Measurement of the electric dipole moments for transitions to rubidium Rydberg states via Autler–Townes splitting

M J Piotrowicz\textsuperscript{1}, C MacCormick\textsuperscript{1,3}, A Kowalczyk\textsuperscript{1}, S Bergamini\textsuperscript{1,3}, I I Beterov\textsuperscript{2} and E A Yakshina\textsuperscript{2}

\textsuperscript{1}Department of Physics and Astronomy, The Open University, Walton Hall, Milton Keynes, MK6 7AA, UK
\textsuperscript{2}Institute of Semiconductor Physics, Lavrentyeva Avenue 13, 630090 Novosibirsk, Russia
E-mail: c.maccormick@open.ac.uk and s.bergamini@open.ac.uk

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Abstract. We present the direct measurements of electric dipole moments for $5\text{P}_{3/2} \to n\text{D}_{5/2}$ transitions with $20 < n < 48$ for rubidium atoms. The measurements were carried out in an ultracold sample via observation of the Autler–Townes splitting in a three-level ladder scheme, commonly used for two-photon excitation of Rydberg states. To the best of our knowledge, this is the first systematic measurement of the electric dipole moments for transitions from low excited states of rubidium to Rydberg states. Due to its simplicity and versatility, this method can be easily extended to other transitions and other atomic species with few constraints. The good agreement seen between the experimental results and the theory proves the reliability of the measurement method.

\textsuperscript{3}Author to whom any correspondence should be addressed.
1. Introduction

The spectroscopic properties of the transitions between excited states of atoms and molecules are important in a wide variety of contexts, ranging from experiments with cold Rydberg atoms [1] to astrophysical studies and plasma physics [2]. Numerous theoretical models have been developed for the calculation of dipole moments, transition probabilities and photoionization cross sections for alkali-metal atoms. These models range from simple estimates based on the relative $n^{a(-3/2)}$ scaling of the dipole moments [3] to complicated relativistic methods [4]. The experimental verification of the different models is fundamental, but work in this direction has so far been lagging behind due to the non-triviality of the task. For alkali-metal atoms in particular, very few experimental measurements have been reported. The most recent measurements of the oscillator strengths for principal series of rubidium, potassium and cesium were reported in [5, 6]. Discrepancies were observed between experimental measurements and theoretical values and could be later explained [7] by introducing several corrections to the relativistic model-potential method. Measurements of the oscillator strength for lithium and sodium, reported in the recent papers [8, 9], highlighted once more significant discrepancies between experiment and theory. Certainly further experimental verification of the theoretical models is strongly needed. Recently, measurements of the effective lifetimes of Rydberg states [10, 11] were used for the indirect verification of the theory [12]. However, these measurements cannot reveal the spectroscopic features of each particular transition; therefore, they are less indicative of the model’s limitations.

It is therefore clear that further measurements of the dipole moments for transitions between excited states of alkali-metal atoms are necessary for the verification of theoretical models. In particular, measurements of the dipole moments for transitions between the first excited and Rydberg states, which are important in many experiments with ultracold Rydberg atoms, have never been made.

In this paper, we describe a simple and versatile method (similar to that used previously in [13] for molecular transitions) that allowed us to measure for the first time the dipole moments for Rb $5P_{3/2} \rightarrow nD_{5/2}$ transitions with $20 \leq n \leq 48$ with high accuracy and can be easily extended to different transitions or atomic species. This method is based on the spectroscopic observation of the Autler–Townes (AT) splitting in a three-level ladder scheme [14], which is also commonly used in electro-magnetically induced transparency (EIT) experiments with Rydberg atoms [15, 16].
Figure 1. Schematic representation of the three-level ladder scheme. $\Omega_c$ and $\Omega_p$ are the Rabi frequencies associated with the coupling laser and probe laser, respectively.

2. Method

We observe EIT and AT splitting [17] in a sample of ultracold Rb Rydberg atoms by the measurement of the absorption profile of the weak (probe) laser radiation scanned across the resonance $5S_{1/2} \rightarrow 5P_{3/2}$ in the presence of the stronger (coupling) laser radiation tuned to transition $5P_{3/2} \rightarrow nD_{5/2}$, following the scheme suggested in [14–16].

The long-lived states $|1\rangle$ and $|3\rangle$ are coupled to a short-lived state $|2\rangle$ by a weak probe laser and an intense coupling laser, as shown in figure 1, and we assume that the states $|1\rangle$ and $|3\rangle$ are not coupled by electric dipole transition. $\Omega_c$ and $\Omega_p$ are the Rabi frequencies associated with the coupling laser and probe laser, respectively.

With the atoms initially populating state $|1\rangle$ and the coupling laser tuned to the $|2\rangle$ to $|3\rangle$ resonance, EIT appears as a dip in the center of the absorption lineshape of the probe beam, which results from the destructive interference between excitation pathways and is observed by sweeping the probe laser through the $|1\rangle \rightarrow |2\rangle$ transition. With increasing coupling laser intensity the transparency dip becomes wider, until it eventually resembles two individual lines (AT splitting), resulting from the dynamic splitting of the middle energy level due to the strong coupling field.

To obtain an expression for the absorption lineshape of the probe beam, we solve the optical Bloch equations for the system of three atomic levels and two coupling lasers, taking the probe laser as a weak perturbation so that the population of the two states $|2\rangle$ and $|3\rangle$ are set to 0. This is ensured by keeping the probe laser intensity $I_p \ll I_{sat}$ for the transition to $|5P_{3/2}\rangle$. In this limit, only a negligible fraction of atoms will populate the Rydberg state, so that any effects arising from level shifts induced by the interaction due to the presence of Rydberg atoms are insignificant, as discussed in [16]. We find that the cross-section for absorption of the probe laser, $\sigma_p$ is then

$$\sigma_p = \sigma_0 \left[ \left( \Gamma + 2i\delta \right) + \frac{\Omega_c^2}{\gamma S + 2i(\delta + \Delta)} \right]^{-1} + \text{c.c.}. \tag{1}$$
where $\sigma_0$ is the cross-section for absorption in the absence of the coupling laser, $\Gamma$ is the width of the $|2\rangle$ state, $\delta$ is the detuning of the probe beam, $\Delta$ is the detuning of the coupling beam and $\gamma_3$ is the dephasing rate of the $|3\rangle$ state. In our experiment, state $|1\rangle$ corresponds to the $5S_{1/2} F = 2, m_F = 2$ state, $|2\rangle$ to $5P_{3/2} F = 3, m_F = 3$ and state $|3\rangle$ to one of the Rydberg states $nD_{5/2}, F = 4, m_F = 4$, where $n$ lies between 20 and 48. It has to be mentioned that other states (e.g. $nD_{3/2}$) can also be selected, by tuning the frequency and changing the polarization of coupling and probe beam. For the $5S_{1/2} F = 2 \rightarrow 5P_{3/2} F = 3$ transition, $\sigma_0 = 2.90 \times 10^{-13}$ m$^{-2}$.

By scanning the frequency of the probe beam around the $|1\rangle \rightarrow |2\rangle$ resonance, we obtain the spectrum $\sigma_p(\delta)$, from which we determine $\Omega_C$. By investigating the dependence of $\Omega_C$ on the power of the coupling beam for a variety of Rydberg states, we experimentally determine their transition dipole moments. We find that this is a highly reliable and precise method of determining dipole moments. As will be shown in the next section, the method allows for a strongly focused coupling beam to be used, such that $\Omega_C \rightarrow \Omega_C(x, y)$, provided that an accurate determination of the laser beam profile and power is performed.

3. Experimental details

We prepare an ensemble of cold atoms at a density of $6 \times 10^9$ cm$^{-3}$ and a temperature of 10 $\mu$K, by collecting $3 \times 10^6$ atoms from the background vapor in a magneto-optical trap and allowing further cooling in optical molasses. The sample is prepared in the $|5S_{1/2}, F = 2, m_F = 2\rangle$ magnetic sub-level by optical pumping, with an efficiency of 80%.

The atoms are subsequently illuminated by an intense coupling laser and a weak, counterpropagating probe laser, as shown in figure 2(a), which are $\sigma^+/\sigma^+$ polarized. The coupling laser is provided by a frequency-doubled diode laser (TOPTICA SHG), which is frequency locked to the center of the $5P_{3/2} \rightarrow nD_J$ transition at around 480 nm using the scheme based on EIT described in [18]. The beam is focused onto the atom cloud to a small waist in...
order to maximize its signature on the atomic sample. The weak ‘probe’ beam is provided by a commercial diode laser frequency stabilized, which is imaged onto the cold atoms cloud by a singlet lens (L1) focusing to a waist of 364 ± 10 μm. Its frequency can be swept around the $5S_{1/2} \rightarrow 5P_{3/2}$ transition at 780.24 nm. After passing through the atoms, a system of two lenses formed an image of the probe beam on either a CCD camera or a photodiode, as shown in figure 2(a). We are able to address $nD_{5/2}$ and $nD_{3/2}$ states by tuning the laser appropriately, since the <1 MHz linewidth of our lasers is much smaller than the fine-structure splitting for the range of $n$ in this study. However, we cannot resolve the hyperfine structure of these states. Thus, in the general case, we do not have a true three-level system. By choosing both laser beams to have $σ^+$ polarization, we drive transitions such that $Δm_F = 1$, according to the scheme in figure 2(b). As the atoms initially populate the $|5S_{1/2}, F = 2, m_F = 2\rangle$, the choice of polarization of the probe selects the $|5P_{3/2}, F = 3, m_F = 3\rangle$ state. For the $nD_{5/2}$ state this scheme selects only the $F = 4, m_F = 4$ sub-level.

The atoms are illuminated for 1 ms by switching on both the 480 nm coupling and the 780 nm probe laser. During this time the probe laser frequency is swept across the $5S_{1/2}F = 2$ to $5P_{3/2}F = 3$ transition, while a 200 mG magnetic field is kept on in order to preserve the projection of the atoms’ magnetic moment. Finally, we record the total laser power for each realization of the experiment on a calibrated photodiode, accurate to better than 5%, also accounting for the response of the photodiode due to wavelength variation. We correct for the variations of the probe laser intensity while the frequency is swept. In this way, we determine the probe absorption cross section as a function of its frequency using Beer’s law.

It must be pointed out that the Rabi frequency $Ω_C$ that appears in equation (1) is, in general, a function of position $Ω_C \to Ω_C(x, y)$. In many experiments, a homogeneous Rabi frequency across the sample is necessary and this requires proper shaping of the coupling laser beam. The analysis method that we employ allows us to avoid the reshaping of the beam, thus enabling us to maximize the intensities involved by focusing the laser beams.

Our method, therefore, does not allow us to resolve the distribution $Ω_C(x, y)$ of the coupling Rabi frequency due to the Gaussian intensity profile. Instead it provides an integrated signal and an ‘average’ cross section $σ_p(δ)$ across the spatial profile. Taking this into account in the analysis of the spectra is non-trivial, as an analytic solution to equation (1) cannot be found. Thus, the signal is numerically evaluated by considering the contributions from areas of equal intensity and integrating over the beam profile (see below). By taking into account the spatial profile $E(x, y)$ and therefore $Ω_C(x, y)$, we extract from each scan a value for $Ω_C^{\text{max}}$ that corresponds to the peak value of $E(x, y)$, i.e. the amplitude of the Gaussian distribution. Figure 3 shows how the modeling of the lineshape well adapts to the measured spectra for $n = 22$ and $n = 44$.

For each chosen Rydberg state $nD_{3/2}$, we take scans at different coupling powers, while the probe laser is kept at very low intensity (typically $0.01 I_{\text{sat}}$) to minimize the probability of populating the $5P_{3/2}$ state, as mentioned earlier. This allows us to build a linear trend for each $n$ of $Ω_C^{\text{max}}$ versus $\sqrt{P}$. From the measurements we can therefore extract a value for the transition dipole moment for each $n$, as $Ω_C \propto μ \sqrt{P}$.

4. Analysis

We have analyzed the probe absorption lineshapes to determine the Rabi frequency $Ω_C^{\text{max}}$ of the upper transition. For an accurate analysis, it is fundamental for us to account for the
inhomogeneity of the intensity profile of the coupling laser beam. A useful feature of our setup is that we may take absorption images of the atoms in the presence of the coupling beam, which allows for an accurate determination of the blue laser waist at the position of the atoms. The atomic cloud casts a shadow on the probe beam that is imaged on the camera. The presence of the coupling beam affects the optical depth of the atomic cloud, so that the cloud becomes transparent to the probe beam. By imaging the center of the atomic cloud, the optical depth of the atoms in the region illuminated by the coupling beam should reflect its intensity profile. Figure 4 shows a typical variation in the intensity of the light transmitted through the sample. This allowed us to accurately determine the elliptical coupling laser waists as \( w_{\text{maj}} = (240 \pm 10) \, \mu m \) and \( w_{\text{min}} = (172 \pm 10) \, \mu m \), along the major and minor axes of the beam cross section. These measurements are mainly limited by the resolution and signal-to-noise ratio of our imaging system. To make sure that the measurements were not biased, we repeated the measurements for different coupling laser powers (80 and 50 mW) and by tuning to different \( n \) states \((n = 26, 40, 44)\). We derived consistent waists in all cases, thus confirming the effectiveness of the method.

Using this determination of the laser beam profile and the measured total power of the beam, we deduced the electric field amplitude \( E(x, y) \) of the coupling laser as a 2D Gaussian distribution of amplitude \( E_{\text{max}} \) and waists \( w_{\text{maj}} \) and \( w_{\text{min}} \), which is then taken into account via numerical methods in the analysis of the spectra.

In order to obtain good AT spectra, we were obliged to work with a very low probe intensity, and consequently we compromised the bandwidth of the photodiode amplifier to 35 kHz in exchange for a higher signal-to-noise ratio. As a result, our spectra are instrumentally broadened and we are unable to observe the narrow EIT features as seen in [16]. We quantified the broadening by analyzing the spectrum obtained when the coupling laser is absent—i.e. by observing the two-level \(|5S_{1/2}, F = 2 \rangle \rightarrow |5P_{3/2}, F = 3 \rangle \) transition. We found that the linewidth \( \Gamma \) is 9 MHz, larger than the value of 6.065 MHz obtained in high-precision measurements. To explain the observed broadening, we modeled the experimental signal by convolving the ideal spectrum with the response of a passive low-pass filter with a corner frequency of 35 kHz. We found that the photodiode bandwidth, combined with the measured laser linewidth of 450 kHz,
Figure 4. Snapshot of the rubidium atoms absorption imaged using the probe and the counterpropagating coupling laser. The hole in the center of the cloud is due to the reduction in cross section for the probe laser where sufficient coupling intensity exists to produce EIT. An accurate measurement of the probe laser waist is obtained by fitting the expected 2D profile to the observed image.

was sufficient to explain the signal broadening. This instrumental limitation determines the width of the EIT feature that we can observe, and limits our measurements of $\gamma$ to 2.5 MHz, much larger than the values expected from the lifetime of any of the Rydberg states used. In order to check that the instrumental broadening did not alter the value of $\Omega_C$ obtained from fitting the lineshape, we studied the effect of the filtering on the AT signal (taking into account the inhomogeneity of the coupling laser). We found that our fitting procedure yielded the correct $\Omega_C^{\text{max}}$ to a high accuracy (better than 1%).

We analyzed the spectra as follows. In general, the photodiode signal is given by Beer’s law as the integral over the atomic cloud:

$$S = I_0 \int \mathrm{d}x \, \mathrm{d}y \, \mathrm{d}z \exp(-\sigma_p(x, y)n(x, y, z)),$$

where the cross section $\sigma_p(x, y)$ (equation (1)) is a function of $x$ and $y$ due to the inhomogeneity of the coupling laser and the atomic density $n(x, y, z)$. We approximate this integral as a sum using as input the measured waist sizes of the coupling beam and the measured cloud dimensions. The spectra are fitted to this sum and we obtain the parameters $\Omega_C^{\text{max}}$, $\gamma$ and $\Delta$, having already determined $\Gamma$ as above. Figure 4 shows examples of the data and the fitted lineshapes.

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Figure 5. The dependence of the Rabi frequency $\Omega$ on the square root of the coupling power $\sqrt{P}$ is used to check if the inhomogeneity of the coupling laser has been correctly accounted for. Error bars represent the uncertainty in the value of $\Omega$ obtained from the fit.

We repeated the measurements for each $n$ state at different coupling laser powers, ranging from 5 to 80 mW. The results obtained for $n = 22$ and $n = 44$ are plotted in figure 5 together with their uncertainty, which is given from the statistics of the fitting procedure, and show a good linear trend. The Rabi frequency is, in fact, directly linked to the square root of the laser power, according to $\Omega_n = \mu E / h \sqrt{\pi w^2 c \epsilon_0} \sqrt{P}$, where $E$ is the amplitude of the electric field in a Gaussian beam of power $P$ and waist $w$. In our case, because of the ellipticity of the beam, $w^2 = w_{\text{maj}} \times w_{\text{min}}$.

The dipole moments for each transition $|5P_{3/2}, F = 3, m_F = 3\rangle \rightarrow |nD_{5/2}, F = 4, m_F = 4\rangle$ (with $n = 22$–44) can therefore be obtained directly from the gradients of the linear trends in figure 5, provided an accurate measurement of $w$ and $P$ is made. From the linear fits, we obtained the values for the gradients with an uncertainty of less than 2% in most cases, with a maximum of 4% for $n = 44$. However, our uncertainties on the measurements of the waist and of the laser power were also to be taken into account when determining the values of the dipole moment $\mu_n$ for the transitions involved, and the total uncertainty on the measured dipole moments is less than 10% for all $n$.

In figure 6, our results are plotted after rescaling as reduced matrix elements, to be representative of the $|5P_{3/2}\rangle \rightarrow |nD_{5/2}\rangle$ transition, by taking into account the three-$J$ and six-$J$ symbols to obtain an angular coefficient of $\sqrt{2/3}$. The plot shows the reduced matrix elements and their uncertainties versus $n$-number.

To demonstrate the validity of our measurement method, we compared the experimental results to theoretical models. Although the alkali-metal atoms have a single valence electron, only the states with small quantum defects exhibit truly hydrogen-like behavior. The oscillator strengths for alkali-metal atoms can strongly deviate from the values for hydrogen. Accurate calculation of the radial integral for transitions between states with a small angular momentum is challenging because the interaction of the valence electron with the atomic core is difficult.
Figure 6. Our results, summarized as a graph of measured reduced matrix elements $\mu$ versus principal quantum number $n$, and comparison to theoretical models.

to be taken into account. *Ab initio* relativistic calculations with corrections for core polarization have been performed only for alkali-metal principal series [7]. It was found that the values of the oscillator strengths for the principal series in heavy alkali-metal atoms can be altered by an order of magnitude due to the polarization of the atomic core. For transitions between excited states, good agreement between experiment and theory is observed in many cases even when approximate methods are used. The Coulomb approximation is based on the idea that the Rydberg electron is localized mostly outside the atomic core, where the potential is Coulombic.

In the numeric Coulomb approximation (NCA) [19], the radial wavefunctions are obtained by numeric solution of the Schrodinger equation with the exact energies of the alkali-metal quantum states, the quantum defects are used as an input parameter for the calculations and the integration is truncated at the inner core radius. Alternative forms of the Coulomb approximation were developed in [20]. The modified Coulomb approximation (MCA) [20] is a generalization of the analytical expression for the hydrogen radial integral for non-integer quantum numbers. It allows direct calculation of the radial matrix elements without numeric integration. Further simplification of the calculations in the Coulomb approximation was achieved by extension of the quasiclassical approximation to the states with low principal quantum numbers (the Kaulakys or Dyachkov–Pankratov (DP) model). The other set of methods is based on the use of the realistic model potentials (the method of model potential (MMP). As well as Coulomb approximation, the potential outside the atomic core is nearly Coulomb-like, while the change of the potential within the atomic core and corrections for core polarizability and spin–orbit interaction can be taken into account [21]. A comparison of the calculated reduced dipole matrix elements [22] with experiment is shown in figure 6. We calculated the dipole moments using non-relativistic DP, NCA, MCA and MMP methods, described above, using the quoted values of quantum defects for rubidium from [24], the energy of the 5P_{3/2} state from [23] and the model potential of Marinescu [21], which takes into account the electric charge of the atomic core and the effect of the induced core electron moments on the valance electron, with corrections for spin–orbit interaction. For MMP calculations the RADIAL program, discussed in [24], was used. The first-order correction for effects of the core polarizability can be done by correcting
the dipole moment operator [25]. Our model potential calculations have been performed without (MMP(a)) and with (MMP(b)) correction for core polarization. We used a ‘chi-squared’ statistical test to assess the agreement between the theoretical models and the experiment. Agreement between experiment and theory is observed for DP, MCA and NCA models, for which the $P$-value is 0.97, 0.92 and 0.83, respectively. While MMP calculations performed only taking into account static dipole polarizability of the positive-ion core (MMP(a)) give a $P$-value of 0.28, substantial improvement can be obtained by including core polarization correction for the transition dipole moment operator (MMP(b)), in which case we obtain a $P$-value of 0.48. The good agreement between experiment and theory shows that non-relativistic models can be used for the calculation of the dipole moments and oscillator strengths for transitions involving low-excited and Rydberg states of alkali-metal atoms. New systematic measurements of the dipole matrix elements for other series of alkali-metal atoms will be of great value for further verification of the theory.

5. Outlook

We made direct measurements of the dipole moments for transitions from the $5P_{3/2}$ state to the high Rydberg state $nD_{J}$ for $^{87}$Rb using EIT/AT in a three-level ladder excitation scheme. These are, to our knowledge, the first direct measurements of the alkali-atom $5P_{3/2} \rightarrow nD_{5/2}$ Rydberg state dipole moments using this method. Good agreement between experiment and theory was observed, and the measurements could be extended to different atomic levels and species, so that this method could prove invaluable for testing current theoretical models that have limited accuracy.

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