Nuclear Charge Radii of $^{6,8,9}\text{Li}$ Determined by Laser Spectroscopy

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The $2S \rightarrow 3S$ transition of $^{6,7,8,9}\text{Li}$ was studied by high-resolution laser spectroscopy using two-photon Doppler-free excitation and resonance-ionization detection. The hyperfine structure splitting and the isotope shift were determined with precision at the 100 kHz level. Combined with recent theoretical work, the changes in nuclear charge radii of $^{8,9}\text{Li}$ were determined. These are now the lightest short-lived isotopes for which the charge radii have been measured. It is found that the charge radii monotonically decrease with increasing neutron number from $^6\text{Li}$ to $^9\text{Li}$. PACS numbers: 32.10.Fn, 21.10.Ft, 21.60.-n

Laser spectroscopy is an important tool for model-independent determination of nuclear properties for short-lived isotopes, giving information on collective and single-particle effects [1, 2]. Nuclear spins and moments can be extracted from the hyperfine structure (HFS) in an optical transition and differences in nuclear charge radii $\delta\langle r^2\rangle$ can be derived from isotope shift (IS) determinations. The latter requires a separation of the effects caused by change in mass and in nuclear-charge volume when comparing the optical transition frequencies for two isotopes. For heavier elements the mass shift is large while the so-called volume or field shift is small: For Li the volume shift is only on the order of $10^{-4}$ of the total IS. In addition, electronic correlation effects have to be taken into account, the so-called specific mass shift, which could not be calculated until recently with sufficient accuracy for atomic systems with more than two electrons. Therefore, determinations of nuclear charge radii of very light atoms ($Z \leq 10$) were restricted to stable isotopes, either by use of electron scattering or by investigating muonic atoms [3], or to hydrogen and helium isotopes by optical spectroscopy.

Recently, it has become possible to perform highly accurate calculations for the three-electron system of neutral lithium [4–6] and to test these results by optical spectroscopy on the stable isotopes $^{6,7}\text{Li}$ [7], where comparison can be made with nuclear charge radii known from electron scattering. This new theoretical development now allows the evaluation of optical IS data and the determination of the charge radii of radioactive lithium isotopes with the prospect of reaching the neutron drip line at $A = 11$ and to determine for the first time the charge radius of $^{11}\text{Li}$, the most prominent neutron halo nucleus.

In order to do so we have developed a novel on-line laser and mass spectroscopic technique which combines extreme accuracy with high sensitivity. Here, we report the first application of this method to short-lived isotopes, namely $^{8,9}\text{Li}$, making these the lightest radioisotopes for which model-independent charge radii have been determined. A decrease in charge radii is observed with increasing mass number, and this is compared with the predictions of various nuclear models.

The experimental arrangement is shown in Fig. 1. The short-lived isotopes with half-life $T_{1/2} = 838\text{ ms}$ ($^8\text{Li}$) and $178.3\text{ ms}$ ($^9\text{Li}$) are produced at the on-line mass separator at GSI, Darmstadt, by directing an 11.4 MeV/u $^{12}\text{C}$ beam from the UNILAC onto a 100 $\mu\text{g/cm}^2$ tungsten target. The fast reaction products enter the hot ion source of the mass separator through a tungsten window and are stopped in a sintered graphite catcher. Atoms diffuse out of the catcher, are surface ionized and extracted through the阳极, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through the catcher, are surface ionized and extracted through...
a small hole. After acceleration to 44 keV the ions are mass separated and delivered to the experiment. Maximum yields were about 200,000/s for $^7$Li and 100,000/s for $^9$Li.

The ion beam from the mass separator is stopped in a thin graphite foil (80 $\mu$g/cm$^2$) that is heated to 1800-1900°C with a CO$_2$ laser (~4 W). The implanted lithium diffuses quickly out of the foil and drifts into the ionization region of a quadrupole mass spectrometer (QMS). Here, the Li atoms are laser-ionized, the ions are mass-analyzed with the QMS, and detected with a continuous dynode electron multiplier (CDEM). The overall excitation scheme was chosen to achieve the required resolution in the $^7$Li($^7$Li) laser, $^9$Li($^9$Li) transition. However, at typical dye laser intensities are needed to approach saturation in both steps of the process. Therefore both lasers are enhanced by a factor of ~100 within a resonant optical cavity. To simultaneously couple both lasers into the cavity, the resonator is locked to the Ti:Sa laser, while the dye laser is locked to the resonator. Hence, the dye laser frequency does not exactly match the resonance frequency of the $2s \rightarrow 3p$ transition. However, at typical dye laser intensities of ~17 W/mm$^2$ the $2p \rightarrow 3d$ transition was broadened to ~8 GHz, and the two $3d$ fine structure components overlap to form a peak with an ~1.2 GHz flat-top region. The free spectral range of the resonator is ~500 MHz and thus a lock point with good ionization efficiency can always be found. The final ionization step can occur by absorption of either a 610 nm or a 735 nm photon.

The Ti:Sa laser is stabilized by frequency-offset locking to a diode-laser reference system that is locked to an iodine line ($X^1\Sigma_u^+ \rightarrow BO^0_u$ R(114) 11-2, $a_1$) using frequency modulation saturation spectroscopy [9]. The beat frequency between the two lasers is detected with a 25 GHz bandwidth photodiode and serves as the basis for a servo-loop controlling the Ti:Sa laser frequency. Typical linewidths of the beat signal were ~1 MHz.

The following measurement procedure was used: The Ti:Sa laser was set to a desired frequency and the ion beam turned on for 10 s. Laser ionized atoms were detected during this period and for an additional 5 s ($^7$Li) or 1 s ($^9$Li). This additional time is larger than five half-lives of the respective isotope and allows decay of radioactive particles implanted into the CDEM. Typical scans recorded data at 1 MHz steps over ±15 MHz range about the expected resonance positions, while inter-peak regions were scanned in a single step without data acquisition to obtain better statistics for the splitting determinations. Reference spectra for both stable isotopes were regularly interspersed among the radioisotope measurements.

Figure 2 shows typical spectra recorded for the Li $2S \rightarrow 3S$ transition. (A) shows the overall structure for $^7$Li with two hyperfine transitions ($\Delta F = 0$, $F = 1, 2$). The fitting function is a sum of a Gaussian pedestal, corresponding to the absorption of two photons from the same direction, and a Voigt profile for the narrow component. The Doppler-free peak has a Lorentzian width of 3 to 5 MHz depending on laser power. Without saturation broadening a width of 2.7 MHz would be expected from the lifetime of the $3S$ state. The Gaussian component showed no power dependence and was on the order of 2 MHz, slightly larger than the typical laser linewidth.

![FIG. 2: Resonance ionization spectra of $^7,^8,^9$Li. (A) Overall spectrum and lineshape for $^7$Li. "Skip-step scans" for splitting determination of $^8$Li (B) and $^9$Li (C). The size of the skip steps are indicated at the breaks. The frequency axis is the beat frequency between the reference laser and the Ti:Sa laser with $\nu_{Ti:Sa} < \nu_{Ref}$ (A,B) and $\nu_{Ti:Sa} > \nu_{Ref}$ (C).](image-url)
The pedestal Gaussian width of \(~ 3\) GHz corresponds to the normal Doppler width constrained by the geometry of the interaction region. Figure 2(B) and 2(C) show scans for \(^{6}\)Li and \(^{4}\)Li, respectively. The solid lines are fits using the same line function as in Fig. 2(A) but with fixed width for the underlying Gaussian profile. To obtain IS values with an accuracy of a few 100 kHz, systematic effects on the line positions must be identified. Therefore, off-line test measurements with the stable isotopes \(^{6,7}\)Li were performed altering various operating conditions. These included intensities of the two excitation lasers, re-locking of the diode laser, and changing the iodine reservoir temperature. Only the intensity of the Ti:Sa laser was found to have a statistically significant influence on the center frequencies. To correct for this power shift, a photodiode was placed behind the end mirror of the cavity and transmitted light intensity was influence on the center frequencies. To correct for this enhanced Ti:Sa laser intensity (Fig. 3). In contrast, the frequency for tions were blue shifted (manifested as a decreased beat recorded during each scan. Spectra were taken with varying laser powers and observed absolute resonance positions were blue shifted (manifested as a decreased beat frequency for \(^{7}\)Li) with linear dependence on the cavity-enhanced Ti:Sa laser intensity (Fig. 3). In contrast, the observed splittings (HFS and IS) were unchanged. The slopes, corresponding to AC-Stark shift coefficients, were found to be equal (within fitting uncertainty of \(~ 3\)% for \(^{6}\)Li, \(^{7}\)Li, and \(^{8}\)Li. A similar dependence is expected for \(^{9}\)Li. The observed HFS splitting \(\Delta \nu_{2S-3S}\) in the transition can be combined with previously measured ground state (gs) splittings \(\Delta \nu_{2S}\) from literature [11–13] to calculate the \(A\) factor of the \(3S\) state:

\[
A_{3S}(iLi) = (\Delta \nu_{2S-3S} - \Delta \nu_{2S}^i)/(I_i + 1/2),
\]

where \(I_i\) is the nuclear spin of the isotope \(i\). Results are listed in Tab. I. The \(^{6,7}\)Li values agree well with previous measurements [7], while the \(^{8}\)Li values have been determined for the first time. In the case of \(^{9}\)Li the accuracy is limited by the rather large uncertainty of the ground state hyperfine splitting [13]. Therefore the \(^{9}\)Li gs splitting was calculated from our results and the gs splitting of \(^{9}\)Li assuming no hyperfine anomaly. The result of 848.28(17) MHz was then used to calculate the \(A\) factor for the \(3s\) state listed in Tab. I. The given uncertainty allows for a possible hyperfine anomaly of \(~ 10^{-4}\) in addition to the measurement uncertainty.

To determine isotope shifts, the center of gravity (cg) of the HFS transitions was calculated using \(\nu_{cg} = (C_F \nu_F - C_F' \nu_{F}')/(C_F - C_F')\), where \(C_F\) and \(C_F'\) are Casimir coefficients for states with total angular momentum \(F\) and \(F'\). For each \(^{6,8,9}\)Li measurement the laser power was determined and the \(^{7}\)Li reference position at this power was calculated from the linear AC Stark shift function shown in Fig. 3. The obtained IS \(\Delta \nu_{exp}\) showed no systematic trends when plotted against time or laser power. Thus, the final IS values listed in Tab. II are the statistically weighted average of all measurements with uncertainties given as the standard error of the mean for sets of 62 \(^{6}\)Li and 104 \(^{8,9}\)Li measurements. An additional uncertainty of 110 kHz has been added linearly to allow for systematic error in the AC-Stark shift correction. The IS for \(^{6}\)Li agrees within 1.3\(\sigma\) with a previous measurement on this transition [7]. The change in nuclear charge radii is related to the measured IS by [4]

\[
\delta (r^2)_{A,7} = \frac{\Delta \nu_{exp}^{A,7}}{\Delta \nu_{MS,abeo}^{A,7}} C,
\]

where \(\Delta \nu_{MS,abeo}\) are the calculated mass shifts and \(C = -1.5661\) MHz/fm\(^2\) is the field shift constant for the Li

<table>
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<th>(^{6})Li</th>
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<th>(^{9})Li</th>
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<td>35.490(28)</td>
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<tr>
<td>35.263(15)</td>
<td>93.106(11)</td>
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<td>[7]</td>
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<tbody>
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<td>0.79(25)</td>
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<td>9 Li</td>
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<tr>
<td>8 Li</td>
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<td>-0.78(17)</td>
<td>2.22(9)</td>
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2S \to 3S \text{ transition taken from [5]. Results for } \delta \langle r^2 \rangle \text{ are included in Table II. For the stable isotopes } \delta \langle r^2 \rangle^{7,6,7} = 0.61(11) \text{fm}^2 \text{ agrees with a previous determination using the same transition } (0.47(5) \text{ fm}^2 [7]) \text{ within the combined uncertainties and is also in agreement with the electron scattering result of } 0.79(25) \text{ fm}^2 [14].

The rms charge radii \( r_{\text{charge}} \) were calculated according to
\[
 r_{\text{charge}}(A \text{Li}) = (r_{\text{charge}}(7 \text{Li}) + \delta \langle r^2 \rangle^{A,7})^{1/2},
\]
where \( r_{\text{charge}}(7 \text{Li}) \) obtained from electron scattering [14] is used as the reference. Results are plotted in Fig. 4 and compared with various theoretical predictions. A clear trend to smaller charge radii with increasing neutron number can be observed. Stochastic variational (SV) calculations [15] and Quantum Monte Carlo calculations (QMC) on light nuclei [16, 17] clearly reproduce this trend and also the absolute values are in good agreement with our results. Large-basis shell-model (LBSM) calculations, besides underestimating the absolute size, expect a minimum of the charge radius at \( A = 8 \) [18]. The dynamic-correlation model (DCM) reproduces the charge radii of the two stable nuclei quite well but the radius for \( 9 \text{Li} \) is predicted to be larger than that of \( 7 \text{Li} \) [19]. Neither of the latter two theories is in accordance with our results. Finally, combining the measured shifts with the absolute frequency of the reference iodine line yields a total transition energy of 27.206.0941(2)(13) \text{ cm}^{-1} \text{ for the } 7 \text{Li} \text{ cg. This is in excellent agreement with the previously reported value of } 27.206.0942(1) \text{ cm}^{-1} [7] \text{ and a recent theoretical evaluation of } 27.206.0926(9) \text{ cm}^{-1} [6]. \text{ This result demonstrates the exactness of the atomic calculations which is crucial for extracting the Li charge radii.}

To conclude, we have demonstrated that high-resolution cavity-enhanced two-photon spectroscopy in combination with resonance ionization and mass spectrometry yields the required sensitivity for on-line experiments. The technique was applied to \( ^{8,9} \text{Li} \) and in combination with advanced atomic calculations the tiny volume effect could be extracted. A steady decrease of the charge radii with increasing mass number was observed, allowing discrimination between different nuclear models. The new method presented here will also allow a determination of the charge radius of \( 11 \text{Li} \) in the near future.

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\[ \text{FIG. 4: RMS charge radii for } ^{6,7,8,9} \text{Li: (+) this measurement with } ^7 \text{Li } r_{\text{charge}} \text{ from electron scattering as reference; (c) LBSM [18]; (△) QMC [16, 17]; (▼) SV [15]; (o) DCM [19].} \]