INVESTIGATION OF THE FIRST IONIZATION POTENTIAL OF YTTERBIUM IN ARGON BUFFER GAS*

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(Received January 9, 2018)

One of the most important atomic properties influencing elements chemical behaviour is the energy required to remove its outermost electron: the first ionization potential (IP). The determination of this basic atomic property is challenging for the transfermium elements with $Z > 100$. Recently, Rydberg states have been observed in nobelium (No, $Z = 102$) inside a buffer gas cell. The buffer gas environment influences the energy of the Rydberg levels and thus the IP extracted from analysing the Rydberg series. Therefore, laser resonance ionization spectroscopy in a buffer gas cell was employed to determine the IP of its chemical homologue, ytterbium (Yb, $Z = 70$) at different buffer gas pressures to characterize the systematics arising from the buffer gas environment.

DOI:10.5506/APhysPolB.49.599

1. Introduction

Atomic structure studies of the heaviest elements with $Z > 100$ is one of the most interesting and challenging disciplines of atomic physics [1]. Rel-

* Presented at the XXXV Mazurian Lakes Conference on Physics, Piaski, Poland, September 3–9, 2017.

ativistic, correlation and quantum electrodynamics effects (QED) have a large influence on the atomic structure of such elements. Relativistic effects manifest in a shrinking of the s- and $p_{1/2}$-electron wave functions leading to a change in the nuclear potential screening by the inner shell electrons. The change alters the binding energies of the valence electrons, which may result in distinct atomic and chemical properties of these elements [2]. The first ionization potential (IP) is one of the most fundamental properties of a chemical element and represents the binding energy of the most weakly bound electron of an atom. The determination of the IP provides an essential test of our understanding of the electronic structure and the chemical properties of an element. Thus, measurements of atomic level energies and IPs of heavy elements provide a benchmark for modern atomic theories.

Transfermium elements with $Z > 100$ are produced solely in accelerator-based nuclear fusion–evaporation reactions at very low production rates. Thus, the study of these elements requires techniques which are sensitive on an atom-at-a-time scale. Recently, we reported on measurements on nobelium where a ground state transition was identified [3], which were performed at SHIP, GSI in Darmstadt. In these experiments, we used an ultra-sensitive experimental method that is based on the RAdiation Detected Resonance Ionization Spectroscopy (RADRIS) technique in a buffer gas environment [4, 5]. This has enabled the measurement of many Rydberg states and the determination of the IP of nobelium with a very high precision by analyzing the convergence of Rydberg series (P. Chhetri *et al.*, manuscript in preparation). The Rydberg states undergo a shift in energy in a gas environment. This prompted an investigation into the pressure dependence of the IP of ytterbium (Yb, $Z = 70$), the chemical homologue of No, where the atomic structure is well-established [6, 7]. In order to transfer the results to nobelium, the Rydberg levels with a similar principle quantum number as observed on-line for No [3] were studied. These measurements were performed at different buffer gas pressures to understand the systematic effects of our measurement technique on the extraction of the IP.

2. Experimental set-up and technique

Off-line measurements were performed on ytterbium with a natural isotope composition, $^{\text{nat}}$Yb. A schematic representation of the used off-line set-up can be found in Ref. [8]. In the gas cell, which was filled with argon gas of 99.9999% purity, a 25 μm thick foil of Yb was clamped onto a tantalum filament of thickness of 12.5 μm. The latter was resistively heated to evaporate the Yb atoms. Using two tunable dye lasers, a two-step photoionization was applied. The dye lasers were simultaneously pumped by an excimer laser at 308 nm. The lasers were operated at a repetition rate of 100 Hz with a pulse energy of around 1 mJ. The light from the dye lasers was

transported to the experimental set-up through optical fibres and collimated into the buffer gas cell. All ions created by photo-ionization were electrically guided to a wire connected to a charge-sensitive preamplifier where the accumulated charge was then detected in coincidence with the laser pulse. The signal amplitude, which is proportional to the number of produced photo-ions, was then recorded as a function of the wavenumber being continuously monitored with a precision of $0.01\,\mathrm{cm}^{-1}$. More information can be found in Refs. [3–5, 8].

3. Results

A two-step photo-excitation scheme was used to excite the Rydberg states in $^{\mathrm{nat}}$Yb. The first step was tuned to $25\,068.22\,\mathrm{cm}^{-1}$ to excite the $^1\mathrm{S}_0 \rightarrow {}^1\mathrm{P}_1$ optical transition in Yb, while the second step was set to states close to the IP. In total, a spectral range from 25 000 to $25\,250\,\mathrm{cm}^{-1}$ for the second excitation step was scanned to locate the Rydberg states. The Rydberg atoms were then subsequently ionized either by residual laser light, black-body radiation or collisional processes. These Rydberg states were excited at 6 different pressures ranging from 5 to 200 mbar. Figure 1 shows the

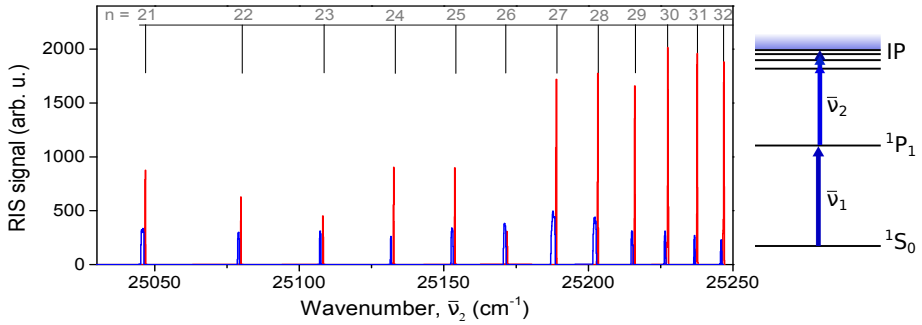

Fig. 1. Left panel: The measured Rydberg states in Yb at 5 mbar (lighter/red line) and 200 mbar (darker/blue line) plotted against the wavenumber ($\bar{\nu}_2$) along with their respective principal quantum number (n) on top. Right panel: Simplified ionization scheme.

observed Rydberg states plotted as a function of the wavenumber ($\bar{\nu}_2$) for two different pressures. It illustrates that the Rydberg states broaden and shift to lower energies for higher argon pressure which is similar to observation reported for the Rydberg states in Sm I [9]. The excitation energies of the Rydberg states (E_n), determined from the centroids of a Gaussian profile fitted to each peak, follows a trend given by the Rydberg formula [10]:

$$E_n = E_{\mathrm{lim}} - \frac{R_m}{(n - \delta(n))}\,. \tag{1}$$

E_{lim} is the convergence limit of the fit, R_m is the reduced mass Rydberg constant for $^{\mathrm{nat}}$Yb, n is the principal quantum number. $\delta(n)$ denotes the quantum defect which is defined by the Ritz expansion in the first order as [11, 12]

$$\delta(n) = \delta_0 + \frac{B}{n - \delta_0} \tag{2}$$

with the constant quantum defect δ_0 and B being fit parameters. Figure 2 shows the position of the observed Rydberg states as a function of their principal quantum number n at a gas pressure of 60 mbar. The best fit to the data showed a convergence at $E_{\mathrm{lim}} = 25\,734.88(3)\,\mathrm{cm}^{-1}$. This together with the first excitation step of $25\,068.22\,\mathrm{cm}^{-1}$ gives an IP of $E_{\mathrm{IP}} = 50\,443.1(3)\,\mathrm{cm}^{-1}$. Similar fits were performed at the other pressures and for each the IP was extracted. Plotting the IP for different pressures clearly shows a systematic shift to lower values of the ionization potential at higher gas pressures, as shown in Fig. 3. This effect can be quantified by a linear fit. The slope of the fit gives a pressure shift of the IP, which in this case amounts to $-0.0060(7)\,\mathrm{cm}^{-1}$ per mbar. The result of the linear fit allows us to determine the IP in vacuum to be $E_{\mathrm{IP_{vac}}} = 50\,443.35(10)\,\mathrm{cm}^{-1}$. This value agrees well with the literature value of $50\,443.2(1)\,\mathrm{cm}^{-1}$ [7].

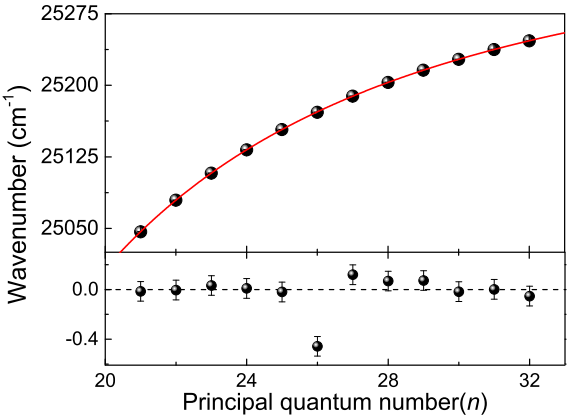

Fig. 2. The identified Rydberg states at a buffer gas pressure of 60 mbar plotted against the corresponding principle quantum number n and a best fit (solid line) according to the Rydberg–Ritz formula, equation (1). The lower graph shows the residuals of the fit.

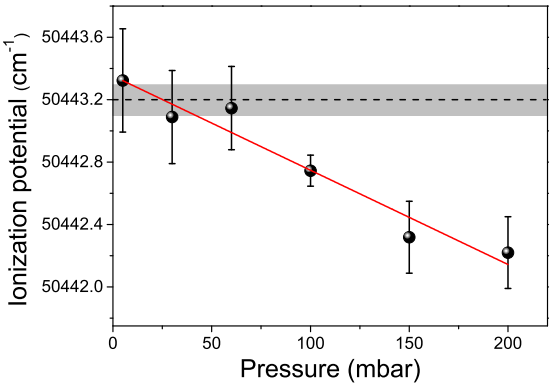

Fig. 3. Extracted IP of Yb plotted as a function of argon pressure along with a linear fit to the data (solid line). Dotted line represents the literature value for the IP with the error (shaded area).

4. Summary

An in-gas cell laser spectroscopy technique was used to extract the IP of Yb at different pressures of argon. This study showed that there is a systematic effect of a buffer gas on the individual Rydberg levels and thus on the IP. Since the on-line measurements for No with the RADRIS set-up at the SHIP separator were performed at 90 mbar argon pressure, the studied systematics in the homologue Yb can be used to estimate the systematic uncertainties for the on-line measurement for nobelium.

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