

Lithium isotope separation with tunable diode lasers

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A laser-isotope-separation study of lithium has been performed with two-step excitation involving UV laser radiation and a visible tunable-diode laser. The method yields a high degree of selectivity by tuning the narrow-linewidth diode laser to the D_1 or D_2 levels of the lithium atom. Selective laser excitation is simplified by the use of the tunable diode laser and the overall approach benefits from the application of a compact mass selector that includes a precision magnetic sector and an ion beam that is designed specifically for light atoms such as lithium. © 2002 Optical Society of America
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1. Introduction

Lithium isotopes are important for fission and fusion reactors. The laser-isotope-separation (LIS) method is considered, in the literature, to be very attractive owing to the high selectivity that can be achieved.^{1–6} This approach applies a two-step selective photoionization method and can be used for nearly all of the elements of the periodic table with commercial tunable lasers.⁷ It can also be used for ultra-sensitive trace-element analysis.⁸

In this paper we present a study of LIS in lithium with simple, compact, and inexpensive tunable diode lasers, which offer excellent spectral characteristics. The overall method allows the complete separation of the different isotopes, even in the case where the lines of the different isotopes overlap.

An integral component of the experimental method is a mass selector that includes a magnetic sector. A fairly detailed description of this relatively simple, compact, and inexpensive apparatus for separating lithium isotopes is given in this paper.

Tunable lasers that are useful to this type of application include the cw dye lasers⁹ and narrow-

linewidth copper-vapor-laser (CVL) pumped-dye lasers.^{10–13} The former type of dye lasers are relatively complex and require fairly sophisticated engineering for building and maintenance. One advantage is that they can yield relatively high CW powers in a single-longitudinal mode. Even higher average powers are available from narrow-linewidth CVL-pumped pulsed-dye lasers. However, this class of laser, although very desirable for this type of application, has been demonstrated and operated only in a handful of laboratories around the world.^{10–14} An incipient alternative is to use tunable external-cavity semiconductor lasers.^{15,16} These lasers are relatively inexpensive and compact, and yield narrow-linewidth single-longitudinal-mode emission. Given our limited resources and the exploratory nature of the experiments, these compact coherent sources are very well suited.

It should be made clear that the experimental approach described here was designed specifically for the selective excitation of light atoms for spectroscopic applications. The scaling of the method using high-power tunable lasers was beyond the scope of this study.

2. Background

The upper limit for the number of ions produced by the lasers at the end of a laser-ionizing pulse can be obtained with the result of an absorption measurement¹⁷

$$N_i = \alpha P_i P_e (1 - T) / A, \quad (1)$$

where $\alpha = \sigma_i T_{UV} / (h^2 v_e v_i) = 17100 \text{ m}^2/\text{W}^2$, P_i is the average UV laser power, P_e is the exciter power, T is the exciter transmittance at the coincidence area A of both lasers in the interaction region, $\sigma_i = 7 \times 10^{-22}$

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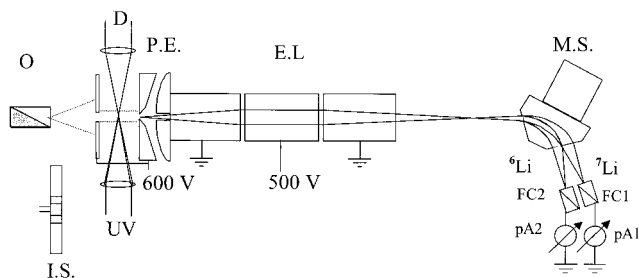


Fig. 1. Diagram of the mass separator. O, Molybdenum crucible with lithium; P. E., Pierce extractor; E. L., Einzel lenses; M. S., magnetic sector; FC1, FC2, Faraday cups; pA1, pA2, picoamperimeters; I. S., lithium ion source. The lithium-ion source can be placed behind the Pierce extractor for nonselective ion production and calibration of the mass separator.

m^2 is the ionization cross section,⁷ T_{UV} is the period of the UV laser pulses of the ionizing laser, h is the Planck's constant, and v_e , v_i are the excitation and ionization frequencies, respectively. The density n of the different isotopes can be obtained with the integral of the absorption coefficient over each line as described elsewhere¹⁸ when the laser intensity is sufficiently low.

3. Experimental Details

The schematics of the experimental set up is depicted in Fig. 1. A beam of lithium atoms enters an optical cell through an entrance aperture. At the optical cell an excitation volume is created by focusing two transverse laser beams of different frequencies, which provide the two-step excitation energy necessary for selective photo ionization. Specifically, the lithium beam is illuminated by a focused UV laser beam and a spatially coincident focused tunable-diode laser. Following the illumination volume the doubly-excited lithium atoms enter a Pierce extractor and proceed to a set of Einzel lenses. The divergent doubly-excited lithium beam then enters a magnetic sector of a mass selector. At this stage the atom beam is separated into two subbeams corresponding to the ⁶Li and ⁷Li isotopes. Each of the isotopic subbeams continues to a separate Faraday cup, where the picocurrent is measured.

A. Laser System

The CW tunable-diode laser used in these experiments was a commercial device (EOSI Model 2010, Environmental Optical Sensors, Inc., Boulder, Colorado 80301) configured in a Littrow grating cavity.^{15,16} The tuning range of this laser is 25 nm centered at 672 nm. This laser emits in a single-longitudinal mode at a linewidth of <100 kHz. The beam divergence is diffraction limited at an output power of 9 mW. This laser was tuned with an electronically controlled servomechanism that rotates the grating. This servomechanism includes a piezoelectric transducer driven by a slow triangular wave generator (HP Model 3310B, Hewlett-Packard Corp., Englewood, Colorado 80155). The time to scan one

complete spectrum was about 15 minutes in order to get enough resolution at the ionization spectra. The emission wavelength was monitored with an optical wavemeter (Burleigh Model WA4500, Burleigh Instruments, Fishers, New York 14453). As noted by previous authors¹⁵⁻¹⁸ tunable semiconductor lasers are ideally suited for this type of spectroscopic task, given their remarkable stability and the absence of thermal and other media-related instabilities that tend to introduce short-term wavelength drifts that require sophisticated control systems.^{13,14}

The tunable-diode laser was focused into the neutral lithium beam with a $f = 0.25$ m lens, while for ionization of the excited atoms we focused the fourth harmonic of a Nd:YAG laser¹ (Lee Laser Model 815TQ, Lee Laser, Inc., Orlando, Florida 32809) deployed in the counter-propagating direction with a fused silica $f = 0.58$ -m lens. Typical average power densities of the exciter and ionizer at the focus were 35 W/cm² at 671 nm and 125 W/cm² at 266 nm, respectively. The Nd:YAG laser has a 5-kHz repetition rate with a KTP* intracavity crystal to produce the green output. We used a temperature-stabilized KD*P crystal (Inrad Model 5-301, Inrad, Northvale, New Jersey 07647) to produce the FHG at 266 nm and a dispersive quartz prism to separate the green from the UV radiation. To increase the UV power density we used a $f = 0.2$ m lens to focus the green into the KD*P crystal and a $f = 0.2$ m fused silica lens to recollimate the beam. We measured the UV pulse length with a photodiode (EGG Model FND100Q, EG&G Optoelectronics, Canada, Vaudreuil, Quebec, J7V8P7, Canada) obtaining 80 ns (FWHM). We measured the focusing area of the red and UV laser using an ICCD (Model 576EMG/RB, Princeton Instruments, Trenton, New Jersey 08619) at different positions near the focal point. We obtained $A = (11.8 \pm 1.3) \times 10^{-9}$ m² for the red laser and $A = (16.8 \pm 0.7) \times 10^{-9}$ m² for the UV, respectively. In this case we are using only the 71% of the UV light for ionization.

B. Isotope Separation Apparatus

The beam of lithium atoms (Fig. 1) was produced through an evaporation of metallic lithium from a heat-pipe cell (Model HP-802, Comstock, Inc., Oak Ridge, Tennessee 37830). The heat pipe used here can reach a temperature of 800 °C with a stability better than 1 °C/min. A detailed description is given elsewhere.¹⁹⁻²⁰ One end of the heat pipe was closed, and the other end was opened and connected to a vacuum chamber containing a mass selector. The aperture used to collimate the beam has a 0.5-cm diameter. The collimator and the Pierce extractor²¹ were held at the same positive potential. The region between them is used as the laser-excitation volume region. The Pierce extractor yields a divergent ion beam that is focused with an Einzel lens system into the entrance of a mass selector. This mass selector was made in-house and is comprised of an ion gun and a magnetic sector.

To calibrate the mass selector we have used a lith-

ium ion cell (STD 250x, HeatWave Labs, Inc., Watsonville, California 95076), which is a ceramic beta eucryptite source containing a 30%/70% mixture of ${}^6\text{Li}$ and ${}^7\text{Li}$ isotopes. The construction and performance of these cells has been disclosed elsewhere.²² We also tested our system with pure ${}^6\text{Li}$ and ${}^7\text{Li}$ cells. The Pierce voltage V_p determines the ion-beam energy. Using the relationship that mass is proportional to the inverse of the applied voltage at a given magnetic field strength and geometry, we can estimate the required V_p . The most convenient value was obtained experimentally at 572 ± 1 V with a stabilized high-voltage power supply (Model EH05P20, Glassman High Voltage, Inc., High Bridge, New Jersey 08829) after focusing the beam at the entrance of the sector. To obtain a well-focused beam we have used a beam profile monitor (Model BPM80, Natural Electrostatics Corporation, Middleton, Wisconsin 53562) and adapted the length of the vacuum chamber to the position of the optimum focus. This focal point is 15 cm from the exit of the ion gun. The best focusing voltage V_f was held at 464 ± 1 V and was determined experimentally by the position of the entrance slit of the magnetic sector.

To measure the mass spectrum we employed a 2.5-mm section copper wire that was moved by a gear system connected to a rotary-motion feedthrough with a stepper motor (Model BRM275-03, MDC, Hayward, California 94545). The ion current reaching the wire was measured by a picoammeter (Model 485, Keithley Instruments, Inc., Cleveland, Ohio 44139) and recorded with a digital storage oscilloscope (Model 9314, LeCroy Corp., Chestnut Ridge, New York 10977). By using a mixed cell we were able to obtain a mass spectrum at one scan of the wire. The positions of the isotopes and the resolution were obtained from the barrel graduation. With this result we could replace the wire at the exit of the sector by two 9-mm-width copper plates separated by 1 mm. The size and position of the plates were determined from the mass spectrum. We adjusted the position of the plates by measuring the current with the ion cell again. In this manner we obtained a collector suitable for mass 6 and mass 7 isotopes. In the experiments, where the ions were produced by the lasers, the ion cell was removed and replaced by the neutral beam and collimator described above. The lasers were focused just behind the Pierce element. The rms value of the current at the ${}^7\text{Li}$ collector was measured with a picoammeter (Keithley Model 485) connected with a general-purpose interface bus to a personal computer and recorded with a Labview 5.0 application. The current of the ${}^6\text{Li}$ isotope was recorded simultaneously with the same software and interface with a more sensitive picoammeter (Keithley Model 595). The time required by our system to take the data for each pair of current values was 652 ms. The total number of ions produced at the ionization area were obtained from an absorption measurement and compared with the ions collected at both plates behind the magnetic sector. In this case the transmittance

was measured at saturation with the same laser intensity that was used at the current measurement. This was done at resonance and slightly off-resonance to avoid etalon effects.

To determine the density of neutral lithium atoms we removed the focusing lens and reduced further the intensity by means of a neutral density filter. The spectrum was recorded with an optical power meter (Model 1815C, Newport Corp., Irvine, California 92606) and a digital-storage oscilloscope (LeCroy Model 9314A). The background light was subtracted. The absorption path length was determined from fluorescence measurements.²³

C. Experimental Overview

The overall experimental setup described in the subsections 3.A and 3.B fits in the space provided by two 1.21×2.43 m commercial optical tables with a total utilized surface area of approximately 4.5 m^2 . The main two items contributing to this reduced area are the tunable-diode laser and the in-house mass separator. The tunable-diode laser is only a fraction of the size of an alternative cw dye laser or a CVL-pumped dye laser. The high-stability precision magnet comprising the mass selector was designed specifically for applications involving light atoms such as lithium. As such, it is only a small fraction of the size of a conventional commercial mass spectrometer. Ease of operation is a further experimental advantage.

4. Results

In these experiments a beam of lithium atoms is produced and illuminated by a two-step selective-laser excitation process. Following passage through a mass-selection apparatus two detectors, Faraday cup 1 and Faraday cup 2, are used to collect the spatially separated isotopes ${}^7\text{Li}$ and ${}^6\text{Li}$, respectively. Spatial separation is shown, through a mass spectrum, in Fig. 2. For our experimental conditions we have a resolution of $\Delta M/M = 3$, which is enough to separate the isotopes of interest.

The isotopic beam detected in Faraday cup 1 gives origin to the resonance ionization mass spectrum of the ${}^7\text{Li } D_1$ and ${}^7\text{Li } D_2$ resolved in doublets (Fig. 3). Note that this spectrum is clear, well resolved, and typical of only this isotope. The isotopic beam detected in Faraday cup 2 gives origin to the resonance ionization mass spectrum of the ${}^6\text{Li } D_1$ and ${}^6\text{Li } D_2$ lines (Fig. 4). Note that this spectrum is characterized by the lower intensity peaks, which correspond to this particular isotope exclusively. For comparison purposes the reader can observe the mixed, or combined, high-resolution spectrum of ${}^7\text{Li}$ and ${}^6\text{Li}$ given elsewhere.¹⁸

Albeit rather insignificant, the average value for the background signal was subtracted in each case from the measured spectra. This average background level was measured when the lithium was cold (heat-pipe cell off) obtaining 0.87 pA and 0.12 pA at the ${}^7\text{Li}$ and at the ${}^6\text{Li}$ collector plate, respectively.

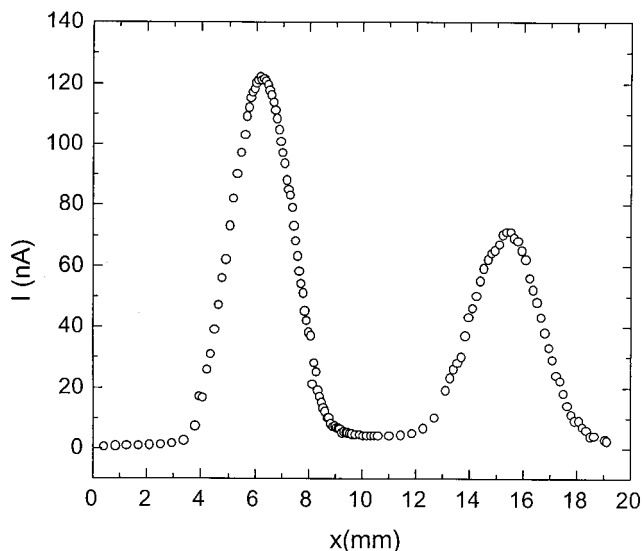


Fig. 2. Mass spectrum of mixed ${}^7\text{Li}/{}^6\text{Li}$ beta-eucryptite source.

This background level is deemed to have a negligible effect on the overall signal.

The hyperfine structure of the isotopes can be distinguished owing to a reduction in the Doppler width produced by the collimation and expansion of the vapor.²⁴ The transmittance T of each hyperfine line of the ${}^7\text{Li } D_2$ line was 0.991 and 0.996, respectively, and the ionization laser power was $P_i = (15 \pm 1)$ mW. In each case we considered the losses at the windows. For these parameters Eq. (1) can be used to estimate the upper limit for the number of ions. In this case those upper limits are estimated to be $N_1 = 8.56 \times 10^5$ and $N_2 = 3.83 \times 10^5$ at the peaks of the lines, respectively.

The density of the neutral lithium beam was determined from the absorption spectra. In this measurement the intensity of the laser used for excitation was kept low enough to avoid saturation effects and

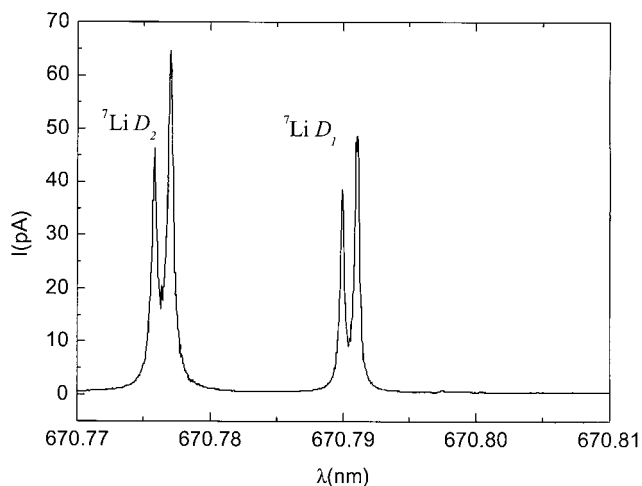


Fig. 3. Resonance ionization mass hyperfine spectrum recorded at the FC1.

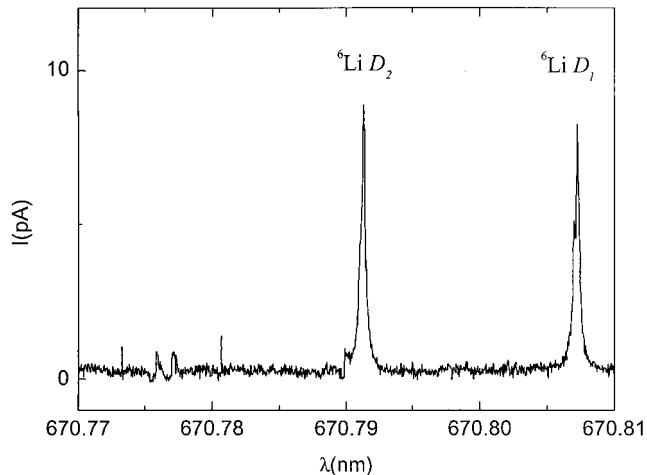


Fig. 4. Resonance ionization mass hyperfine spectrum recorded at the FC2.

optical pumping (1 W/cm^2 is convenient). At 780°C the density of the beam was $n = 2.5 \times 10^{16} \text{ m}^{-3}$ with a lithium-beam diameter of 0.005 m . The collimator installed behind the Pierce extractor has the important function of efficiently repelling the thermal ions arriving from the heat-pipe cell, which could contribute to an increase of the background signal at the spectra, giving a loss of selectivity. Selectivity also could be reduced by collision effects among atoms or ions as excitation transfer, but these effects are quite negligible owing to the low lithium density, which gives a mean free path $\lambda = 1/n\sigma$ of the order of 10 m or more, depending on the cross-section value of each collision process. The ionization by electrons can also affect the selectivity, but this effect is absent because the Pierce extractor repels the electrons of the beam. An additional nonselective effect is the direct ionization of the lithium clusters produced by the UV; this effect can be measured with the experimental arrangement of our previous work.¹⁷ This corresponds to 10^4 clusters/pulse or less. These clusters are filtered by the mass selector and do not contribute to the picocurrent signal.

In these experiments we observe a negligible background signal, and the spectral lines ${}^7\text{Li } D_2$, depicted in Fig. 3, and ${}^6\text{Li } D_1$ depicted in Fig. 4, appear free of the simultaneous signal from the other isotope. This indicates a high selectivity during the resonant ionization process.

5. Discussion and Conclusion

In these experiments we have recorded a high-resolution spectrum corresponding to ${}^7\text{Li}$ in one Faraday cup detector, whereas the spectrum corresponding to ${}^6\text{Li}$ was recorded in a second Faraday cup detector. These two detectors were separated spatially from each other. This spatial mass separation resulted from the selective two-step laser excitation, using a UV laser beam and a visible tunable-diode laser, of an atomic beam of lithium that propagated through a relatively simple mass selector. The in-

tensity ratio and wavelength characteristics of the two separated hyperfine spectra are consistent with known spectroscopic data.

To our knowledge this is the first report of the LIS of lithium utilizing a tunable diode laser. The application of this tunable-diode laser in conjunction with a simple, and compact, mass selector contribute significantly towards the ease of use and the overall compactness of the experimental apparatus for LIS experiments in light atoms.

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