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High-resolution spectroscopy of Rydberg states in an ultracold Cesium gas

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Abstract

Transitions between high Rydberg states of Cs atoms have been studied by high-resolution millimeter-wave spectroscopy of an ultracold sample. The spectroscopic measurements were performed after loading the atoms into a magneto-optical trap. Switching off all trapping fields and compensating the stray electric and magnetic fields to below 1 mV/cm and 2 mG, respectively, prior to the spectroscopic measurement enabled the recording of millimeter-wave spectra of Rydberg states with principal quantum number beyond $n = 100$ under conditions where the inhomogeneous broadening by stray fields is minimal and no dephasing of the Rydberg-atom sample can be detected over measurement times up to $60 \mu s$. The Fourier-transform-limited linewidths of better than 20 kHz enabled the observation of the hyperfine structure of $nS$ and $nP$ Rydberg states of Cs beyond $n = 90$. The analysis of the lineshapes of transitions to Rydberg states with $n \approx 230$ indicated that field inhomogeneities across the atomic sample represent the dominant cause of spectral broadening at high $n$ values. The analysis also revealed that the initial polarization of the atomic sample ($F = 4$, $M_F = 4$) is preserved for several tens of microseconds, the depolarization being caused by slow precession along the magnetic stray fields.

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I. INTRODUCTION

This article describes a spectroscopic study of very high Rydberg states of Cs in a magneto-optical trap (MOT) under conditions where Doppler broadening and line broadening caused by the finite transit time of the atoms in the measurement region are negligible. The residual line broadening is exclusively caused by residual stray electric and magnetic fields and by transitions induced by blackbody radiation. Under these conditions, we demonstrate that it is possible to resolve Rydberg series, including their hyperfine structures, up to high values of the principal quantum number. It is also possible to quantify the effects of weak electric and magnetic fields on the spectral structures.

High Rydberg states are electronically highly excited states that can be regarded, in first approximation, as consisting of a positively charged ion core weakly interacting with a distant electron in an orbital of high principal quantum number \( n \). These states form series that can be described by Rydberg’s formula

\[
\nu_n = \frac{E_1^{\alpha^+}}{h} - \frac{c R_M}{(n - \delta)^2}.
\]

In Eq. (1), \( \nu_n \) represents the spectral position of the Rydberg states in frequency units, \( E_1^{\alpha^+} \) the convergence limit of the series, which represents the energy required to form an ion in quantum state \( \alpha^+ \), \( R_M \) the mass-dependent Rydberg constant (\( R_M = R_\infty M^+/(m_e + M^+) \); \( m_e \) and \( M^+ \) are the masses of the electron and ion core, respectively), \( c \) is the speed of light in vacuum and \( \delta \) is the quantum defect. Most physical properties of Rydberg states scale as integer powers of \( n \), e.g. their lifetime as \( n^3 \), their polarizability as \( n^7 \), the threshold field for field ionization as \( n^{-4} \), and the van der Waals interaction between a pair of Rydberg atoms as \( n^{11} \) [1, 2].

In recent years, high-resolution spectroscopic studies of high Rydberg states have been carried out with the following goals: (1) to obtain information on the electron–ion-core interaction and exploit this information to characterize atomic and molecular photoionization [3] and electron-ion recombination processes [4] that play an important role in astrophysics and plasma physics; (2) to obtain precise values of ionization energies and other related thermochemical quantities [5]; (3) to measure stray fields [6–8]; (4) to obtain information on the interactions between two Rydberg atoms [9, 10] or between a Rydberg atom and a ground state atom [11, 12] which can give rise to the formation of weakly bound molecules with unusual properties [13]; (5) to study many-body effects in dense, ultracold Rydberg...
gases [14, 15] and characterize excitation blockade mechanisms of interest in quantum information science [16, 17]; (6) to study the interaction between Rydberg atoms and molecules and surfaces [18–20]; and (7) to exploit the unusual properties of high Rydberg states in fundamental experiments in quantum optics [21, 22].

The weakly bound nature of the Rydberg electron and the resulting high sensitivity of high Rydberg states to their environment are simultaneously a curse and a blessing: A blessing, because it is the origin of most applications of high Rydberg states, and a curse because artefacts can easily mask the effects one desires to study. For instance, the presence of only a few ions in the measurement volume can alter the line shapes and line positions in spectra of the high Rydberg states [23], which in turn may be interpreted as arising from other effects. Several of the applications listed above rely on the generation and use of dense Rydberg atom samples at very low temperatures, usually alkali metal atoms in MOTs, which offer experimental conditions under which Doppler broadening and transit-time broadening are negligible. Line broadening, if not caused by the fine and hyperfine structure, arise in such samples either as a result of the Stark and Zeeman effects or as a result of interactions of the Rydberg atoms with neighboring atoms. The full interpretation of the high-resolution spectra necessitates to disentangle these sources of spectral broadening, a task that is best achieved in experiments carried out at densities sufficiently low that interparticle interactions play no role. The compensation of stray electric and magnetic fields in MOTs to a level where they do not affect the spectral properties of high Rydberg states is a challenging task. The present article demonstrates how electric and magnetic stray fields can be reduced to below 1 mV/cm and 2 mG, respectively, enabling measurements of the hyperfine structures of Cs Rydberg atoms at much higher \( n \) values than in earlier works.

The article is organized as follows: The experimental setup is described in Section II and Section III presents the experimental procedures used for Rydberg-state excitation and detection, and for stray-field compensation. The results of the measurements of the hyperfine structure of high-\( n \) Rydberg states are presented in Section IV and the article is concluded with a brief summary.
FIG. 1: a) Schematic overview of the laser setup for laser cooling of cesium. The frequency of the master-oscillator power-amplifier (MOPA) system (Toptica TA-Pro) is stabilized by a Pound-Drever-Hall (PDH) lock to a saturated-absorption line of atomic cesium. The frequency modulation of the laser light required for the PDH scheme is performed by a resonantly-driven electro-optical modulator (EOM). Repumping light is provided by an external-cavity diode laser (ECDL, Toptica DL-Pro), which is stabilized to a second saturated-absorption line of cesium atoms using a lock-in detection scheme. b) Overview over the relevant transitions coupling the hyperfine components $F$ of $6S_{1/2}$ and $F'$ of $6P_{3/2}$: (A) light for absorption imaging, (C) light for laser cooling, (R) light for repumping.

II. EXPERIMENTAL SETUP

The central element of the experimental setup is a MOT for cesium atoms. The optical layout is shown schematically in Fig. 1a. The main source of narrow-band laser light at 852.3 nm is a master-oscillator power-amplifier (MOPA) system. Repumping light is provided by an additional external-cavity diode laser. Variable beamsplitters and acousto-optical modulators (AOMs) are used to generate intensity- and frequency-controlled light from these two lasers. The light for laser cooling is detuned by $\approx 2.8 \Gamma$ from the $6S_{1/2}, F = 4 \rightarrow 6P_{3/2}, F = 5$ transition and is split into three beams with roughly equal intensities which are coupled into three polarization-maintaining single-mode fibers. The repumping light is resonant with the $6S_{1/2}, F = 3 \rightarrow 6P_{3/2}, F = 4$ transition. It is spatially...
FIG. 2: a) Schematic top view of the setup. Pairs of fiber output couplers and retro reflectors deliver the cooling and repumping light (see text for details). UV light excites ground-state atoms into Rydberg states, and millimeter-wave radiation from a backward-wave oscillator (BWO) drives transitions between Rydberg states. Field plates (see Fig. 3) can be used to apply electric fields across the photoexcitation region. b) Schematic side view of the setup showing how the atom cloud is imaged onto a charge-coupled-device (CCD) chip by a matched pair of achromatic lenses.

The magnetic-field gradient for the MOT is produced by a pair of coils in anti-Helmholtz configuration. Each coil consists of 35 windings of copper band (width 15 mm, thickness 0.45 mm) with a mean radius of 53 mm. The coils are mounted at a distance of 65 mm from each other outside of the vacuum chamber in re-entrant view ports. To produce gradients on the order of 10 G/cm, a current of 8.0 A is passed through the coils.

Cesium atoms are provided by high-purity resistively-heated dispensers (Alvasource AS-
3-Cs-100-F) placed at a distance of roughly 15 cm from the MOT. The MOT can also be loaded from cesium vapor produced by light-induced atomic desorption (LIAD) [24] of cesium atoms from the walls and view ports of the chamber. The UV light for the LIAD process is produced by three high-power light-emitting diodes (LEDs, Edixeon EDEV-SLC1-R), each emitting approximately 580 mW of light at a central wavelength of 402 nm. The LEDs are installed outside of the vacuum chamber and the light enters the chamber through a fused-silica window. This loading scheme leads to a significant increase in the number of trapped atoms.

A. Absorption imaging

We determine the number, the density, and the temperature of the trapped atoms by absorption imaging of the atom cloud. The radiation for absorption imaging is provided by the master laser and can be independently modulated in frequency and intensity by an AOM. It is overlapped with the cooling laser beam in the vertical direction, and the two light fields are coupled into the fiber with orthogonal polarization. In front of the chamber, these polarizations are converted into left- and right-handed circular polarizations by a quarter-wave plate (see Fig. 2b for a schematic drawing). After the chamber, a combination of a quarter-wave plate and a polarizing-beamsplitter cube is used to separate the two frequency components based on their state of polarization. The plane containing the trapped atoms is imaged by a pair of identical achromatic lenses onto a charge-coupled-device (CCD) chip. The scale of the acquired image was calibrated before the final assembly by imaging a ruler at the position of the MOT. In order to obtain an absorption image, the cooling light is switched off using an AOM and an exposure of the CCD chip is triggered. Then, the absorption light is switched on for about 100 µs and the image of the atom cloud is read out digitally and processed by software. All timing sequences for controlling the MOT are generated by a DAQ-Card (National Instruments PCI-6259) which is programmed using LabView. The atomic samples used for the spectroscopic measurements described in this article had a typical peak density of $5 \times 10^9$ atoms/cm$^3$, a $1/e$ sample diameter of 150 µm, and a temperature of 10 µK, as determined from the analysis of the absorption images.
FIG. 3: (color online) Schematic description of the three potential configurations for the eight electrodes generating homogeneous electric fields at the position of the MOT (blue circle). Electrodes with positive potentials are presented in white and electrodes with a negative potential in black. The arrows indicate the direction of the electric field vectors. The configuration on the left-hand side generates an electric field in $z$ direction, the configuration in the middle generates a field in $x$ direction and the configuration on the right-hand side generates a field in $y$ direction.

B. Electrode geometry

The electrode structure for the manipulation and detection of Rydberg atoms consists of two segmented, ring-shaped electrodes (called “field plates” in the following) spaced by 40 mm and an ion lens for imaging the Cs$^+$ ions onto a multichannel-plate (MCP) detector. The shape and positions of the elements are indicated in Fig. 2 and were designed for the creation of homogeneous fields at the position of the MOT. Each field plate consists of four electrodes. These electrodes can be used to apply compensation, polarization, or ionization and ion-extraction fields. For the generation of compensation and polarization fields, voltages of a few volts are generated by a computer-controlled DAQ-Card (Measurement Computing USB-3105) and are, after analog filtering and down-scaling by a 1:6 voltage divider, directly applied to the electrodes. The potential configurations used to apply compensation electric fields along the $z$, $x$ and $y$ directions are schematically depicted in Fig. 3a, b and c, respectively.

Rydberg atoms can be ionized by applying positive high-voltage pulses either to the four field plates on the opposite site of the MCP (see Figs. 2 and 3c) or to the ring electrodes in front of the MCP. In the former case, four home-built high-voltage switches are used to switch from a potential configuration for the compensation of stray electric fields to a
potential configuration for the ionization of Rydberg atoms. Applying the high-voltage pulses to the ring electrodes enables one to avoid applying pulsed voltages to the field plates, which reduces transient stray fields in the region where the atom cloud is located. The resulting ionization fields are smaller and only suitable to detect Rydberg states with $n > 50$. Consequently, we apply the ionization-voltage pulses to the ring electrodes to detect Rydberg states with $n > 50$ and to the field plates for the detection of Rydberg states in the range $50 > n > 30$. Electric fields of sufficient strength to field ionize Rydberg states below $n \approx 30$ cannot be generated.

C. Magnetic coils

Three pairs of coils have been installed around the chamber to compensate external magnetic fields or to apply well-defined magnetic fields to the atomic sample. The compensation field in $z$ direction is produced by a pair of coils in Helmholtz configuration. The coils are wound around the gradient coils of the MOT and consist of 13 copper-band windings with a radius of 67.5 mm located at a distance of 33 mm from the center of the MOT. In the $y$ direction, the coils are mounted on flanges of the vacuum chamber. The coils consist of 64 windings of wire (ribbon cable) with a radius of 57 mm located at a distance of 63 mm from the center of the MOT. Because of the geometry of the chamber, a pair of asymmetric coils is used to apply magnetic fields in the $x$ direction. The first (second) coil consists of 27 windings (150 windings) with a mean radius of 57 mm (78 mm) located at a distance of 100 mm (195 mm) from the center of the MOT. The ratio of the windings of these two coils has been chosen so as to obtain a homogeneous magnetic field at the position of the atom cloud when the same current passes through both coils. The number of windings of all three pairs of coils has been adjusted to generate a magnetic field of about 1 G at the position of the MOT for a current of 1 A through each pair of coils. Under these conditions, a magnetic field variation of $\approx 0.5 \text{ mG}$ results over the 100 $\mu$m large atom cloud.

D. Rydberg spectroscopy

A pulsed frequency-doubled dye laser (DCM in ethanol, 0.05 cm$^{-1}$ bandwidth) is used to excite a fraction of the ultracold cesium atoms to a high Rydberg state. The dye laser is
pumped by a 10 Hz Nd:YAG laser operated at a wavelength of 532 nm and a pulse energy of $\approx 70$ mJ. After frequency doubling in a BBO crystal, the laser wavelength is centered around 320 nm and can be tuned to induce transitions from the $6S_{1/2}$ electronic ground state of cesium to the Rydberg series converging to the $^1S_0$ ground state of Cs$^+$. The pulsed UV radiation is linearly polarized along the $y$ axis and the pulse energy can be adjusted between 5 and 80 $\mu$J/pulse by using an attenuator consisting of a half-wave plate and a UV polarizer.

Spectra of transitions from the initially prepared Rydberg state to higher-lying Rydberg states were recorded with millimeter-wave radiation. As continuous millimeter-wave source, we used the phase-locked backward-wave oscillator (BWO) described in Ref. [25]. Its frequency can be continuously tuned in the range of 240 – 380 GHz and its bandwidth is narrower than 1 kHz [25]. The output horn of the BWO is placed 40 cm away from the center of the MOT and the millimeter-wave radiation intersects the UV-laser beam at right angles (cf. Fig. 2).

The output of the BWO is stabilized in a phase-lock loop to $f_{RF} + Nf_{LO}$, where $Nf_{LO}$ is the $N$th harmonic of the frequency of a local oscillator (Wiltron 6769B, discontinued product) and $f_{RF}$ is the frequency of an additional radio-frequency (RF) generator (Agilent 8647A, Palo Alto, USA). The output frequency is tuned by changing $f_{LO}$, which can be scanned in minimum frequency steps of 1 kHz. Because the 10th to 26th harmonics of $f_{LO}$ are used for the frequency stabilization, the minimum step size by which the output frequency can be varied is $10 - 26$ kHz, which is too large for the narrowest lines observed in the experiments. As an extension of the performance of this source as compared to the description in reference [25], a frequency step size below 1 kHz could be obtained by scanning the frequency $f_{RF}$ of the reference signal (350 MHz in normal operation) instead of $f_{LO}$. In this way, the frequency step size could be reduced to 1 Hz.

III. EXPERIMENTAL PROCEDURE

A. Photoexcitation

The experiment runs in a pulsed mode at a frequency of 10 Hz given by the repetition rate of the Nd:YAG laser used to pump the dye laser. After a MOT-loading phase of
≈ 92 ms, the magnetic fields in the MOT are turned off to enable spectroscopy of Rydberg states under field-free conditions. To avoid eddy currents influencing the measurements, the electric currents in the gradient coils are turned off 7.4 ms before application of the laser pulse used to prepare the Rydberg states. After a pseudo-molasses phase, the MOT lasers are turned off 400 µs (cooling laser) and 50 µs (repumping laser) before Rydberg excitation to avoid the generation of ions through direct photoionization of atoms in the 6P3/2 state by a UV photon. Because the cooling laser is turned off before the repumping laser, the cesium atoms are prepared in the F = 4 hyperfine component of the 6S1/2 state. To obtain a purely oriented sample of atoms in the MF = 4 state, the atomic sample is exposed to a beam of circularly polarized laser light propagating along the z direction and resonant with the 6P3/2, F = 5 ← 6S1/2, F = 4 transition for 100 µs. This laser is then turned off 20 µs prior to Rydberg excitation.

Because of a Cooper minimum in the photoexcitation cross section from the 6S1/2 state to the nP (eP) states (continuum) of cesium just above the ionization threshold [26], nF1/2 states with n > 40 are very difficult to access by one-photon excitation from the electronic ground state. The UV laser pulse energy is adjusted so as to only excite a small fraction, typically less than 10%, of the cesium atoms to a selected nF3/2 Rydberg state with n in the range 30 < n < 150. Under these conditions, the number of atoms excited to Rydberg states remains low and many-body interactions are negligible. For the measurement of millimeter-wave spectra, the UV laser frequency is kept fixed at the position of a selected nP3/2 ← 6S1/2, F = 4 transition. The laser linewidth of ≈ 0.05 cm⁻¹ enables the excitation of isolated Rydberg states up to n ≈ 140. All hyperfine transitions allowed by electric-dipole selection rules are excited.

The interaction time of the millimeter-wave radiation and the Rydberg states is determined by the delay time between the UV-laser pulse used for the initial excitation and the application of the electric-field pulse used for detection of the transitions. Because the UV pulse is short compared to this delay time and the electric-field pulse immediately shifts the transitions out of resonance, the effect of the millimeter-wave radiation inducing the Rydberg-Rydberg transitions can be described to a good approximation by a square pulse.

The maximal output power of the millimeter-wave source is on the order of tens of milliwatts [25]. The transition dipole matrix elements for transitions between Rydberg states scale as n² so that the output power had to be lowered to its minimum in order to
avoid saturation broadening of the investigated transitions. The intensity of the millimeter-wave radiation interacting with the atoms had to be further reduced by placing a stack of 500 sheets of paper between the source and the atom cloud. This measure effectively blocked the direct optical path, and the transitions were induced by reflected or scattered radiation. Under these conditions, the intensity was low enough to avoid saturation broadening which enabled us to fully exploit the narrow bandwidth of the millimeter-wave source. However, the polarization of the millimeter-wave radiation interacting with the Rydberg atoms was not well defined.

B. Detection of the Rydberg atoms

The Rydberg atoms were detected by monitoring the field-ionization signal induced by a slowly rising negative potential $U_{\text{ion}}$ applied to all ring electrodes and the grid in front of the MCP over a simple RC low-pass filter ($R = 8.2 \, \text{k}\Omega$, $C = 100 \, \text{pF}$). The resulting field ramp ionized all Rydberg states in the volume of the MOT and extracted the positively-charged $\text{Cs}^+$ ions towards the MCP detector. The time-of-flight traces were read out by an oscilloscope and processed further using a LabView program. $U_{\text{ion}}$ was adjusted to the $n$ values of the investigated Rydberg states such that initial and final Rydberg states appeared as separate signals in the time-of-flight traces. All timings relevant for Rydberg state excitation and detection were controlled by a high-precision delay generator (Quantum Composer 9528). To reduce the effects of shot-to-shot intensity and frequency fluctuations of the UV laser, the field-ionization signal of the final state of the transitions was normalized to the total ion yield. At low number densities of excited atoms, this ratio is independent of the absolute number of excited Rydberg atoms. To increase the signal-to-noise ratio, the signal of 80 experimental cycles was averaged at each spectral position.

C. Measurement and compensation of stray fields

Stray electric fields were compensated by applying potentials to the field plates described in Sect. II B (see Fig. 2 and Fig. 3). The voltages were applied to the electrodes such that the electric potential at the position of the MOT remains zero. By changing the polarity and the magnitude of these three potentials, any homogeneous electric field can be generated in
the central region of the electrodes.

The compensation fields were optimized by minimizing the Stark shifts of \( n'S_{1/2} \leftarrow nP_{3/2} \) transitions recorded with the millimeter-wave radiation. Because the states experience a quadratic Stark effect at small field strengths, the stray field can be compensated in each spatial dimension independently. First, a rough compensation of the stray field was performed at low \( n \) values. Then the procedure was repeated at progressively higher \( n \) values, exploiting the increasing sensitivity to stray fields resulting from the \( n^7 \) scaling of the polarizability [2]. In Fig. 4, a millimeter-wave spectrum of transitions from the \( 93P_{3/2} \) state to well-resolved Rydberg states with \( n \sim 230 \) is shown. At such high \( n \) values, the linewidths could not be reduced by varying the voltages applied to the compensation electrodes and was found to be independent of the ground- and Rydberg-state density. The linewidths increased, however, with increasing polarizabilities of the coupled states. Consequently, the line broadening is attributed to an electric field gradient originating from the residual stray field or from ions embedded in the atom cloud. Ions could result from collisions of Rydberg atoms, from blackbody-induced ionization, and from two-photon ionization of ground states atoms by combination of a UV photon and an IR photon (all IR beams are turned off by AOMs, but a small amount of light can leak through). From the experimental linewidth of \( \approx 2 \text{ MHz} \) for the \( 230S_{1/2} \leftarrow 93P_{3/2} \) transition and calculated Stark shifts of the Rydberg states involved, it can be excluded that the residual electric stray field varies by more than 1 mV/cm over the atomic sample. Thus, initial stray fields of about 250 mV/cm could be reduced to less than 1 mV/cm. The spectrum displayed in Fig. 4 illustrates the possibility to fully resolve Rydberg states of very high principal quantum numbers.

The magnetic stray field was compensated using the three pairs of magnetic coils described in Sect. II.C. In contrast to the quadratic Stark effect, the Rydberg atoms show a linear Zeeman effect in a magnetic field. Consequently, the components of the magnetic stray field along the different axes cannot be compensated independently, which complicates the stray field compensation procedure. We first exploited the mechanical Hanle effect [28] for a coarse compensation of the magnetic field in two directions. Then the remaining magnetic stray field was compensated by measuring the Zeeman effect in millimeter-wave spectra. In this way, initial magnetic stray fields of about 550 mG could be reduced to below 2 mG. This value was estimated by comparing spectra calculated for different magnetic field strengths to experimental millimeter-wave spectra (see next section) and is limited by the accuracy of
FIG. 4: Millimetre-wave spectrum of Rydberg states around 231S\(_{1/2}\) from the initial 93\(P_{3/2}\) state. The dashed vertical lines are calculated line positions based on the extrapolated quantum defects of Ref [27]. The appearance of the dipole-forbidden transitions \(nP_{1/2,3/2} \leftarrow 93P_{3/2}\) and the apparent line broadening are due to the residual electric fields.

1 mA with which the currents applied to the compensation coils can be adjusted.

IV. MEASUREMENT OF THE HYPERFINE STRUCTURE OF RYDBERG STATES

Stray electric and magnetic fields below 1 mV/cm and 2 mG, respectively, have almost no detectable effect on Rydberg states with \(n < 100\) at our resolution. Consequently, the hyperfine structure of high-lying Rydberg states could be investigated with high precision. Exemplary millimeter-wave spectra of \(n'S_{1/2} \leftarrow nP_{3/2}\) transitions are shown in Figure 5. The hyperfine splitting of the \(F = 3\) and \(F = 4\) components of the \(nS_{1/2}\) states is clearly resolved in these spectra for \(n\) up to 90. The two components are broadened by the hyperfine structure of the initial \(nP_{3/2}\), which is only partially resolved for \(n \lesssim 70\).

The experimental spectra could be modeled accurately by taking into account the hyperfine and the Zeeman interactions of initial and final states, and a finite Lorentzian line
FIG. 5: Experimental and simulated spectra of millimeter-wave transitions between \(nP_{3/2}\) and \(n'S_{1/2}\) Rydberg states of cesium (49\(S_{1/2} \leftarrow 45P_{3/2}\), 68\(S_{1/2} \leftarrow 59P_{3/2}\), 81\(S_{1/2} \leftarrow 67P_{3/2}\), 90\(S_{1/2} \leftarrow 72P_{3/2}\)). Each point represents an average over 80 laser shots. The error bars show the standard deviation of each measurement. The black line is a simulated spectrum that has been fitted according to the model described in the Appendix.

The model used for the calculation of spectra is described in detail in the Appendix. The adjustment of the calculated to the measured spectra allowed us to determine the spectral resolution, the hyperfine coupling constants for the involved states, and the initial polarization of the atomic sample. As an example, the experimental spectrum of the 68\(S_{1/2} \leftarrow 59P_{3/2}\) transition is compared to a spectrum calculated with the adjusted model and an assignment of the different spectral features in Fig. 6. From the adjusted model, we obtain a line profile with a full width at half maximum (FWHM) of 17(3) kHz. This is in good agreement with the Fourier transform limited linewidth of the 60 \(\mu\)s long millimeter-wave pulse (FWHM 17.2 kHz) and a lifetime-limited atomic linewidth of 2.5 kHz, taken from Ref. [29]. The experimental linewidth increases from 17(3) kHz to 19(3) kHz and 24(3) kHz for the millimeter-wave spectra of the transitions to the 81\(S_{1/2}\) and 90\(S_{1/2}\) states,
FIG. 6: Millimetre-wave spectrum of the $68S_{1/2} \leftarrow 59P_{3/2}$ including a fitted simulation (thick black line) and an assignment of the single hyperfine transitions (black arrows on dashed vertical lines). The inverted stick spectrum, on which the simulation is based, is presented in arbitrary intensity units and shows the splitting of the $F' \leftarrow F$ transitions into their $M_F$ components at the residual magnetic field of 2 mG.

respectively. This is in qualitative agreement with a Stark broadening caused by a residual electric field gradient of 100 mV/cm².

For each spectrum depicted in Fig. 5, the magnetic dipole hyperfine coupling constant $A_{\text{hfs, } P_{3/2}}$ of the $nP_{3/2}$ states was obtained by adjusting the model to the two hyperfine components of the $nS_{1/2}$ state separately. The magnetic dipole constant $A_{\text{hfs, } S_{1/2}}$ of the $nS_{1/2}$ state was then obtained in a second step by leaving $A_{\text{hfs, } P_{3/2}}$ unchanged and fitting the overall amplitude and frequency shift of the model to the complete experimental spectrum. The hyperfine coupling constants of these high Rydberg states can be compared to reported values measured at lower principal quantum number [27, 30–37]. The hyperfine structure results from the Fermi contact interaction of the Rydberg electron with the nucleus and is therefore proportional to the probability density of the Rydberg electron at the nucleus. It scales with $(n^*)^{-3}$, where $n^* = n - \delta_\ell$ is the effective principal quantum number and $\delta_\ell$ is the $\ell$-dependent quantum defect. One can thus introduce a reduced hyperfine coupling $A_{\text{hfs, } \ell}$ constant as $A_{\text{hfs, } n^*, \ell} = \frac{A_{\text{hfs, } \ell}}{n^*}$. The so-obtained reduced coupling constants were compared to previously published values [27, 30–37] in Fig. 7 (quantum defects are taken from [27]).
FIG. 7: Plot of the fitted magnetic dipole constants of the hyperfine structure of $nS_{1/2}$ (a) and $nP_{3/2}$ (b) Rydberg states of cesium atoms weighted by $(n^*)^3$ vs $n^*$ (our measurements are shown in black). The grey bar represents a weighted average including a 95% confidence interval of all four measurements and should guide the eye for the comparison with the literature data [27, 30–37] (shown in grey) at lower principal quantum number.

Because the hyperfine structure of the $nP_{3/2}$ states could not be resolved, the magnetic dipole coupling constants of the $nP_{3/2}$ states were only deduced from the line shape and linewidth and are therefore associated with larger uncertainties, whereas the hyperfine coupling constants of the $nS_{1/2}$ states could be determined with an accuracy comparable to, or exceeding the accuracy of the measurements at low $n^*$. The weighted averages of the reduced coupling parameters for both $nP_{3/2}$ and $nS_{1/2}$ states are $A_{\text{hfs},P_{3/2}}^* = 0.80(8)$ GHz and $A_{\text{hfs},S_{1/2}}^* = 13.53(12)$ GHz, respectively. For $A_{\text{hfs},P_{3/2}}^*$, we obtain systematically higher values than previously reported for lower $n$ values. The discrepancy could be the result of line-broadening effects by the residual electric field and the gradients of both electric and magnetic fields, which are not included in our model.

The modeled line shapes are sensitive to the initial population of $M_F$ states in the $6s^2S_{1/2}, F = 4$ ground state, i.e., to the orientation of the initial atomic sample before Rydberg excitation. Experimentally, we polarize the sample by applying a circular polarized pump laser pulse, which is switched off 20 µs before the excitation to Rydberg states (see Section III.A). In the model, the initial polarization was treated as a global parameter being constant for all spectra presented in Figs. 5, 6, and 8. The optimal agreement with the experimental spectra was obtained assuming that 95% of the atoms remain in the $M_F = 4$ component until excitation to Rydberg states. The timescale for depolarization is limited...
FIG. 8: Experimental and simulated spectra of millimeter-wave transitions from the $59P_{3/2}$ Rydberg state to the $66D_{3/2}$ and $66D_{5/2}$ states of cesium.

in our experiment by the precession of the magnetic moment of the cesium atoms around the axis of the stray magnetic field with the Larmor period $T_L = \left( \frac{\mu_B g_F B_{\text{res}}}{h} \right)^{-1}$, where $\mu_B$ is the Bohr magneton, and $g_F = 1/4$ the $g$-factor of the $6S_{1/2}, F = 4$ state. By taking a value of $B_{\text{res}} = 2 \text{ mG}$ for the residual magnetic field, we obtain $T_L = 1.4 \text{ ms}$, which explains why we do not observe a significant depolarization of the atomic sample during the $20 \mu s$ delay between optical pumping and Rydberg excitation.

We also recorded spectra of transitions from the $59P_{3/2}$ state to the $66D_{3/2}$ and $66D_{5/2}$ states, which are depicted in Fig. 8. The asymmetric line profiles result from the combined hyperfine structure of the initial and final states, which is not resolved. However, because we have independently determined the hyperfine coupling constant of the $59P_{3/2}$ state, we can extract the hyperfine coupling constants of $A_{\text{hfs},66D_{3/2}} = 2.6(5) \text{ kHz}$ and $A_{\text{hfs},66D_{5/2}} = 0.10(45) \text{ kHz}$ for the $D$ states.

V. CONCLUSION

A new experimental setup for high-resolution spectroscopy of cesium Rydberg states has been presented. Its performance was demonstrated by the measurement of the hyperfine structure of $nS_{1/2}$ states at $n = 45–90$ using millimeter-wave spectroscopy and the excitation of very-high-$n$ Rydberg states with $n = 230$ in the noninteracting regime (low ground- and Rydberg-state density). For $n < 100$, the hyperfine splitting of the $nS_{1/2}$ Rydberg states could be resolved in the measured millimeter-wave spectra. From the modelling of the
experimental spectra, magnetic dipole coupling constants for the hyperfine structure of the \(nS_{1/2}\) and \(nP_{3/2}\) states were obtained which are in agreement with measurements at lower \(n\) values. To achieve the required resolution, electric and magnetic stray fields of 250 mV/cm and 560 mG had to be compensated so that the residual fields were below 1 mV/cm and 2 mG, respectively. Our experiments exploited the fact that the factors that usually limit the resolution of spectroscopic measurements are negligible in our setup. Indeed, the Doppler broadening is well below 1 kHz in samples of ultracold atoms at low frequencies. Moreover, the low temperature of the atomic sample enables measurement times extending beyond 100 \(\mu s\). The control over the stray electric and magnetic fields demonstrated in this article was essential for the determination of the hyperfine structure of Rydberg states of Cs at high \(n\) values and to retain a full coherence and avoid a significant depolarization of the atomic sample over a time scale of up to 60 \(\mu s\).

**Appendix: Calculation of spectra**

The simulation of the experimental spectra requires the calculation of line positions and line intensities and a convolution with the instrumental line profile. In order to calculate the line positions, the energies of the hyperfine levels in a magnetic field of the initial and the final Rydberg state are obtained by diagonalizing the Hamiltonian matrix [38]

\[
\hat{H} = \hat{H}_{\text{hfs}} + \hat{H}_Z = A_{\text{hfs}}(\hat{I} \cdot \hat{J}) + g_J \mu_B B_z \hat{J}_z.
\]  

(A.1)

The Hamiltonian consists of the hyperfine-structure Hamiltonian \(\hat{H}_{\text{hfs}}\) in the magnetic dipole approximation and the Zeeman Hamiltonian \(\hat{H}_Z\) for the electron spin. Neglecting the Zeeman effect of the nuclear spin is justified because the electron \(g\)-factor is three orders of magnitude larger than the \(g\)-factor of the proton. Neglecting higher multipoles in \(\hat{H}_{\text{hfs}}\) is also justified, because the magnetic quadrupole constant of Cs is two orders of magnitude smaller than the magnetic dipole constant [39]. The Hamiltonian matrix is evaluated in the \(|F, M_F\rangle\) basis, in which \(\hat{H}_{\text{hfs}}\) is diagonal, with matrix elements

\[
\langle F | \hat{H}_{\text{hfs}} | F \rangle = \frac{1}{2} A_{\text{hfs}} [F(F + 1) - I(I + 1) - J(J + 1)].
\]  

(A.2)

The Zeeman Hamiltonian \(\hat{H}_Z\) is diagonal in the basis of \(|I, M_I, J, M_J\rangle\) and the matrix elements in this basis are given by

\[
\langle I, M_I, J, M_J | \hat{H}_Z | I, M_I, J, M_J \rangle = g_J \mu_B B_z M_J,
\]  

(A.3)
where the Landé $g$-factor was taken as $g_J = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)}$ [40]. The basis transformation from the $|I, M_I, J, M_J\rangle$ to the $|F, M_F\rangle$ basis was then applied to $\hat{H}_Z$, the resulting matrix was added to $\hat{H}_{hfs}$, and the total Hamiltonian matrix was numerically diagonalized. The transition frequencies were obtained as differences between the eigenenergies of the final and initial states.

For the calculation of the line intensities, the initial distribution of $M_F$ levels in the $F = 4$ hyperfine component of the $6S_{1/2}$ state of cesium and the $\Delta M_F$ selection rules for both Rydberg excitation and Rydberg-Rydberg transitions were taken into account. The UV laser is linearly polarized with its polarization axis perpendicular to the quantization axis of the atoms after optical pumping. Consequently, the UV laser drives $\Delta M_F = \pm 1$ transitions. The polarization of the millimeter-wave radiation interacting with the cesium atoms was not controlled in the experiments and the ratio of $\Delta M_F = \pm 1$ to $\Delta M_F = 0$ transitions has been determined empirically in a global analysis of all spectra. A ratio of 70% $\Delta M_F = 0$ and 30% $\Delta M_F = \pm 1$ yielded best overall agreement with the experimental results. This ratio was kept unchanged in the simulation of all spectra presented in Figs. 5, 6, and 8.

The transition strengths could then be evaluated using the spherical angular-momentum dipole-matrix elements

$$\langle FM_F|er_0|F'M'_F\rangle = \langle J||er||J'\rangle(-1)^{2F'+J+M_F+I}. \tag{A.4}$$

Because only the relative transition strength between the different $|F, M_F\rangle$ levels is observable in the experimental spectra, and the reduced matrix element $\langle J||er||J'\rangle$ gives a constant contribution for all transitions with given $\Delta l = 1$ and $\Delta J = 0,1$ values, this term was not evaluated in the calculation of the transition dipole moments. The line intensities were obtained as the product of the relative population in a given $M_F$ level of the $P_{3/2}$ Rydberg state and the spherical matrix elements for the transitions to the $S$ and $D$ states. For calculations including a nonzero magnetic field, the line intensities were further weighted by the admixture of initial and final states (in the unperturbed $|F, M_F\rangle$ basis) to the eigenstates of $\hat{H}$ in the magnetic field. The magnitude of the residual magnetic field was treated as global fit parameter. The best agreement between experimental spectra and simulation was obtained for a residual magnetic field of 2 mG.
The convolution of the Fourier transform of the square excitation pulse (a sinc function) and the Lorentzian line-profile of the atomic transition was found to be adequately described by a Lorentzian line-profile. Thus the stick spectra obtained with the calculated line positions and intensities were convoluted with this line-profile function. The overall amplitude and an arbitrary frequency shift of the resulting spectra were fitted separately to each experimental spectrum, as well as the magnetic dipole constants of the initial and final state and the Lorentzian linewidth.

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