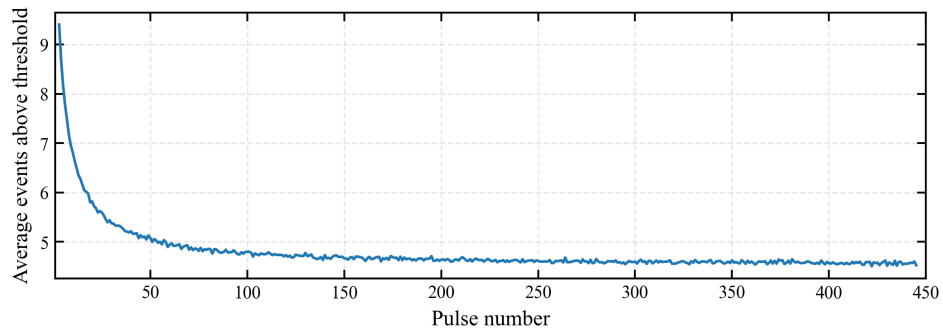


Figure 6.11: Mean number of above-threshold events per dark frame within a 16×16 pixel ROI, plotted as a function of pulse number. The threshold is set to 60. The dark-event rate is increased during the first approximately 50-100 pulses per cycle.

Conclusion and Outlook

In this thesis, effective photon-photon interactions in an ultracold atomic gas under EIT conditions were experimentally investigated. Their influence was observed as power-dependent changes in the transverse distribution of the transmitted probe light. First, the theoretical concepts of EIT and effective photon-photon interactions mediated by Rydberg excitations were introduced. Next, the experimental setup and the integration of the EMCCD camera were presented. The photon numbers estimated with the SPCMs and the EMCCD for few-photon pulses showed a reproducible difference of 25%-30%. Possible contributions to the difference in photon number include detector quantum efficiencies that could not be independently verified, multimode-fiber coupling, and charge-transfer losses during vertical shifting of the EMCCD, but no dominant source was identified.

Because the shape of the probe beam, which is expected to be modified by Rydberg-Rydberg interactions, is crucial for the detection of transverse interactions, a dedicated test setup for the probe beam was built to investigate beam imperfections introduced by the optical components. It was investigated how the different optical elements influenced the beam profile close to the focal plane where the probe photons interact with the atomic cloud. It was shown that the main source of the astigmatism that was observed in the experiment is caused by the lens alignment while the polarization optics and dichroic mirrors contribute only to a minor extent. It was shown that modifying the fiber outcoupler could increase the quality of beam collimation. Furthermore, a second modified outcoupler can be used to collect the probe photons for detection on single-photon counters. Combined with precise alignment of the two probe lenses, this increases the fiber coupling efficiency from 80% to almost 90% and thereby almost halves the coupling losses into the single-mode fiber.

The possibility of resolving the transverse probe photon distribution increased the demand for precise atomic cloud and control beam positioning to exclude diffraction effects due to imperfect alignment. Therefore, piezo-actuated mirror mounts were included in a previous project [41] to improve the alignment precision. It was demonstrated that the previously used SPCM-based alignment method provides limited accuracy and may even lead to systematic misalignment of the position of the atomic cloud under certain conditions. Two alternative approaches were therefore introduced to improve the cloud alignment which are direct alignment based on near-resonant diffraction observed with the EMCCD camera and an alignment method based on cloud splitting measured by absorption imaging. For the control beam alignment, the same two approaches can be used. However, instead of creating cloud splitting by shooting a hole

through the atomic cloud with a resonant beam, the control beam is aligned by overlapping its repulsive potential with the attractive potential of the dipole trap and detecting the splitting by absorption imaging. Furthermore, it was shown that the optical depth can be extracted from EMCCD measurements without additional photon loss caused by near-resonant diffraction, because of the large field of view of the camera.

Finally, the effect of photon-photon interactions on the transverse beam profile was measured. Therefore, a control detuning of -20 MHz was chosen. The probe detuning was chosen to maximize the difference between the real part of the two- and three-level susceptibilities, corresponding to the point at which the transmission reaches 50% of the EIT peak transmission. In the mean-field regime, the photon-photon interaction results in a power-dependent change in the transverse beam profile. Due to the opposite signs of the real parts of the two- and three-level susceptibilities, a transition from focusing to defocusing relative to the free-space beam was observed. Furthermore, it was shown that the interaction strength can be varied by changing the control beam Rabi frequency and the principal quantum number of the Rydberg state. Finally, preliminary steps towards spatial correlation measurements were presented, including thresholding, short-pulse analysis, and the suppression of dark-count correlations.

Outlook

The next step for the mean-field measurements is to numerically simulate the propagation of the probe beam through the three-level atomic medium, including the Rydberg-mediated nonlinearity, and to compare the results with the experimental observations. Such a comparison would provide a direct test of the applicability of mean-field theory in the few-photon regime.

The next goal of this experiment is to measure transverse photon-photon correlations. It was shown that the number of clock-induced charge events significantly increased compared to measurements when the camera was first used. Since this represents a major limitation for the planned correlation measurements, the origin of the increased clock-induced charge rate needs to be determined. A useful first step is to repeat the measurement using the same experimental conditions and camera settings as in [54]. This will help determine whether the increased clock-induced charge rate is caused by ageing of the shifting registers or by the current camera settings. Furthermore, pulse shortening is required to prevent the formation of a polariton train that would wash out the correlations in the recorded images. Depending on the final conclusions regarding the EMCCD dark counts, another possibility is to use a different type of spatially resolving single-photon detector. A possible option is a single-photon avalanche diode (SPAD) array. SPAD arrays have already been demonstrated to be a useful tool for correlation measurements [59–61]. Compared with EMCCD cameras, SPAD arrays provide higher temporal resolution and do not suffer from clock-induced charge, while their quantum efficiency is typically lower.

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Appendix

A.1 Atom loss during pulse sequence

This appendix presents an investigation of the influence of atom loss on the measurement results. During normal operation, the dipole-trap pulsing results in atom loss over the course of the pulse sequence, which leads to a decrease in OD. This effect is shown for different pulse windows using the SPCMs and the EMCCD camera in Figure A.1.

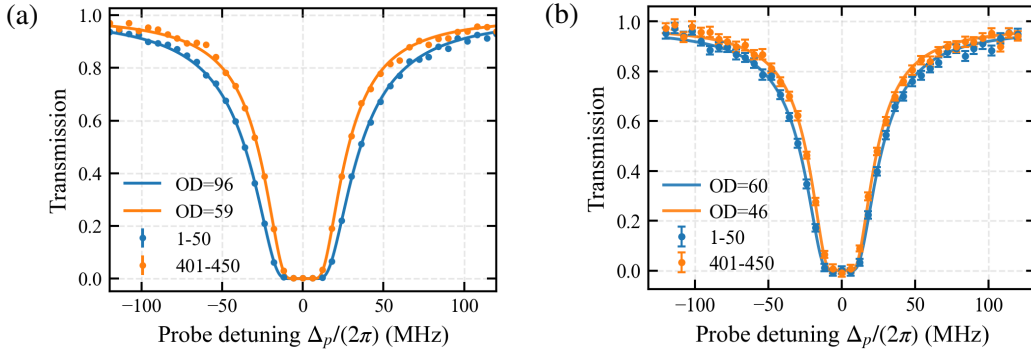


Figure A.1: Probe-detuning scans for a two-level system including only the first 50 pulses (blue) and the last 50 pulses (orange), with Lorentzian fits using Equation 5.4 to extract the OD. The decrease in OD is caused by atom loss throughout the pulse sequence. The shown pulse windows are extracted from the full data set presented in Figure 5.8. (a) OD scan using the SPCMs. (b) OD scan using the EMCCD camera.

To keep the optical depth approximately constant, the power of the optical dipole trap was linearly increased during the pulse sequence. This reduces the width of the atomic cloud and increases the atomic density near its center, thereby compensating for the decrease in OD caused by atom loss. However, this approach comes at a cost. As shown in Figure A.2, the temperature at the end of the pulse sequence increased by more than a factor of two due to the dipole-trap power ramp.

To compare the SPCMs and EMCCD results for a constant OD with a third method, the measurements had to be repeated on the same day as the estimation with the pulse delay was performed. This result is

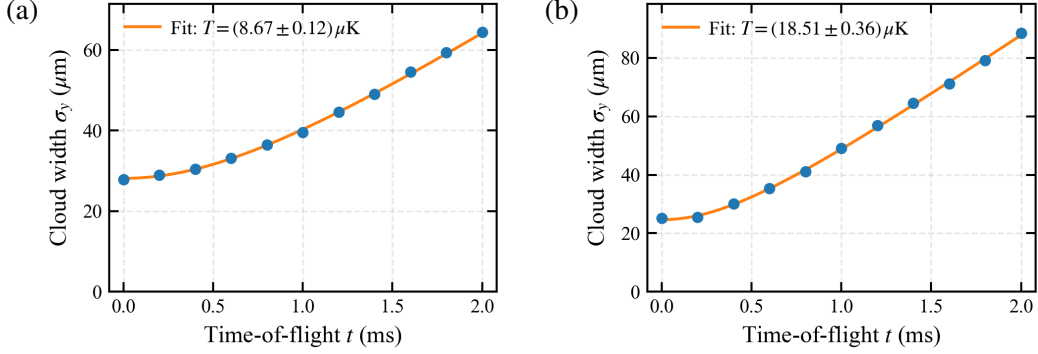


Figure A.2: Measurement of cloud size after different time-of-flights. Equation 5.7 is used for fitting the data and extracting the temperature (a) Without changing the dipole trap power. (b) With linear increase of the dipole trap power throughout the pulse series.

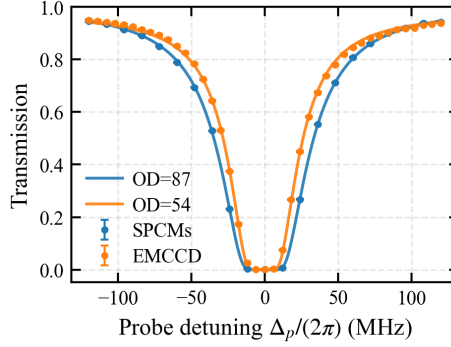
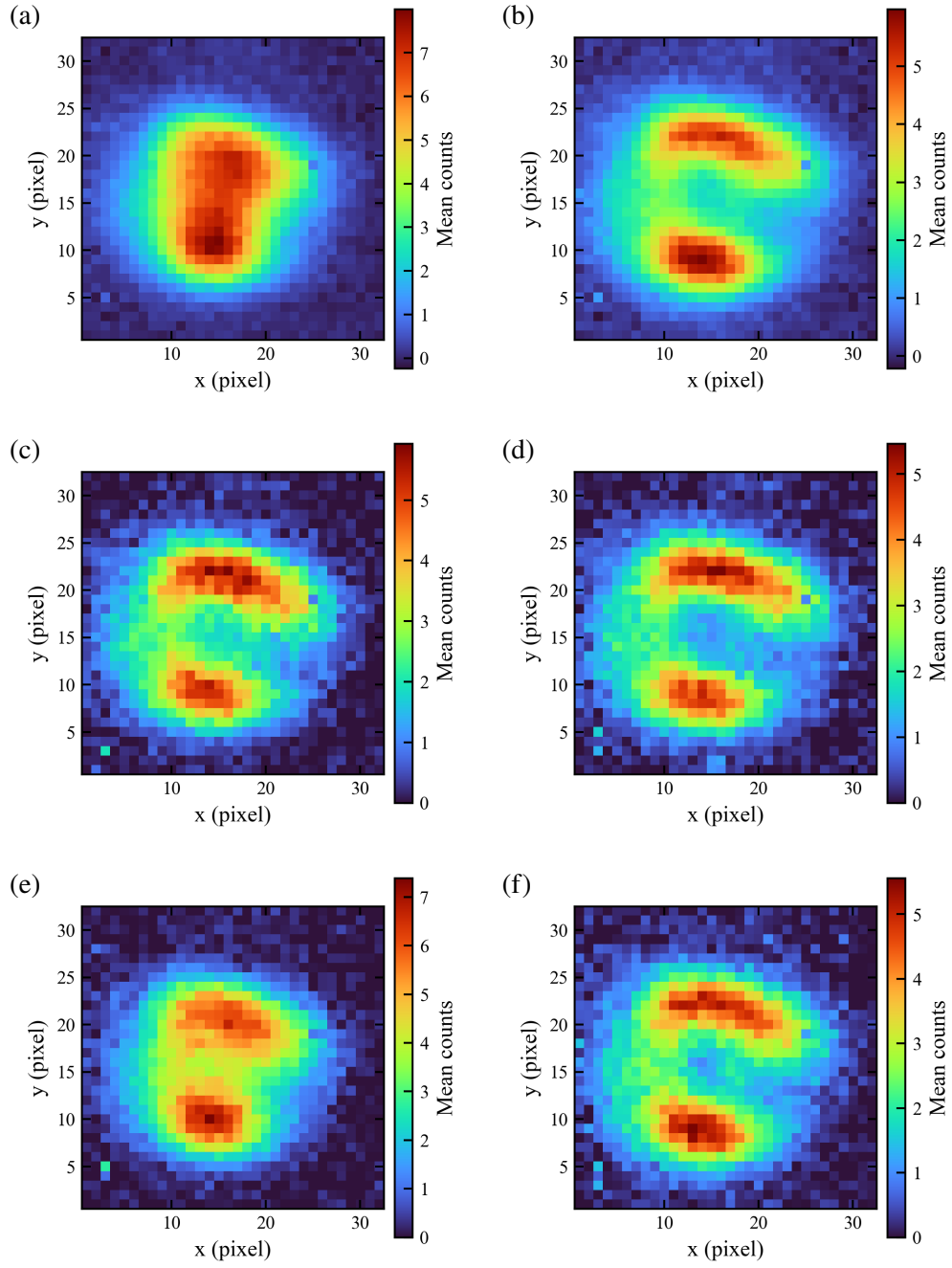


Figure A.3: Probe-detuning scans for a two-level system with SPCMs (blue) and EMCCD (orange) with Lorentzian fit from Equation 5.4 to extract the OD. The data was taken for comparison with the OD extracted from pulse delay as shown in Figure 5.10.

shown in Figure A.3.

Finally, the two approaches, with and without a linear increase in the dipole-trap power, were compared using the EMCCD camera. The corresponding images are shown in Figure A.4 for the complete pulse sequence and for different pulse windows. Without the dipole-trap power ramp, the decreasing OD results in a significant change in the beam profile throughout the pulse sequence. Consequently, averaging over the entire pulse sequence obscures the beam profile observed during the first 90 pulses. In contrast, when the OD is kept approximately constant by increasing the dipole-trap power, the beam profiles remain much more similar between the different pulse windows.



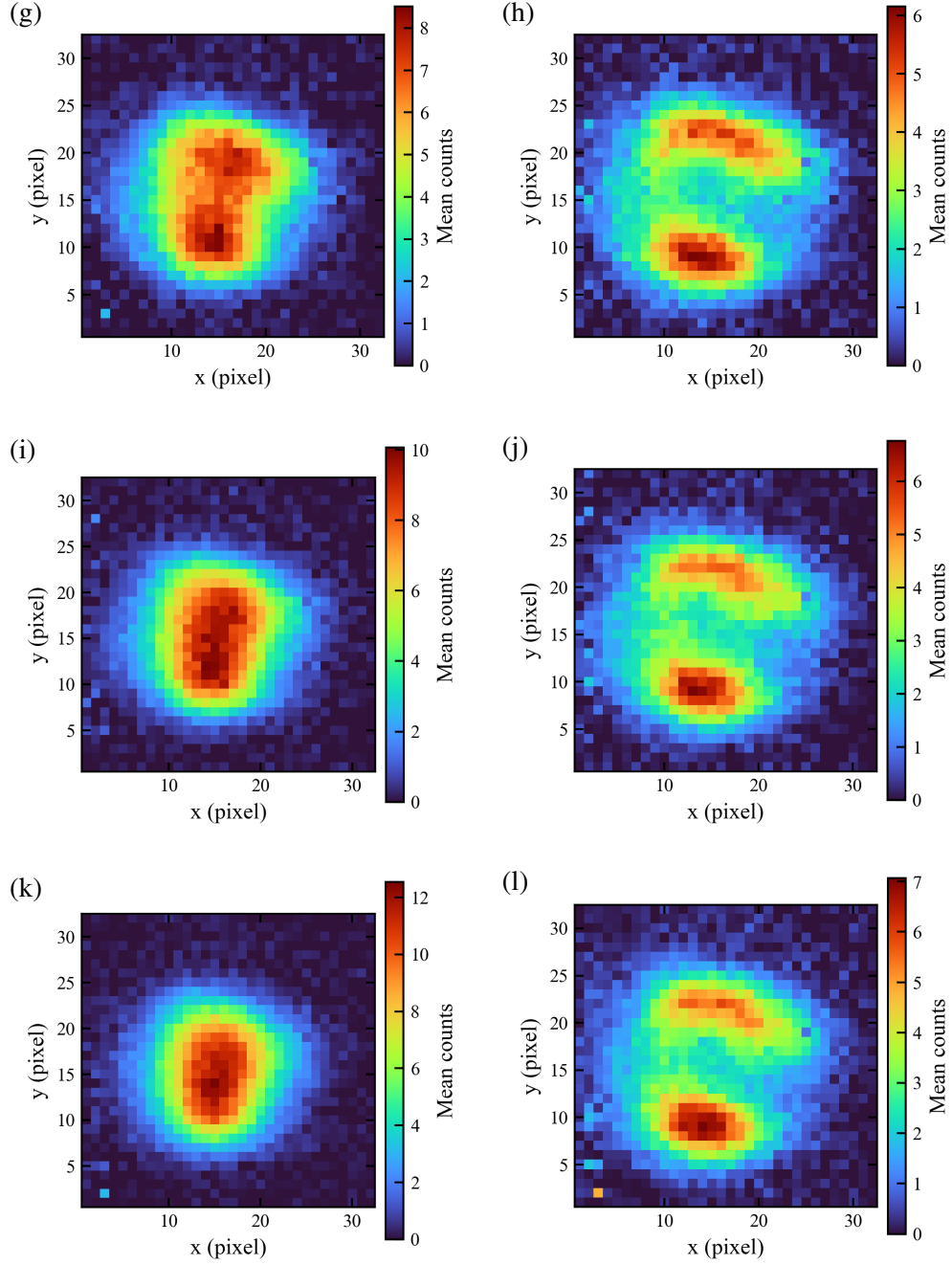


Figure A.4: EMCCD images at a probe detuning of 20 MHz for $OD \approx 65$. The images compare measurements with (a,c,e,g,i,k) and without (b,d,f,h,j,l) a linear increase in the dipole-trap power throughout the pulse sequence. The lower limit of the color scale was set to zero to simplify visual comparison. (a,b) All 450 pulses. (c,d) Pulses 1–90. (e,f) Pulses 91–180. (g,h) Pulses 181–270. (i,j) Pulses 271–360. (k,l) Pulses 361–450.

Acknowledgements

First of all, I want to thank Sebastian Hofferberth for giving me the opportunity to do my master's thesis in his group and Prof. Wang for agreeing to be my second appraiser.

I want to thank Daniil for giving me the opportunity to work together with him in the Rubidium experiment and for teaching me a lot about operating the experiment, EMCCD cameras, Rydberg polaritons and much more. I really enjoyed our numerous scientific discussions and working with you in general.

I also want to thank all other RQO team members. I want to thank Benedikt for his help with all kinds of questions. I especially enjoyed our after-work chess battles. I also want to thank the other students in the RQO team, Philipp, Enrico, and Max, who were fun to work with, as well as Wolfgang for his helpful suggestions whenever problems came up.

Special thanks go to Lukas and Nina, with whom I only worked for a shorter period during my master's thesis, but who were very helpful during both my bachelor's and master's theses and taught me a lot about quantum optics and beyond.

Also thanks to Fabian Maucher for the interesting discussions about the theory of the mean-field measurements.

I want to thank Daniil, Benedikt, Sam, Riccardo, Julia, Lukas and Nina for proofreading parts of my thesis.

I want to thank the whole group for the nice working atmosphere. Lastly, I want to thank my family.