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On the first ionization potential of lithium

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Abstract
New experimental data on the highly excited Rydberg states of lithium have been acquired, using a two-step laser excitation technique in combination with a thermionic diode ion detector. The atoms are prepared in the 3s $^2S_{1/2}$ intermediate level by two-photon excitation from the ground state, whereas the np $^2P_{1/2,3/2}$ levels have been approached via single photon absorption. The new observations include much extended np $^2P_{1/2,3/2}$ Rydberg series ($15 \leqslant n \leqslant 60$). The Rydberg relation fit to the new data yields the quantum defect for the np $^2P_{1/2,3/2}$ series as 0.051(4) and the binding energy of the 3s $^2S_{1/2}$ level as 16 281.19(2) cm^{-1}. Adding the energy of the 3s $^2S_{1/2}$ level and its binding energy reveals the first ionization potential of lithium as 43 487.26(3) cm^{-1}.

1. Introduction

Atoms in the highly excited states have been extensively studied since the availability of narrow bandwidth dye lasers and the results have been discussed in a number of books (Letokhov 1987, Gallagher 1994, Demtroder 1996, Connerade 1998). Lithium, possessing a single valence electron, is the simplest element after hydrogen and helium, but not much attention has been paid to the study of its highly excited levels. Study of the Rydberg series in atoms and molecules up to higher principal quantum numbers yields accurate value of the ionization potential. The first ionization potential of lithium was determined by Johansson (1959) as 43 487.150(5) cm^{-1} who fitted the data of the ns ($2 \leqslant n \leqslant 8$), np ($2 \leqslant n \leqslant 8$), nd ($3 \leqslant n \leqslant 9$) and the nf ($4 \leqslant n \leqslant 9$) levels to the Ritz formula. Subsequently, Lorenzen and Niemax (1983) used the data of Johansson (1959) and derived this value as 43 487.15(3) cm^{-1} based on a modified Ritz relation. Gallagher (1994) used the data for the np series ($5 \leqslant n \leqslant 15$) listed in the NBS tables (Moore 1971) and derived the quantum defects and the ionization potential as 0.046(1) and 43 486(1) cm^{-1}, respectively. A more accurate and reliable value of the ionization limit can be extracted by observing the highly excited states of an atom close to the series limit. In lithium, there have been a few attempts to observe the highly excited $\ell = 0$ and 2 levels via two-photon excitation, because strong lithium molecular absorption due to the

$X\,^1\Sigma_g^+ \rightarrow B\,^1\Pi_u$ band system made it almost impossible to detect these levels near the series limit. Young *et al* (1979) used a thermionic diode detector and managed to observe the ns $^2S_{1/2}$ and nd $^2D_{3/2,5/2}$ series up to $n = 32$ and $n = 49$, respectively, via two-photon absorption. Young *et al* (1979) neither reported the energies of these levels nor made any attempt to improve the value of the first ionization potential of lithium. The latest NIST compilation includes the np $^2P_{1/2,3/2}$ series data up to $n = 42$ which is based on an unpublished report by Kelly (1985). To our knowledge laser based data on the highly excited states for the $\ell = 0, 1, 2$ and 3 levels are not available for lithium. The motivations of the present experimental work have been to record the $\ell = 0, 1, 2$ and 3 levels, to compile accurate energies and quantum defects and to extract the first ionization potential of lithium.

The related laser based work includes the saturated three-step resonance ionization of the ground state of lithium and photo-ionization cross section of the excited state $n = 3$ by Kramer *et al* (1979) and the radiative lifetime measurements of 15 levels by Hansen (1982). Recently, Baig *et al* (1994) reported the nf $^2F_{5/2,7/2}$ ($13 \leqslant n \leqslant 48$) series using a two-colour three-photon resonant excitation technique. More recently, Ballard *et al* (2001) reported the two-photon and three-photon excitation spectra of ^{7}Li and their observations include 2s $^2S \rightarrow n\,^2P$, 2s $^2S \rightarrow n\,^2F$, 2p $^2P \rightarrow n$p ^2P and 2p $^2P \rightarrow n$f ^2F Rydberg series. However, the work of Ballard *et al* (2001) suffered AC Stark shift and broadening and they did not comment on the ionization potential of lithium.

In the present paper, we report new studies on the highly excited levels of lithium using two-step laser excitation in conjunction with a thermionic diode detector. Using 3s $^2S_{1/2}$ as the intermediate level the np $^2P_{1/2,3/2}$ series has been considerably extended ($15 \leqslant n \leqslant 60$) revealing an accurate value of the binding energy of the 3s $^2S_{1/2}$ level as 16 281.19 (2) cm^{-1}. Adding the energy of the intermediate level as 27 206.066 cm^{-1} (Radziemski *et al* 1995), the first ionization potential of lithium is determined as 43 487.26(3) cm^{-1}.

2. Experimental details

The experimental arrangements are the same as described in our earlier papers (Baig *et al* 1994, 2004). The laser system comprised a Q-switched pulsed Nd: YAG laser (Quantel, Brilliant), 10 Hz repetition rate, pulse duration $\approx$4 ns, energy 180 mJ per pulse at 532 nm. The line width of the laser is 1.4 cm^{-1} at 532 nm. The output of the SHG of the Nd: YAG laser was used to pump two dye lasers, a Quanta-ray, PDL-2 and a home built with a cavity pioneered by Hanna *et al* (1975). The cavity was formed between a flat mirror and a 2400 lines mm^{-1} holographic grating and the wavelength tuning was achieved by rotating the grating by a computer controlled stepping motor.

The ion detector was a single wire thermionic diode configuration which comprised a 48 cm long stainless steel tube having 3 cm diameter and 1 mm wall thickness. About 23 cm of the central zone of the detector was placed in a clamp-shell furnace. Both ends of the furnace tube were water-cooled and sealed with 25 mm diameter quartz windows. The ultimate temperature was monitored by a Ni–Cr–Ni thermocouple and it was maintained within $\pm2\%$ by a temperature controller. A molybdenum wire 0.25 mm in diameter stretched axially was used as a cathode that was heated by a separate regulated dc power supply. About 2 g of spectroscopically pure lithium sample was placed in the central heating zone. The cell was evacuated up to 10^{-6} Torr and subsequently filled with argon gas at about 0.2 Torr. The buffer gas provided a uniform column of the vapour, and it also protected the quartz windows from the chemical attack of the hot vapours. The thermionic diode was then heated up to 800 K temperature corresponding to a lithium vapour pressure of 0.01 Torr.

The dye laser wavelength was initially determined by a monochromator (Ocean Optics, HR2000) equipped with a 600 lines mm^{-1} grating. The wavelength calibration was achieved by recording simultaneously the output from the thermionic diode, the optogalvanic spectra of neon from a hollow cathode lamp and etalon rings from a 1 mm thick fused silica Fabry–Perot etalon (FSR 3.33 cm^{-1}) via three boxcar averagers (SR 250). The optogalvanic effect into the neon hollow cathode lamp provided well-distributed spectral lines of neon in the region of interest that serve as wavelength standards listed in the MIT table (Harrison 1982). The interference fringes from the etalon were used to interpolate between the neon lines. Each data point is an average of five laser shots. The spectrum was recorded with a 0.05 cm^{-1} step of the scanning dye laser that governs the uncertainty of the measurement of the line position. From the location of the peak signal positions, the transition energies have been determined within an accuracy of ± 0.15 cm^{-1}.

Two dye lasers have been used to record the resonance enhanced two-step excitation spectra of lithium. The first dye laser pumped by the 532 nm was tuned at 735.1 nm to populate the 3s $^2S_{1/2}$ level via two-photon excitation. The dye laser beam was focused in the centre of the thermionic diode with a 50 cm focal length convex lens. The second dye laser, delayed by a few nanoseconds, was tuned in wavelength region from 634 nm to 613 nm. The ionization signal was optimized by adjusting the cathode current and the buffer gas pressure. The ionization signal was registered as a change in the voltage across a 100 kΩ load resistor. The three signals were simultaneously recorded on a computer through a GPIB card for subsequent processing and analysis.

3. Results and discussion

The ground state of lithium comprises a single valence electron occupying a 2s orbital. The excited states are of the form ns $^2S_{1/2}$, np $^2P_{1/2,3/2}$, nd $^2D_{3/2,5/2}$ and nf $^2F_{5/2,7/2}$ that can be approached via single-photon, two-photon or three-photon excitation from the ground state. The energy levels split into two components due to the spin–orbit interaction, except for $\ell = 0$ levels. In the present work 3s $^2S_{1/2}$ was selected as the intermediate level that was populated by two-photon absorption from the ground state, in accordance with the $\Delta\ell = 0, \pm 2$, $\Delta J = 0$, ± 2, even $\leftrightarrow$ even or odd $\leftrightarrow$ odd parity selection rules. The first dye laser was fixed at 735.1 nm and the second laser was scanned from 634 nm to 613 nm to cover the energy region up to the ionization limit. The np $^2P_{1/2,3/2}$ Rydberg series have been observed from $n = 15$ to 60 according to the following excitation process:

$$2s\,^2S_{1/2} \xrightarrow{2\hbar\omega} 3s\,^2S_{1/2} \xrightarrow{\hbar\omega} np\,^2P_{1/2,3/2}.$$
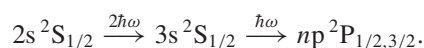
The identifications of the observed levels are confirmed by applying the appropriate selection rules for the stepwise excitation. A schematic energy level diagram and the relevant transitions are shown in figure 1. The lifetime of the 3s $^2S_{1/2}$ intermediate level is 30 ns. The population of this level might get redistributed through the available decay channels. The transition probability of the 3s $^2S_{1/2}$ level to the 2p $^2P_{1/2,3/2}$ level is 3.49×10^7 s^{-1} (Wiese *et al* 1966). The 2p resonance levels are also optically connected to the ground level with 3.72×10^7 s^{-1} transition probability. The lifetime of the aforementioned level is much larger than that of the dye laser, 4 ns. Therefore, the temporal delay of the second dye laser is not very critical.

Figure 2 shows the spectrum of atomic lithium recorded in the energy region 15 760 cm^{-1} to 16 300 cm^{-1}. The upper trace shows the well-distributed optogalvanic spectrum of neon that serves as wavelength standards for determining the energies of the unknown spectral
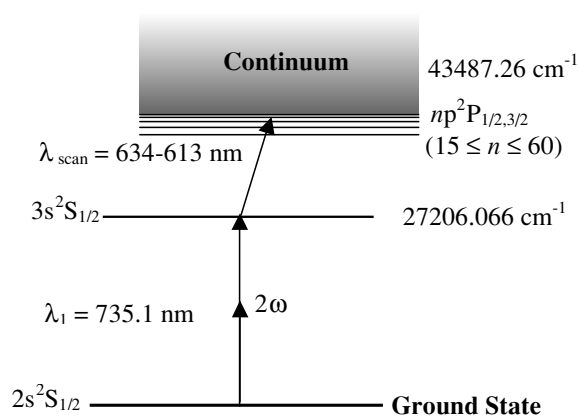
Figure 1. Energy level diagram of lithium to study the two-step excitation spectrum.
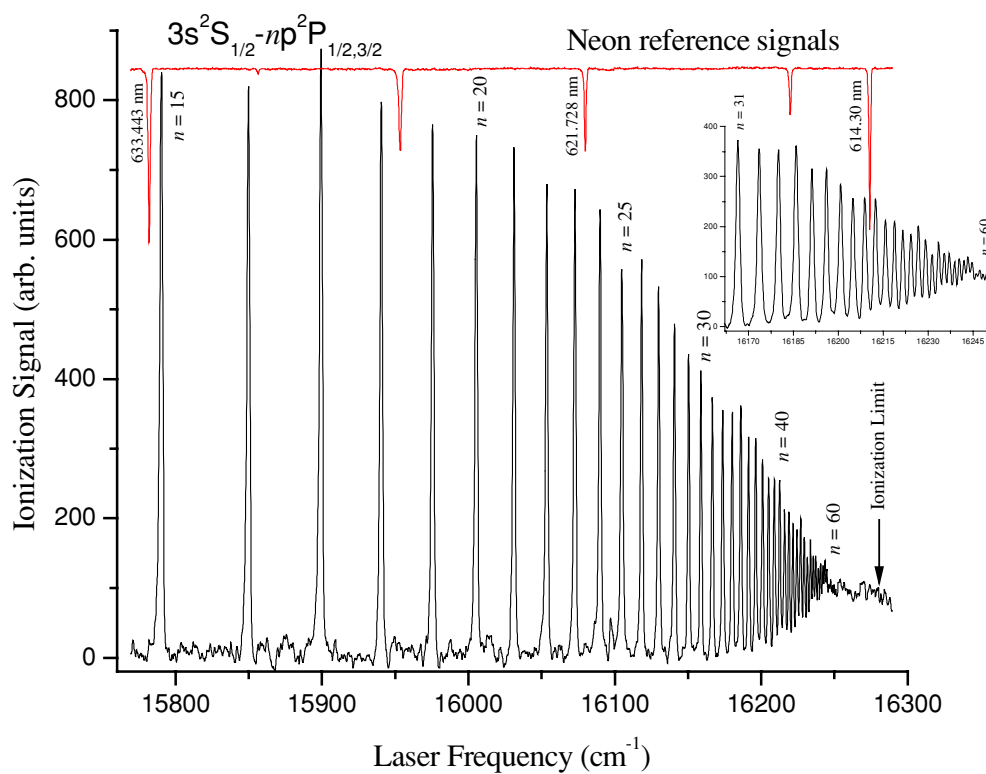

Figure 2. A portion of the spectrum of lithium showing the $np\ ^2P_{1/2,3/2}$ series obtained from the $3s\ ^2S_{1/2}$ intermediate level, populated in the first step by two-photon excitation, and scanning the second laser in the energy region from $15\,760$ cm^{-1} to $16\,300$ cm^{-1} corresponding to $n = 15$ up to the first ionization threshold. An enlarged section of the spectrum close to the limit is shown as an inset.

lines of lithium. An enlarged portion of the spectrum near the series limit is shown as an inset. It is evident that after $n = 20$ the signal intensity decreases with the increase of the

principal quantum number. The last member of the observed series lies only 30 cm^{-1} below the series limit whereas the energy difference between the adjacent levels is $\approx$1 cm^{-1}. We also noted that there is no systematic broadening of lines with increasing principal quantum number indicating a field-free environment. The widths of the observed lines are nearly three times higher than the dye laser line width. The larger width of the observed lines may be attributed to the power broadening or Doppler broadening. The energy of the dye laser was 1 mJ, the pulse duration was 4 ns and the diameter of the dye laser beam was 2 mm. Therefore, the intensity turns out to be $\approx$8 $\times$ 10^{10} W m^{-2}, the field is about 8 $\times$ 10^{6} V m^{-1} and the corresponding power broadening is estimated about 0.2 cm^{-1}. Similarly the contribution of the Doppler broadening is about 0.13 cm^{-1}. Adding all these factors the expected widths of the lines turn out to be comparable to those of the observed lines.

The energy levels of an unperturbed series can be represented by the extended Rydberg–Ritz formula:

$$E_n = \text{SL} - \frac{R_y}{(n - \mu_\ell)^2},$$

where E_n is the energy of the level with principal quantum number n, SL is the series limit, R_y is the mass corrected Rydberg constant of an atom ($R_{\text{Li}} = 109\,728.64$ cm^{-1}) and μ_ℓ is the quantum defect. The quantum defect is directly related to the interaction of the excited electron with the ion core and describes how much a Rydberg series departs from the behaviour of an atomic hydrogen Rydberg series that can be written as

$$\mu_\ell = a + \frac{b}{(n - a)^2} + \frac{c}{(n - a)^4} + \cdots.$$

We equate the transition energies (scanning laser frequency) of the 3s ^{2}S$_{1/2}$ $\rightarrow$ np ^{2}P$_{1/2,3/2}$ transitions to E_n. This is equivalent to measuring the energies of the np ^{2}P$_{1/2,3/2}$ levels with 3s ^{2}S$_{1/2}$ as the reference level. We then fit the Rydberg formula to the entire measured series of transitions (15 $\leqslant$ n $\leqslant$ 60) and determined the 3s ^{2}S$_{1/2}$ series limit as 16 281.19(2) cm^{-1}. The difference between the observed and calculated transition energies does not exceed 0.05(3) cm^{-1}. The term energies are then calculated by adding the measured transition energies to the accurately known energy of the 3s ^{2}S$_{1/2}$ level. Radziemski *et al* (1995) reported the energy of this level for ^{6}Li and ^{7}Li as 27 205.7129 cm^{-1} and 27 206.0952 cm^{-1} whereas the NIST Database (2004) gives the mass corrected value as 27 206.066(1) cm^{-1} that has been used here. The corresponding ionization potential of lithium is then 43 487.26(3) cm^{-1}. In table 1 we have tabulated the measured and calculated transition energies, the difference between the observed and calculated transition energies, term energies and effective quantum numbers of the observed series.

Johansson (1959) used the measurements of Meissner *et al* (1948, 1949) for the four lowest nd ^{2}D terms and using the Ritz formula determined the ionization potential of lithium as 43 487.150(5) cm^{-1}. Goy *et al* (1986) used this limit value and reported precise values of quantum defects for the ns and np Rydberg levels with n ranging from 18 to 40. Lorenzen and Niemax (1983) used the same data reported by Johansson (1959) and derived the series limit as 43 487.15(3) cm^{-1} based on a modified Ritz relation. Gallagher (1994) applied a graphical approach using data for 5p to 15p lines listed in the NBS table (Moore 1971) and determined the quantum defects and ionization potential of lithium as 0.046(1) and 43 486(1) cm^{-1} respectively. Baig *et al* (1994) reported the nf ^{2}F$_{5/2,7/2}$ (13 $\leqslant$ n $\leqslant$ 48) odd parity Rydberg series using a two-colour three-photon resonant excitation technique and extracted the ionization potential as 43 487.42(8) cm^{-1}. It is worth mentioning that Baig *et al* (1994) used the 2p ^{2}P$_{1/2,3/2}$ intermediate level and the nf ^{2}F$_{5/2,7/2}$ levels were excited via two-photon excitation. Since the spin–orbit splitting of the 2p level is 0.335 cm^{-1} and these

Table 1. A comparison of the measured and calculated level energies and calculated effective quantum number for the np ^{2}P$_{1/2,3/2}$ series in lithium using the Rydberg formula.

n	Laser frequency measured (cm^{-1})	Laser frequency calculated (cm^{-1})	Difference	Term energies (cm^{-1})	n^*
15	15 790.24	15 790.19	0.05	42 996.31	14.950
16	15 849.85	15 849.83	0.02	43 055.92	15.950
17	15 899.29	15 899.23	0.06	43 105.36	16.951
18	15 940.52	15 940.60	−0.08	43 146.59	17.947
19	15 975.58	15 975.60	−0.02	43 181.65	18.949
20	16 005.46	16 005.47	−0.01	43 211.53	19.950
21	16 031.20	16 031.16	0.04	43 237.27	20.951
22	16 053.47	16 053.43	0.04	43 259.54	21.951
23	16 072.79	16 072.84	−0.05	43 278.86	22.945
24	16 089.92	16 089.88	0.04	43 295.99	23.952
25	16 104.85	16 104.91	−0.06	43 310.92	24.947
26	16 118.21	16 118.23	−0.02	43 324.28	25.948
27	16 130.02	16 130.10	−0.08	43 336.09	26.942
28	16 140.71	16 140.72	−0.01	43 346.78	27.948
29	16 150.29	16 150.25	0.04	43 356.36	28.953
30	16 158.80	16 158.85	−0.05	43 364.87	29.943
31	16 166.59	16 166.63	−0.04	43 372.66	30.938
32	16 173.62	16 173.69	−0.07	43 379.69	31.939
33	16 180.13	16 180.12	0.01	43 386.20	32.952
34	16 185.92	16 185.98	−0.06	43 391.99	33.938
35	16 191.32	16 191.35	−0.03	43 397.39	34.943
36	16 196.23	16 196.28	−0.05	43 402.30	35.930
37	16 200.86	16 200.81	0.05	43 406.93	36.960
38	16 205.01	16 204.99	0.02	43 411.08	37.953
39	16 208.93	16 208.86	0.07	43 415.00	38.969
40	16 212.49	16 212.43	0.06	43 418.56	39.966
41	16 215.78	16 215.75	0.03	43 421.85	40.959
42	16 218.89	16 218.83	0.06	43 424.96	41.969
43	16 221.68	16 221.70	−0.02	43 427.75	42.941
44	16 224.32	16 224.38	−0.06	43 430.39	43.927
45	16 226.92	16 226.88	0.04	43 432.99	44.967
46	16 229.25	16 229.22	0.03	43 435.32	45.964
47	16 231.46	16 231.41	0.05	43 437.53	46.975
48	16 233.49	16 233.46	0.03	43 439.56	47.964
49	16 235.44	16 235.39	0.05	43 441.51	48.975
50	16 237.21	16 237.21	0.00	43 443.28	49.951
51	16 238.98	16 238.92	0.06	43 445.05	50.988
52	16 240.52	16 240.53	−0.01	43 446.59	51.944
53	16 242.08	16 242.05	0.03	43 448.15	52.970
54	16 243.48	16 243.49	−0.01	43 449.55	53.945
55	16 244.83	16 244.85	−0.02	43 450.90	54.937
56	16 246.15	16 246.13	0.02	43 452.22	55.962
57	16 247.36	16 247.35	0.01	43 453.43	56.954
58	16 248.53	16 248.51	0.02	43 454.60	57.966
59	16 249.63	16 249.61	0.02	43 455.70	58.967
60	16 250.68	16 250.66	0.02	43 456.75	59.973

levels were not resolved, therefore an average value of the 2p levels, based on the statistical weight of the fine structure components, was used to determine the first ionization potential.
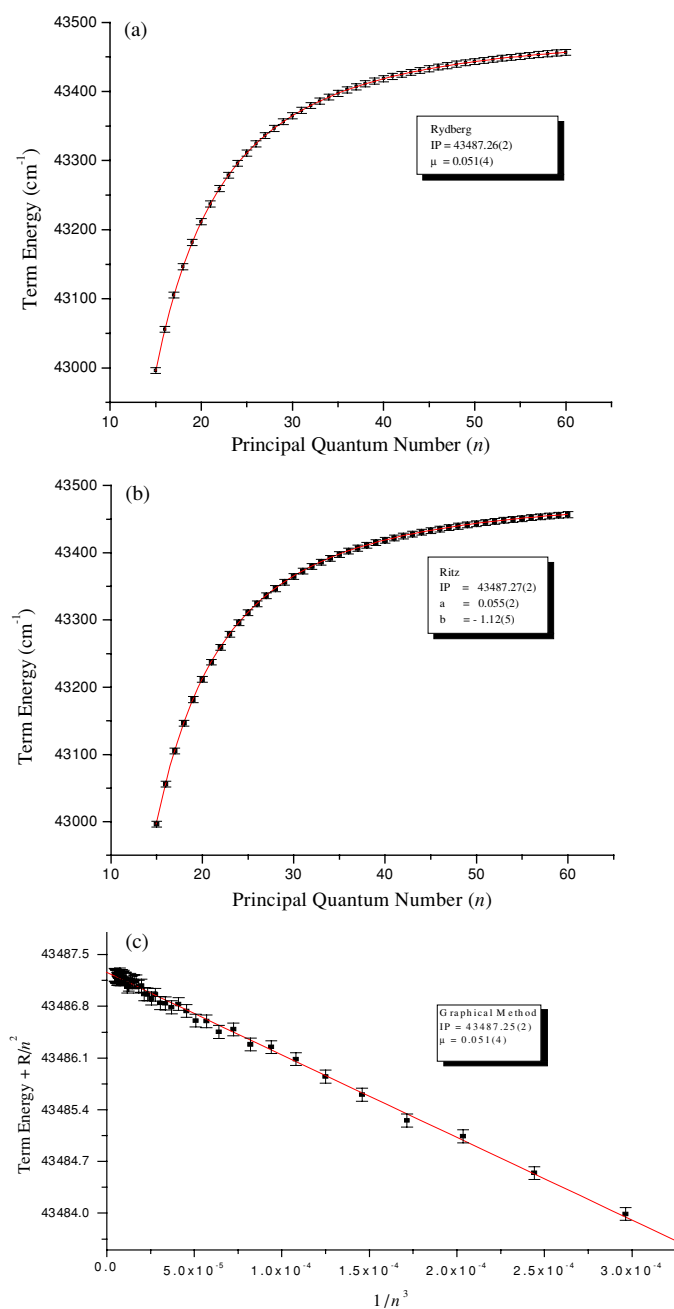
Figure 3. A least-squares fit to the experimental data, term energies of the np ^{2}P$_{1/2,3/2}$ series versus principal quantum number (n) is reproduced using the Rydberg, Rydberg–Ritz and graphical methods. The 0.002% error bars are also inserted in the graph.

Since the quantum defect approaches a constant value at larger principal quantum number, therefore the Rydberg–Ritz relation can also be used to extract the series limit and the quantum defect. The Ritz formula is particularly important if only lower members of a Rydberg series are
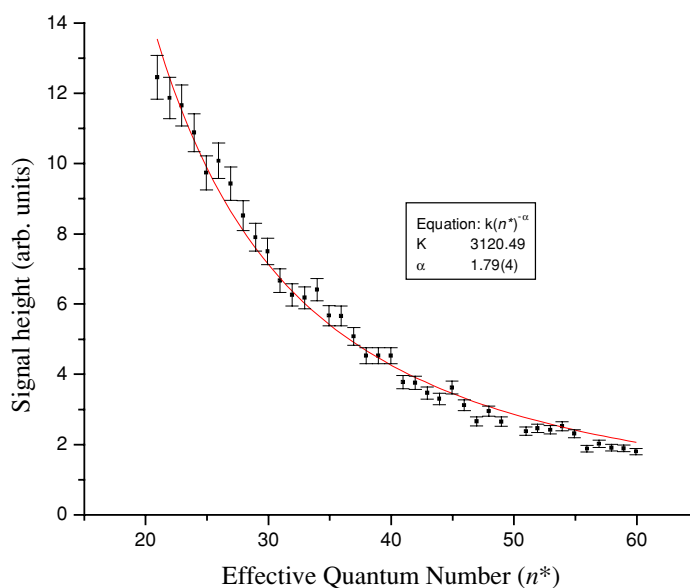
Figure 4. A plot of the relative intensities of the Rydberg series np ^{2}P$_{1/2,3/2}$ versus the effective quantum number (n^*). The solid line which passes through the data points represents an empirical relation: intensity $\propto (n^*)^{-\alpha}$ which yields the proportionality constant 3120.48 and $\alpha = 1.79(4)$. The 5% error bars are also inserted.

Table 2. A comparison of the calculated binding energies, ionization potentials, constants and calculated effective quantum number for the np ^{2}P$_{1/2,3/2}$ series in lithium using the Rydberg formula, Ritz formula and graphical method.

	3s ^{2}S$_{1/2}$ binding energy (cm^{-1})	Ionization potential	Constants
Rydberg	16 281.19(2)	43 487.26(2)	$\mu = 0.051(4)$
Ritz	16 281.21(2)	43 487.27(2)	$a = 0.055(2)$
			$b = -1.12(5)$
Graphical method	16 281.19(2)	43 487.25(2)	$\mu = 0.051(4)$

known whereas the Rydberg relation is applicable when series close to the limit are observed. The fit of the above-mentioned relation to the observed transitions gives the ionization potential of lithium as 43 487.27(2) cm^{-1} along with the constants: $a = 0.055(2)$ and $b = -1.12(5)$. The graphical method of **Gallagher 1994**, to plot ($T_n + R_y/n^2$) against $1/n^3$, has also been applied to determine the series limit and quantum defect. The intercept of this line along the y-axis gives the value of the ionization potential as 43 487.25(2) cm^{-1} and the quantum defect = 0.051(5) is extracted from the slope = $-2R_y\mu$. The fittings of the Rydberg formula, Ritz formula and the graphical method to the term values of the newly observed levels are reproduced in figure 3.

The ionization potential and the corresponding constants using the Rydberg, Rydberg–Ritz and graphical methods are collected in table 2. Keeping all the possible inherent errors into account, the first ionization potential of lithium is reported as 43 487.26(3) cm^{-1} and the quantum defect for the np ^{2}P$_{1/2,3/2}$ series as 0.051(4). The value of the ionization potential

is 0.11 cm^{-1} higher than that reported by Lorenzen and Niemex (1983) and NIST database. These ionization limits and the quantum defect values are in good agreement, within the experimental error, with the earlier work by Johansson (1959), Gallagher (1994) and Goy *et al* (1986).

Figure 4 represents the signal intensities of the observed Rydberg series plotted against their effective quantum numbers. The solid line that passes through the experimental points is the fit of an equation $f_n = K(n^*)^{-\alpha}$. Here f_n is the ionization signal strength, K and α are constants. The value of α is expected to be equal to 3 as relative intensities in an unperturbed Rydberg series scale as $1/(n^*)^3$. The fit to the observed line intensities yields α equal to 1.79(4). Interestingly, Mende and Kock (1997) reported the *f*-values of the Rydberg series in strontium and noted that the *f*-values scale as $1/(n^*)^{2.5}$ instead of the $1/(n^*)^3$ scaling law. Since the lifetimes of the Rydberg series also scale as $1/(n^*)^3$, it will be interesting to inspect the behaviour of the lifetimes of the Rydberg levels. Theodosiou (1984) reported this anomalous behaviour in the lifetimes of the np ^{2}P$_{1/2,3/2}$ Rydberg series in lithium and potassium. The difference in the signal height scale may be attributed to the presence of the Cooper minima near the ionization limit.

In conclusion, using two-step laser excitation, we have been able to present new observations of the higher Rydberg members of the np ^{2}P$_{1/2,3/2}$ series that yield an improved value of the first ionization potential of lithium. The Rydberg series has been extended up to $n = 60$ and it is noted that the spectral line widths remain constant and the signal strength decreases as $(n^*)^{-1.8}$.

Acknowledgments

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