Trap-loss spectroscopy of Rydberg states in ytterbium

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Abstract

We present an experimental study of the Rydberg $^1S_0$- and $^1P_1$- series of ytterbium for principal quantum numbers in the range of $n = 70–90$. The study is performed using trap loss spectroscopy in a magneto-optical trap operating on the $6^1S_0 \rightarrow 6^1P_1$ transition at 399 nm. Compared to the commonly used Rydberg spectroscopy method using field-ionization and ion detection, trap loss spectroscopy is significantly simpler and requires a less sophisticated experimental setup. Using this method we determine relative values of the scalar and tensor electric polarizabilities of both, $6sns^1S_0$- and $6snp^1P_1$- Rydberg states.

Keywords: Rydberg, ytterbium, spectroscopy

1. Introduction

Due to their special properties, Rydberg atoms have attracted the attention of experimental atomic physicists for a long time [1]. In recent years the possibility to produce Rydberg atoms with kinetic temperatures in the mK - or even µK - regime has opened up fascinating new possibilities for the precise control of Rydberg atoms, in particular to exploit the long-range dipole-dipole interaction [2]. This strong interaction may be used to observe a dipole blockade which can lead to a highly-correlated state in a mesoscopic atom cloud [3] or to entangle individual atoms [4, 5] offering the possibility to use Rydberg atoms for quantum information processing [6].

In the majority of experiments with ultracold Rydberg atoms, alkaline atoms such as rubidium have been used, where only one electron can be easily excited to a Rydberg state. For alkaline earth atoms or other atoms with two-electrons in the outermost shell, such as ytterbium (Yb), new possibilities arise since the atomic core remains optically active if only one electron is excited to a Rydberg state [7]. For example, for Rydberg states with low angular momentum so-called isolated-core excitation of the second electron can lead to auto-ionization of the Rydberg electron. This phenomenon can be used to control Rydberg excitations [8, 9] or directly image Rydberg atoms without the need to apply high-voltage pulses [10, 11]. Another attractive application is trapping of Rydberg atoms [12]. Very recently, several research groups have started to exploit the particular properties of two-electron Rydberg atoms for applications in quantum technology. The most promising platform consists of tweezer arrays in which atoms can be trapped at single sites and manipulated using transitions to Rydberg states as has recently been demonstrated for strontium [13, 14] and Yb [15, 16].

For all applications using Rydberg atoms, a precise knowledge of the level structure and the specific properties of the Rydberg states is essential. For Yb, several low-resolution studies have been performed in hot atomic samples [17–20]. Here we present a precise measurement of the binding energies and polarizabilities of the most abundant isotope $^{174}$Yb using simple and efficient trap-loss spectroscopy in a magneto-optical trap (MOT). In particular, we determine the binding energies of the $6sns^1S_0$- and $6snp^1P_0$- Rydberg series.
for principal quantum numbers ranging from from \( n = 70 \) to \( n = 90 \) and measure the relative polarizabilities. Our measurements complement recent results on Yb Rydberg energy levels obtained in significantly more complicated setups \cite{16, 21}, which make use of complicated electrode setups, to ionize the Rydberg atoms, micro-channel-plates for the ion detection and fast counting units for the evaluation. The study presented in this manuscript is a first step towards experiments in which we plan to investigate the special properties of Yb Rydberg atoms with a particular focus on the optical trapping of Yb Rydberg atoms.

2. Experimental setup

In our experiment, trap-loss spectroscopy is performed in a standard six-beam magneto optical trap using the isotope \(^{174}\)Yb. The MOT operates on the broad \( 6s^2 \, ^1S_0 \rightarrow 6s6p \, ^1P_1 \) transition at 399 nm which is continuously loaded from a Zeeman slower. The laser light for the MOT and the Zeeman slower is provided by a home-built diode laser system in master/slave configuration (Nichia NDHV310ACAEI/Nichia NDV4313E) (see figure 1(b)). The master laser is frequency-stabilized to a Doppler-free Yb spectroscopy. For precise control of the laser frequencies acousto-optical modulators are used.

Rydberg atoms can be produced in the MOT by a two photon excitation scheme, as illustrated in figure 1(a). The first photon with a wavelength of \( \lambda_{\text{MOT}} \approx 399 \) nm corresponds to the \( 6s^2 \, ^1S_0 \rightarrow 6s6p \, ^1P_1 \) MOT transition and is provided by the MOT laser. To transfer the atoms from the \( 6s6p \, ^1P_1 \) - state into a Rydberg state of the \( 6s_{\text{nS}}^1S_0 \) - or the \( 6snp \, ^1P_1 \) - series, the MOT is illuminated with an additional so-called Rydberg laser with a wavelength of \( \lambda_R \approx 394 \) nm. The nominally forbidden transitions \( 6s6p \, ^1P_1 \rightarrow 6snp \, ^1P_1 \) can be excited, if an external electric field is applied which results in an admixing of even-parity states to the odd-parity \( 6snp \, ^1P_1 \) - states. The Rydberg laser light is generated using a commercial diode laser system (Toptica DLpro) which is frequency doubled to \( \lambda_n \), using a Lithium Triborate (LBO) crystal integrated in a home-built bow-tie resonator. The output of the doubling resonator is delivered to the experiment using an optical fiber and is focused onto the MOT with a beam waist of \( \rho_{\text{max}} \approx 10^9 \) cm\(^{-3}\). A relatively homogeneous electric field at the MOT position can be created by simply placing an electrode on the top viewport of the grounded vacuum chamber at a distance of \( 75 \) mm from the MOT position. The electrode is a copper disk with a diameter of \( 150 \) mm and a \( 25 \) mm bore in the center which is used for the vertical MOT beam. Typically, a voltage of \( 2 \) V is applied to the electrode leading to electric fields up to a few \( 10 \) V m\(^{-1}\) at the position of the MOT.

3. Measurement procedure

A cycle of the sequence, which we use for the spectroscopic determination of the excitation energy of Rydberg levels of \(^{174}\)Yb, is illustrated in figure 2(a). The cycle is initialized by turning off the slower and MOT light for \( 100 \) ms to remove all remaining atoms from the previous cycle. In the loading phase, the slower and the MOT light are switched on with the MOT light detuned by \( \approx 22 \) MHz from the atomic resonance. The MOT is loaded for \( 3.5 \) s, to ensure saturation of the number of atoms in the MOT. Subsequently, the slower light is switched off and the Rydberg laser is switched on during the spectroscopy phase. If the Rydberg laser is resonant with a transition from the \( 6s6p \, ^1P_1 \) - state to a Rydberg state, atoms are removed from the MOT and correspondingly the MOT fluorescence is reduced. To determine the relative loss induced by the Rydberg laser, fluorescence images of the MOT are taken right before the Rydberg laser is turned on and after \( 1 \) s of illumination. During this time, the frequency of the Rydberg laser is ramped over 10 MHz. The whole cycle is repeated between 50 and 100 times with varying frequency of the Rydberg laser in order to take a spectrum over a specific frequency range. Every fifth cycle is a calibration cycle, where the Rydberg laser is not switched on in the spectroscopy phase. This calibration is required to account for long-time fluctuations of the number of atoms trapped in the MOT.

A typical trap-loss spectrum is depicted in figure 2(b). The excitation frequencies (given in wave numbers) are calculated by summing the excitation frequency of the intermediate \( 6s6p \, ^1P_1 \) - state and the measured frequency of the Rydberg laser which has an error of \( \approx 0.005 \) cm\(^{-1}\) due to the uncertainty of the wavemeter. This includes the error of the wavemeter calibration, using a frequency locked laser, the wavelength measurement itself and the wavemeter drift over a day. Because
of frequent calibrations, the day to day drift is canceled out. Other sources of error such as the Zeeman shift due to the MOT magnetic field are determined to be significantly smaller. With a magnetic field gradient of $\approx 40 \text{ G cm}^{-1}$ and a MOT size of $\approx 2 \text{ mm}$ the variation of the magnetic field in the MOT volume is $\approx 8 \text{ G}$ leading to an uncertainty of $\approx 11 \text{ MHz}$, which is much smaller than the wavemeter error and can thus be neglected.

To determine the polarizability of a specific Rydberg state, line spectra are taken at various values of an externally applied electric field $\vec{F}$. The external field is generated by applying a voltage to the electric field plate which is located on top of the vacuum chamber. Due to the complex geometry of our chamber and the uncertainty in the exact position of the MOT relative to grounded surfaces the corresponding electric field has a large uncertainty which is estimated to be more than 50%. Therefore, we only give the value of the externally applied voltage in figure 2. Nevertheless, the experimental geometry ensures that the electric field $\vec{F}$ points predominantly in the vertical direction at the position of the atoms.

In figure 3, typical spectra are shown for the $6s83s^1S_0$ - state (a) and the $6s83p^1P_1$ - state (b), where spectra for different values of the applied voltage are combined into a single graph and the maxima are fitted to a function which is quadratic in the applied voltage (see equation (3)). For the relatively small values of the applied voltage, only Rydberg states experience a noticeable electric-field (Stark) shift and hence an observed line shift corresponds directly to an energy shift of the Rydberg state. Hence, we will refer to plots as shown in figure 3 as Stark maps.

A notable feature of the obtained Stark maps is that the $6s83s^1S_0$ - line is only shifted by the external electric field while the $6s83p^1P_1$ - line is additionally split into two components. This reflects the fact that the Stark shift depends on the absolute value $|m_J|$ of the magnetic quantum number which can take the values $|m_J| = 0, 1$ for the $6s83p^1P_1$ - state while for the $6s83s^1S_0$ - state only $|m_J| = 0$ is possible. At the apex of the $6s83p^1P_1$ Stark map, we observe almost no excitation of the atoms into the Rydberg state, in agreement with selection rules that forbid transitions between the two odd-parity states $6s6p^1P_1$ and $6s83p^1P_1$. At finite electric field, even-parity states are admixed to the $6s83p^1P_1$ - state and transitions become possible.

In addition, the apex of the Stark maps is not at zero external field and is not even at an identical position for different Stark
maps. We attribute this to background electric fields in the chamber which are most likely originating from patch charges on the top window. Those patch charges may vary on the time scale of days. To remove these charges, at least partially, we regularly illuminate the top window with a 340 nm LED \[\text{[22]}\] in between experimental runs. Vanishing of the $6s^3p^1P_1$ - line at the apex of the Stark map indicates that the applied external electric field which is oriented vertically compensates the background field to a large extent and we may assume that the background field is also oriented vertically.

The broadening of the spectra if an electric field is applied in figure 3 is attributed to a saturation of the trap loss due to a Rydberg excitation probability which is increasing with applied electric field. This saturation effect is significantly larger for the allowed transitions to the $6s^3P_1$ -states. However, this broadening mechanism has little effect on the determination of the position of the line centers which is relevant for the determination of polarizabilities as described below.

### 4. Results and discussion

Our results on the excitation energies and polarizabilities of the $6sns^1S_0$ and the $6snp^1P_1$ series of $^{174}$Yb in the range from \(n = 70\) to \(n = 90\) are summarized in table 1. For each line a complete Stark map was recorded to determine the polarizability and the zero-field excitation energy. The excitation energies for the $6snp^1P_1$ series are extrapolated from quadratic fits to the Stark maps (see figure 3).

In a system with two valence electrons like Yb a full theoretical description of the Rydberg series can be obtained using Multi-Channel Quantum Defect Theory \[\text{[21, 23]}\]. However, for large principal quantum numbers and in the absence of perturbations by doubly excited states, the excitation energies of a Rydberg series are to a good approximation described by a single quantum defect according to

\[
E_{\text{exc}} = E_{\text{ion}} - \frac{R_y}{(n - \delta_{L,J})^2} = E_{\text{ion}} - \frac{R_y}{n^{*2}}. \tag{1}
\]

Here \(R_y = 10973696.959\, \text{m}^{-1}\) is the mass corrected Rydberg constant and \(E_{\text{ion}} = 50443.07041(25)\, \text{cm}^{-1}\) the first ionization energy \[\text{[21]}\]. The effective principal quantum number \(n^* = n - \delta_{L,J}\) is determined by the quantum defect \(\delta_{L,J}\), which describes the deviation from the hydrogenic case. It is due to the incomplete shielding of the charge of the nucleus by the inner electrons, which is particularly important for small \(L\) \[\text{[1]}\].

In figure 4, the measured excitation energies are plotted as a function of the principle quantum number. The values for the $6sns^1S_0$ series that we have obtained using the simple trap-loss method are in good agreement with previously reported values by the group at Laboratoire Aimé-Cotton \[\text{[21]}\] that have been measured using field-ionization of Rydberg states, which are also shown in the table. From fits of the measured excitation energies to equation (1) we get the quantum defects \(\delta_{0,0} = 4.2721(25)\) and \(\delta_{1,1} = 3.9535(9)\), where the value for the $6sns^1S_0$ series is again in good agreement with \[\text{[21]}\].

If Rydberg spectroscopy is performed in a small external electric field \(\vec{F}\) a quadratic Stark shift

\[
\Delta E = -\frac{1}{2}\alpha|\vec{F}|^2 \tag{2}
\]

has to be taken into account where \(\alpha\) is the state-dependent atomic polarizability. The total excitation energy is then given by

\[
E_{\text{exc}}(F) = E_{\text{ion}} - \frac{R_y}{n^*} - \frac{1}{2}\alpha|\vec{F}|^2. \tag{3}
\]

The polarizability \(\alpha\) of a Rydberg state with principal quantum number \(n\), total angular momentum \(J\) and magnetic quantum number \(m_J\) can generally be written as \[\text{[24]}\]

\[
\alpha(n, J, |m_J|) = \alpha_0(n, J) + \alpha_2(n, J) \frac{3m_J^2 - J(J + 1)}{J(2J - 1)}, \tag{4}
\]

with the scalar polarizability \(\alpha_0(n, J)\) and the tensor polarizability \(\alpha_2(n, J)\). Thus, the external electric field splits a Rydberg
Table 1. Experimental excitation energies of the 6sns1S0 and the 6sns3P1 Rydberg series of 174Yb and polarizabilities relative to the measured polarizability of the 6s70P1S0 state. The stated values for $E_{\text{exc}}$ are the measured energy differences between the 6s23S0 ground state and the respective Rydberg state in cm$^{-1}$. $E_{\text{ref}}$ are the corresponding energies measured in [21]. For the 6ns$^3$S$^0$ series, $\alpha_0$ is the experimentally determined relative scalar polarizability. For the 6ns$^3$P$^1$ series $\alpha_s$ and $\alpha_t$ are the experimentally determined relative polarizabilities corresponding to $|m_J| = 0, 1$ and $\alpha_0$ and $\alpha_2$ are the derived values of the scalar and tensor polarizabilities according to equation (4). The accuracy of the excitation energies is limited by the accuracy of our home-built wavemeter and is estimated to be 150 MHz or correspondingly 0.005 cm$^{-1}$. For the measured polarizabilities the main error results from the quadratic fit and is around 10%.

| State       | $E_{\text{exc}}$ | $E_{\text{ref}}$ | $\alpha_0$ | State       | $E_{\text{exc}}$ | $\alpha_s$ | $\alpha_t$ | $\alpha_0$ | $\alpha_2$ |
|-------------|-----------------|-----------------|------------|-------------|-----------------|------------|------------|------------|------------|
| 6s70P1S0    | 50417.693       | 50417.6654      | 1.00       | 6s70P1S0    | 50417.915       | 0.85       | 0.38       | 0.54       | −0.16      |
| 6s71S1S0    | 50418.420       | 50418.4212      | 0.88       | 6s71P1S1    | 50418.661       | 1.00       | 0.37       | 0.58       | −0.21      |
| 6s72S2S0    | 50419.142       | 50419.1438      | 1.02       | 6s72P1S2    | 50419.372       | 1.00       | 0.43       | 0.62       | −0.19      |
| 6s73S3S0    | 50419.835       | 50419.8350      | 1.18       | 6s73P1S3    | 50420.051       | 1.59       | 0.56       | 0.90       | −0.34      |
| 6s74S4S0    | 50420.497       | 50420.4966      | 1.37       | 6s74P1S4    | 50420.705       | 1.20       | 0.54       | 0.76       | −0.22      |
| 6s75S5S0    | 50421.136       | 50421.1305      | 1.80       | 6s75P1S5    | 50421.328       | 1.27       | 0.42       | 0.70       | −0.28      |
| 6s76S6S0    | 50421.737       | 50421.7379      | 1.77       | 6s76P1S6    | 50421.928       | 1.59       | 0.56       | 0.90       | −0.34      |
| 6s77S7S0    | 50422.326       | 50422.3208      | 1.55       | 6s77P1S7    | 50422.505       | 1.60       | 0.65       | 0.97       | −0.32      |
| 6s78S8S0    | 50422.881       | 50422.8797      | 1.53       | 6s78P1S8    | 50423.058       | 1.78       | 0.63       | 1.01       | −0.38      |
| 6s79S9S0    | 50423.422       | 50423.4164      | 2.09       | 6s79P1S9    | 50423.588       | 2.10       | 0.72       | 1.64       | −0.46      |
| 6s80S10S0   | 50423.935       | 50423.9322      | 2.83       | 6s80P1S0    | 50424.095       | 2.19       | 0.76       | 1.24       | −0.48      |
| 6s81S11S0   | 50424.432       | —                | 2.45       | 6s81P1S1    | 50424.585       | 2.65       | 0.79       | 1.41       | −0.62      |
| 6s82S12S0   | 50424.907       | —                | 2.64       | 6s82P1S2    | 50425.053       | 3.05       | 1.01       | 1.69       | −0.68      |
| 6s83S13S0   | 50425.365       | —                | 2.84       | 6s83P1S3    | 50425.510       | 3.13       | 1.03       | 1.73       | −0.70      |
| 6s84S14S0   | 50425.805       | —                | 3.54       | 6s84P1S4    | 50425.945       | 3.22       | 1.02       | 1.75       | −0.73      |
| 6s85S15S0   | 50426.228       | —                | 3.17       | 6s85P1S5    | 50426.364       | 3.69       | 1.45       | 2.20       | −0.75      |
| 6s86S16S0   | 50426.638       | —                | 3.70       | 6s86P1S6    | 50426.765       | 3.78       | 1.51       | 2.02       | −0.76      |
| 6s87S17S0   | 50427.031       | —                | 3.92       | 6s87P1S7    | 50427.160       | 4.41       | 1.45       | 2.43       | −0.99      |
| 6s88S18S0   | 50427.413       | —                | 4.92       | 6s88P1S8    | 50427.532       | 4.41       | 1.57       | 2.52       | −0.95      |
| 6s89S19S0   | 50427.781       | —                | 4.69       | 6s89P1S9    | 50427.898       | 4.67       | 1.56       | 2.60       | −1.04      |
| 6s90S20S0   | 50428.137       | —                | 5.60       | 6s90P1S10   | 50428.244       | 3.80       | 1.07       | 1.98       | −0.91      |

Figure 4. Excitation energies for the 6sns1S0 (a) and 6sns3P1 (b) Rydberg-series of 174Yb. The solid lines correspond to fits according to equation (1) with $\delta_{2,1}$ being the only free fitting parameter. The obtained values for the quantum defect are $\delta_{0,0} = 4.2721(25)$ and $\delta_{1,1} = 3.9535(9)$. The y-axis has an offset of 50 400 cm$^{-1}$.

Using these formulas, the values for $\alpha_0(n,1)$ and $\alpha_2(n,1)$ given in table 1 are calculated under the assumption that the tensor polarizability $\alpha_2(n,1)$ is negative implying
Figure 5. Relative scalar and tensor polarizabilities of the 6sns$^1$S$_0$ (a) and the 6sn$p^1$P$_1$ (b) Rydberg-series of $^{174}$Yb. The scalar polarizability $\alpha_0(n, 0)$ of a state 6sns$^1$S$_0$ is directly determined from a quadratic fit to the external electric field (see figure 3 (a)). The Scalar polarizabilities $\alpha_0(n, 1)$ (black dots) and the tensor polarizabilities and $\alpha_2(n, 1)$ (red dots) of the 6sn$p^1$P$_1$ - states are determined using quadratic fits and making use of equation (4). All polarizabilities are normalized to the measured scalar polarizability of the 6s70s$^1$S$_0$ state. Solid lines are fits of the polarizabilities to functions of the type $(a(n - \delta_{LJ}))^3 = (an^*)^3$.

that the measured component with the larger polarizability $\alpha_2(n, 1)$ for a given state of the 6sns$^1$P$_1$ series corresponds to $J = 1, |m_J| = 0$. This assumption seems justified as it resembles the behavior of the polarizabilities in highly excited $^2$P$_{3/2}$ - Rydberg states of rubidium [25, 26]. In figure 5, the determined scalar and tensor polarizabilities for all states of the 6sns$^1$S$_0$ (a) and the 6sn$p^1$P$_1$ series of $^{174}$Yb in the range of $n = 70$ to $n = 90$ are shown. The error bars result from quadratic fits to the line center of each spectrum and are on the order of 10%. The error of the relative electric field values can be neglected compared to the fit errors, since the voltage supply has an accuracy of better than 1%, leading to a maximum error contribution of 2% to the polarizability. The solid lines in the figure are fits of the polarizabilities to functions of the type $(a(n - \delta_{LJ}))^3 = (an^*)^3$, which reflect the expected scaling with $n^3$ of the polarizabilities of highly excited Rydberg states [1]. Due to the large uncertainty of the value of the external electric field at the position of the atoms all polarizabilities are normalized to the measured polarizability of the 6s70s$^1$S$_0$ - state.

5. Conclusion

In this manuscript, we have presented a spectroscopic measurement of the 6sns$^1$S$_0$ and the 6sn$p^1$P$_1$ Rydberg-series of $^{174}$Yb which is based on simple trap-loss spectroscopy in a magneto-optical trap. Using this method, we have obtained excitation energies for Rydberg states in the range of $n = 70$ to $n = 90$ which are in agreement with previously reported results on the 6sns$^1$S$_0$ series and extend those results to larger principal quantum numbers and the 6sn$p^1$P$_1$ series. In addition, we have performed an investigation of the Stark effect in low electric fields allowing us to infer scalar and tensor polarizabilities which exhibit the expected $n^3$ scaling behavior. These results will be valuable for further experiments using Yb Rydberg atoms. The method described in this manuscript can also be used to study other Yb isotopes. Specifically, we plan to investigate the fermionic isotopes $^{171}$Yb and $^{173}$Yb where interesting effects due to the hyperfine coupling are expected [27].

While trap-loss spectroscopy is a very useful and simple method for the spectroscopy of Rydberg states it is not suitable for experiments involving time-resolved measurements or coherent manipulation of Rydberg states. Such experiments will require conservative trapping of the atoms to get rid of the perturbing influence of the MOT light. In addition, other detection methods such as field-ionization or detection by electromagnetically induced transparency [28] will have to be applied. In order to be able to perform such exciting experimental studies we are upgrading our apparatus accordingly.

Data availability statement

Research data supporting this publication are available from the corresponding author upon reasonable request.

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