An atomic beam of $^6\text{Li}$ – $^7\text{Li}$ for high resolution spectroscopy from matrix isolation sublimation

A N Oliveira$^1$, R L Sacramento$^2$, B A Silva$^2$, F O Uhlmann$^3$, W Wolff$^2$ and C L Cesar$^2$

$^1$ INMETRO, Av. Nossa Senhora das Graças, 50 25250-020 Duque de Caxias, RJ, Brazil
$^2$ Instituto de Física, Universidade Federal do Rio de Janeiro, Caixa Postal 68528, 21941-972 Rio de Janeiro, RJ, Brazil
$^3$ Beuth University of Applied Science, Zip Code 13353, Berlin, Germany

E-mail: anunes@inmetro.gov.br

Abstract. We propose the Matrix Isolation Sublimation (MISu) technique for generating cold lithium atoms for the measurement of the $^6\text{Li}$ – $^7\text{Li}$ isotope shift in D1 and D2 transitions. The technique is capable of generating cold $^6\text{Li}$ and $^7\text{Li}$ beams at 4 K with forward velocity of 125 m/s. Using this beam we offer a distinguished source of lithium atoms for transitions measurements, adding a new possibility to make high resolution spectroscopy towards improving the experimental checks of the theory.

1. Introduction
The energy structure of lithium can be solved from ab initio calculations and their predictions are compatible with experimental results. The fact that both results, experimental and theoretical, are within the same accuracy makes lithium atoms of great interest. In 2000 Yan and Drake [1, 2] proposed a combined theoretical and experimental approach to determine the relative nuclear charge radii based on the Splitting Isotopic Shift (SIS - difference for D1 or D2 line between different isotopes) (figure 1). Unfortunately, theoretical and experimental results reveals discrepancies as discussed in reference [3]. Since previous measurements where done using an effusive atomic beam at temperature around $s 400^\circ\text{C}$, including the recent one made in 2013 [4], we propose the use of a cold beam of Li atoms instead.

The present work shows that the MISu can produce simultaneous beams of $^6\text{Li}$ and $^7\text{Li}$. Cryogenic beams of Cr, $^7\text{Li}$, Ca, Cs and Li$_2$ molecules have been previously produced from the sublimation of Ne or H$_2$ matrices [5, 6, 7, 8, 9, 10].

2. Experimental setup
The basic experimental setup has been described in detail in [5] and a simplified diagram is shown in figure 2. The system consists of a closed-cycle cryostat that reaches 3 K at its cold plate with 1 W cooling power at 4 K. A sapphire substrate, with a deposited resistive NiCr film, is thermally connected, through a thermal link, to the cold plate. This configuration permits fast variations of the sapphire substrate temperature without affecting the cryostat temperature.

The gas to grow the matrix (Ne or H$_2$) is delivered through a 2 mm diameter tube whose exit points towards the sapphire substrate. The flow rate of the gas is controlled by the pressure in a
reservoir followed by a high impedance line, achieving 1–10 mmol/h. The growth of the matrix film is monitored by a CW laser beam – designated “longitudinal” – through the interference fringes (with 10–12% visibility) on the etalon formed by the Ne film together with the ~40% reflectivity of the NiCr resistive film on the sapphire substrate. This longitudinal beam comes from the bottom upwards, reflects off the Ne film and NiCr/sapphire mirror at near normal incidence, and is monitored by photodiode as indicated in figure 2. A typical time for growing a 330 nm thick Ne layer is around 40 s, but it can be reduced to less than 1 s. Once the cryostat starts cooling and cryopumping the vacuum line below 5 x 10⁻⁶ Torr the external vacuum pump line is closed.

After the matrix has been grown, or during its growth, a high-power pulsed laser (~10 mJ in 5 ns at 532 nm) promotes ablation of the solid precursor (Li or LiH), implanting the atoms into the matrix. Completed the matrix with the isolated atoms, we apply a heat pulse to the sapphire causing the matrix, together with the implanted atoms, to sublimate into vacuum. The atomic spectrum in flight is registered using the laser beam propagating in longitudinal and transverse direction, while scanning the laser frequency. The longitudinal beam is both co-propagating and counter-propagating with the sublimation plume. This leads to a splitting of a resonance line due to a positive and a negative Doppler effect, allowing a direct measure of the sublimation plume drift velocity (figure 5), as well as the sample’s longitudinal temperature. The other component from the same laser designated transverse, propagates horizontally (transverse to the propagation direction of the sublimation plume). The transverse beam is insensitive to the Doppler shift from the drift velocity of the plume while allowing for the characterization of the transverse temperature of the sample. In the data presented here the CW laser is tuned to the ⁷Li D1 and ⁶Li D2 transitions at 670.795 nm. It is scanned at 2 kHz covering 4 GHz around the center frequency which is enough to detect both transitions. The frequency scan is monitored by a Fabry-Perot interferometer allowing corrections on the non-linear scan due to the PZT response of the laser.

### 3. Experimental results

The scan, around the central frequency 446 800 000 MHz, allows to cover the transitions labelled 5–10 in figure 1 since the frequency range of those transitions span around 1 GHz. We have used a LiH substrate and the expected natural abundance between ⁶Li and ⁷Li is respectively 7.6% and 92.4% [12].

Figure 3 shows a typical transverse absorption signal where the atoms transverse temperature

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**Figure 1.** Lithium 6 and 7 isotopes energy levels, frequency shift relative to the state center of gravity energy \( E_{cg} = \sum_F (2F + 1)E_F / \sum_F (2F + 1) \) are shown in MHz [11].

| State | Energy Shift (MHz) |
|-------|--------------------|
| ⁶Li   | +2.762             |
| ⁷Li   | +1.255             |
| ⁶Li   | -1.757             |
| ⁷Li   | +17.394            |
| ⁶Li   | +76.068            |
| ⁷Li   | +152.136           |

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\begin{align*}
E_{cg} &= \frac{\sum_F (2F + 1)E_F}{\sum_F (2F + 1)} \\
\end{align*}
\]
Figure 2. Schematics of a typical setup for MISu showing the cryostat with optical access, and the laser beams for absorption spectroscopy. The zoom in the central region shows the sapphire substrate with its NiCr film resistor and the deposited matrix of Ne. For the study of lithium, the atoms are implanted via laser ablation (dashed green line) on a solid Li or LiH precursor. The spectroscopy laser beams (solid red line and identified as $L_{\text{long}}$ and $L_{\text{trans}}$), propagate along or perpendicular to the direction of the sublimation plume expansion. The longitudinal laser is also used to monitor the matrix film thickness. The lasers’ transmissions through the system are monitored by photodiodes ($PD_{\text{long}}$ and $PD_{\text{trans}}$).

was 2.5 K. Using the frequency difference between transition lines 5 to 10 from literature [13] and estimating the relative intensity by considering the isotopes proportion, it was possible to adjust the lineshape demonstrating a clear presence of $^6$Li and $^7$Li.

The advantage of the present technique is based on its versatility of operation on the choice of parameters governing the three steps of the method: matrix formation, ablation and sublimation. In order to achieve the best beam configuration for the experiment several sets of parameters can be chosen until exhausting the freedom of the method. The present set of parameters is not necessarily the best configuration for a high resolution experiment, the beam condition can be improved and made to last longer, achieving a quasi-cw condition.

In figure 4, a lithium beam is observed in the 5 to 15 ms range with a temperature between 3 – 5 K and longitudinal velocity of 120 – 130 m/s. A neon matrix of $\sim$ 170 nm thickness was grown in 210 s and 200 ablation pulses were applied during its formation. After matrix completion with implanted atoms we sublimated it in a slow regime [7], using a heat pulse of 4.0 W for 15 ms. In this regime the matrix takes awhile to start the sublimation ($\sim$ 5 ms). After
Figure 3. Single scan using the transverse laser converted to optical density. The full absorption signal is composed by 6 gaussian distributions: 2 gaussians originate from the $^6$Li D2 transition – 5 and 6 (gray curves) – and 4 gaussians from $^7$Li D1 transition – 7, 8, 9, and 10 (dashed gray curves).

this time the beam reaches an almost constant optical density regime, until the heat pulse is finished. The temperature was extracted from both transverse and longitudinal signal while the forward velocity was derived from the longitudinal signal. In figure 5 only one scan is depicted for illustration.

Figure 4. Processed data with absorptions converted to optical density. The peaks appear as the laser is continuously scanned through the resonances.

4. Conclusion and prospects
We observed the creation of a cold and slow atomic beam of $^6$Li and $^7$Li, suitable for high resolution spectroscopy. Due to the flexibility of MISu technique we can further improve the beam properties, in particular, its lasting time can be increased. Transverse temperature can be also reduced if we use collimating orifices in the beam path. Our purpose is to use an optical frequency comb available in Inmetro in combination with this “MISu-beam” to measure the lithium isotopic shift. This proposal will be the first step of a joint project based on MISu technique and high resolution spectroscopy in Inmetro Optical Division (DiOpt). Another goal is placed on measuring the hydrogen 1S-2S transition.

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Figure 5. Processed data for a single scan of the longitudinal laser signal. Based on the Doppler shift the forward velocity is determined. Gaussian width is used to extract temperature. 12 gaussian distributions were used in this fit. The same parameters used in the analysis of figure 3 are used here to determine the two shifted distributions.

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