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Specific mass shift of potassium $5P_{1/2}$ state

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ABSTRACT

We have measured the isotope shift between ^{41}K and ^{39}K in the $4s_{1/2} \rightarrow 5p_{1/2}$ transition at 405 nm using saturation spectroscopy. Our measured isotope shift is 456.1 ± 0.8 MHz, implying a residual isotope shift (sum of specific mass shift and field shift) of −52.7 ± 0.8 MHz. We deduce a specific mass shift of −40 ± 5 MHz, which would imply that the $5p_{1/2}$ state has a considerably larger specific mass shift than the $4p_{1/2}$ state. We have in addition measured the $5p_{1/2}$ hyperfine splitting for ^{41}K.

1. Introduction

1.1. Isotope shifts

The isotope shift is the difference in frequency of the transition between the same pair of atomic states of two different isotopes of the same element. It is due primarily to the differing atomic mass and the finite density of the electron wavefunction at the nucleus [1]. The shift in transition frequency between any two states $\delta\nu$ for different mass numbers A', A can be written in terms of three components, the normal mass shift, specific mass shift and the field shift:

$$\delta\nu^{A,A'} = \delta\nu_{NMS} + \delta\nu_{SMS} + \delta\nu_{FS}.$$

The normal mass shift is a result of the difference in reduced mass between two isotopes due to their nuclear masses. With m the electron mass, M_1 and M_2 the nuclear masses of the lighter and heavier isotopes, and ν_1 the frequency of the transition in the lighter isotope, the normal mass shift is [1]

$$\delta\nu_{NMS} = \frac{m(M_2 - M_1)}{M_1(M_2 + m)}\nu_1.$$

For the $4s_{1/2} \rightarrow 5p_{1/2}$ transition in ^{39}K and ^{41}K, the normal mass shift is 508.8 MHz. Subtracting the exactly known normal mass shift from the measured isotope shift leaves the experimentally measured quantity, termed the residual isotope shift, $\delta\nu_{RIS} = \delta\nu_{SMS} + \delta\nu_{FS}$.

The residual mass shift consists of two terms of quite different origin. The specific mass shift $\delta\nu_{SMS}$ comes from the change in reduced mass due to the correlated motion of the electrons in the atom. It is proportional to the two-body momentum correlation $\langle\sum_{i>j}\mathbf{p_i}\cdot\mathbf{p_j}\rangle$, where the sum is over all electrons in the atom. To calculate $\delta\nu_{SMS}$ is a difficult many-body calculation. There is considerable interest in $\delta\nu_{SMS}$ in many atoms because it is needed to extract the field shift, which allows the extraction of differences in nuclear charge radii from optical isotope shifts [2]. For example, $\delta\nu_{SMS}$ has been calculated very accurately in the lithium atom three-body system [3,4], but such calculations for atoms with more than three electrons involve considerable difficulties. Systematics of isotope shifts in various levels of potassium were explored in Ref. [5]. In the lithium atom, the specific mass shifts for excited states are found to decrease smoothly with increasing principle quantum number [1]. Note that unlike the normal mass shift, the specific mass shift does not simply scale with the transition frequency. The two-body momentum correlations are better thought of as producing an energy shift of the individual states.

The specific mass shift of the $4s_{1/2} \rightarrow 4\ p_{1/2}$ transition is not completely understood. An empirical value of $\delta\nu_{SMS}$ of 19.9 ± 4.9 MHz has been derived from comparison of the measured optical

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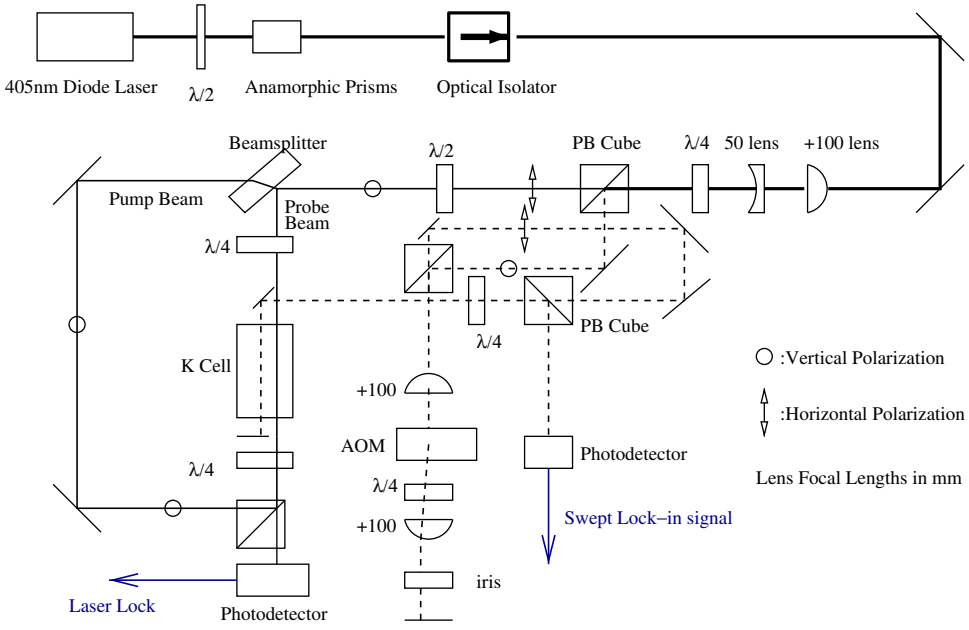

Fig. 1. Dithered saturation spectroscopy setup. The solid line shows the pair of pump and probe beams to whose signal the laser is frequency-locked. The dashed line shows the pair of pump and probe beams which are offset by a double-pass AOM (acousto-optic modulator).

isotope shifts with muonic X-rays [6] to assist with the extraction of the charge radii of potassium isotopes. Calculations to 2nd-order in many-body perturbation theory have been made of the specific mass shift in potassium $4s_{1/2}$ and $4p_{1/2}$ states [1,6]. There was good agreement for the 4s and 4p states, though for the 4p states this was regarded as fortuitous because several similar-sized terms cancelled to produce a smaller answer [1,6]. Similar many-body perturbation theory calculations have been done more recently to 3rd-order, producing $\delta\nu_{SMS} = 62$ MHz [7], and the authors of Ref. [7] conclude that a complete calculation including all 4th-order terms would be needed. A recent more general formulism applied to many atoms using a different basic approach with hoped-for better convergence and done to 2nd-order in many-body perturbation theory produces a calculated value and error of $\delta\nu_{SMS} = -2 \pm 21$ MHz [8], in agreement with experiment. The result is that $\delta\nu_{SMS}$ remains the largest systematic error in the charge radii of the chain of potassium isotopes.

No calculations have been made of the $4s_{1/2} \rightarrow 5\,p_{1/2}$ transition. Our measurement implies a considerably more negative value for this transition than for the $4s_{1/2} \rightarrow 4p_{1/2}$, and therefore a more negative value of the specific mass shift for the $5p_{1/2}$ state itself compared to the $4p_{1/2}$. Since the calculation of the $4p_{1/2}$ is difficult due to accidental cancellations of similar terms, we hope that our measurement of the $5p_{1/2}$ will be of some utility as an additional test for future calculations.

We note that a complete analysis would be helped by a measurement in ^{40}K, so that two pairs of isotope shifts could be used to construct a King plot [9]. However, the sensitivity of this work is inadequate to measure a 10^{-4} abundance isotope.

2. Experiment

We produce Doppler-free signals by standard saturation spectroscopy [10]. Two separate pairs of overlapping probe/pump beams originating from a commercial 405 nm diode laser traverse the same potassium spectroscopy cell (Fig. 1). Heating the cell to 70 °C produces sufficient potassium vapor density for absorption signals in this transition. One pumped probe signal is used to frequency-lock the laser to a ^{39}K transition. The other probe/pump pair is frequency shifted by an acousto-optic modulator, whose offset is precisely swept over the locations of other hyperfine-split lines to determine the isotope shift. The Doppler-broadened 1.5 GHz full-width-half-maximum background produces a small slope, which is recorded separately by misaligning the pump beam and subtracted off.

A Zeeman dither technique [11] is used to produce phase-sensitive detection signals with enhanced sensitivity. The current in a solenoidal coil wound around the cell is varied sinusoidally at 3 kHz, producing a varying field of approximately 1.0 G in the cell. (The background B field in the direction of laser light was measured to be less than 0.2 G.) The pump and probe beams are circularly polarized, so the resonant atomic frequencies are shifted sinusoidally as well, and a signal recovered by phase-sensitive detection.

The zero-crossing lock-in detector signal recovered from one pump–probe pair is passed through a commercial PID (proportional-integral-differential) [12] circuit to frequency-lock the laser to the crossover resonance [10] midway between the $F = 2 \rightarrow F' = 1$ and $F = 2 \rightarrow F' = 2$ lines. A typical lock-in detector signal from the

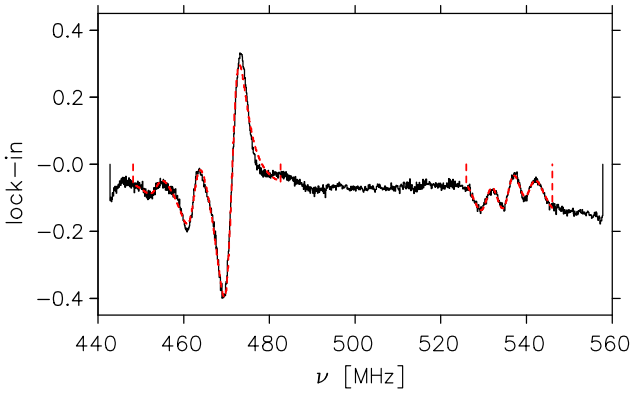

Fig. 2. Lock-in signals, showing transitions populated from ^{39}K $F_g = 1$ and ^{41}K $F_g = 2$ ground states. The solid histogram is the data, and the dashed line shows the fit and demarks the fitting region (see text).

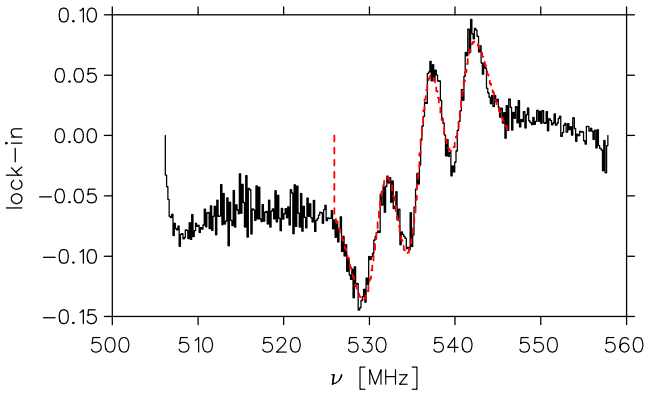

Fig. 3. Expansion of lock-in signal for transitions populated from the ^{41}K $F_g = 2$ ground state.

other pump–probe pair is shown as a function of the acousto-optic modulator (AOM) shift frequency in Fig. 2, as it is swept over ^{39}K and ^{41}K lines.

The lower-frequency lines shown in Fig. 2 are from the ^{39}K F = 1 to F′ = 1 and F′ = 2 real transitions and their crossover resonance. These are fit by the derivatives of three evenly spaced Lorentzian functions with common width, along with a linear background. The result is compared to the known ^{39}K ground-state hyperfine splitting [13] to accurately determine the frequency of the laser lock-point, which was 0.70 ± 0.35 MHz lower than the exact crossover peak frequency.

2.1. Experimental frequency shift and errors

The higher-frequency lines in Fig. 2 are from the ^{41}K F = 2 to F′ = 1 and F′ = 2 real transitions and their crossover resonance. Their location is similarly fit by derivatives of three Lorentzians and a linear background. The weighted average centroid of four fits (each a sum of 10 sweeps) to the ^{41}K F = 2 lock-in peaks was 535.69 MHz with variance 0.08 MHz (see Figs. 2 and 3).

Table 1
Sources of 1 σ errors in measurement of isotope shift.

Source of error	ϵ (MHz)
Variation between runs	0.08
Circular polarization sign	0.15
Lock point uncertainty	0.35
Subtraction of background	0.10
Opposite sweep directions	0.07
Total	0.75

Table 2
Summary of isotope shift values for previous and present results.

Results	$4s_{1/2} \rightarrow 4p_{1/2}$	$4s_{1/2} \rightarrow 5p_{1/2}$
$\delta\nu_{IS}$ [MHz]	235.27 ± 0.33 [18]	456.1 ± 0.8[e]
$\delta\nu_{RIS}$ [MHz]	−32.23 ± 0.33 [6,18]	−52.7 ± 0.8[e]
$\delta\nu_{SMS}$ [MHz]	−19.9 ± 4.9[a] [6]	−40 ± 5[e]
	−24[b] [1]	
	−62[c] [7]	
	−32 ± 21[d] [8]	
$\delta\nu_{FS}$ [MHz]	−12.9 ± 4.9 [6]	−12.9 [assumed]

[a] Experimental: comparison of optical isotope shifts and muonic X-rays.
[b] Second-order many-body perturbation theory.
[c] Third-order many-body perturbation theory.
[d] More general theoretical approach applied to many atoms.
[e] Present measurement.

We summarize the systematic errors in this measurement in Table 1.

We discussed above the lock point uncertainty. The result shifts by 0.1 MHz if we do not subtract the Doppler-broadened background, and we assign this as an uncertainty.

Changing the sign of circularly polarized light of the pump and probe beams produced a small variation of ∼0.15 MHz. This is within the variance of runs but possibly indicates a small constant B field producing a Zeeman shift, so we conservatively take that as an uncertainty.

The frequency was swept up and down to ensure that we were not exceeding the lock-in detection bandwidth, and we take the difference of 0.07 MHz as an uncertainty.

These errors are not necessarily random, and we conservatively add them linearly to produce a total uncertainty of 0.75 MHz [14].

2.2. Isotope shift result

Using literature values of 230.86 MHz and 127.006934 MHz for the ground state $A_{s1/2}$ hyperfine splitting constants of ^{39}K and ^{41}K $A_{4s1/2}$ (summarized in [13]), we deduce an isotope shift between ^{41}K and ^{39}K of $\delta\nu^{A,A'} = 456.1 \pm 0.8$ MHz for the $4s_{1/2}$ to $5p_{1/2}$ transition. Given the normal mass shift of 508.8 MHz, this implies a residual isotope shift $\delta\nu_{RIS} = \delta\nu^{A,A'} - \delta\nu_{NMS} = -52.7 \pm 0.8$ MHz. This constitutes our main experimental result.

2.3. Aside: hyperfine splitting of ^{41}K $5p_{1/2}$

Using the same error analysis, we measure the hyperfine coefficient $A_{5p1/2}$ for ^{39}K to be 8.93 ± 0.69 MHz. This is in agreement with measurements in the literature of 8.99 ± 0.15 MHz [15] and 9.30 ± 0.5 MHz [16] (summarized in [13]).

We measured $A_{5p1/2}$ for ^{41}K to be 4.96 ± 0.17 MHz, a previously unmeasured quantity. This is in agreement with the result from scaling $A_{p1/2}$ for ^{39}K by the nuclear magnetic moments, i.e. as would be expected, there is no evidence at this accuracy for a hyperfine anomaly [17].

2.4. Specific mass shift

From our measured residual mass shift, we can then make simple extrapolations of the known field shift of the $4s_{1/2} \rightarrow 4p_{1/2}$ transition to deduce the specific mass shift of the $4s_{1/2} \rightarrow 5p_{1/2}$ transition.

The field shift for the $4s_{1/2} \rightarrow 4p_{1/2}$ transitions has been extracted in [6]. The field shift scales with the finite density of the electron wavefunction at the nucleus, and Table 2 of Ref. [6] shows their calculated result that the contribution from the $4p_{1/2}$ state is approximately 5% of that of the $4s_{1/2}$. It is therefore a good approximation for us to assume that the field shift is the same for the $4s_{1/2} \rightarrow 5p_{1/2}$ and $4s_{1/2} \rightarrow 4p_{1/2}$ transitions. We then can deduce from our measurement of $\delta\nu_{RIS}$ our result for the specific mass shift $\delta\nu_{SMS} = -40 \pm 5$ MHz for the $4s_{1/2} \rightarrow 5p_{1/2}$ transition. The error is dominated by the uncertainty in the specific mass shift for the $4s_{1/2} \rightarrow 4p_{1/2}$ transition. The result is a factor of two more negative than the specific mass shift for the $4s_{1/2} \rightarrow 4p_{1/2}$ transition.

This is our main derived result. Since the $4s_{1/2}$ state is shared by the two transitions, it implies that the specific mass shift of the $5p_{1/2}$ state is considerably more negative than that of the $4p_{1/2}$ state.

We summarize previous results and our present results in Table 2.

2.5. Supporting measurements at lower accuracy

We tested this result in two other ways. They have less accuracy, so we do not discuss them in detail, but they are in agreement with our $\delta\nu_{RIS}$ result above at ∼2 MHz accuracy.

2.5.1. Saturation spectroscopy of additional transition

Data obtained from the ^{41}K ground-state crossover enhanced absorption peaks was analyzed in a similar fashion. The signal/noise ratio was poorer because of poorer diffraction efficiency of the AOM in the 318–346 MHz range required, and the change in absorption is smaller. The isotope shifts calculated from this data were in agreement with our final result, though with error ~2 MHz, so we do not discuss them in detail.

2.5.2. Laser-cooled atoms

We have also tested this result using laser-cooled atoms. We frequency-locked the laser in the same way and sent the frequency-shifted beam (the dashed beam in Fig. 1) to irradiate atoms that were cooled by a magneto-optical trap (MOT) [19]. Atoms were probed with the MOT light off. The MOT laser beams were chopped on and off in a 40 μs duty cycle to minimize atomic motion, but the quadrupole field was left on. A uniform B field was applied with Helmholtz coils, with the MOT beams deliberately left unbalanced in intensity in this direction so the equilibrium position was not at zero field. The atoms were also spin-polarized by optical pumping on the $4s_{1/2}$ to $4p_{1/2}$ transition to help understand the magnetic field from the Zeeman shifts. The $5p_{1/2}$ population was measured as a function of the 405 nm AOM shift by photoionizing the atoms with a small pulsed 532 nm laser and accelerating the photoions to a microchannel plate (similar to [20]).

Our implementation of this technique has a number of systematic errors. The MOT quadrupole magnetic field makes Zeeman shifts in the lines depending on the atom location, as the cloud is large enough to sample different field strengths. The cloud position and state populations change with the optical pumping polarization. It was not practical to test systematics by trapping ^{39}K at the same time and measuring its resonances with respect to the laser lock. The photoionization laser can itself introduce AC stark shifts. The result for the shift was 456.6 ± 1.6 MHz (with an error dominated by systematics), in agreement with our result from saturation spectroscopy.

These errors are sufficiently small to support our main result, that the specific mass shift of the $5p_{1/2}$ state is considerably more negative than that of the $4p_{1/2}$ state.

3. Conclusion

We have measured the isotope shift of the $4\ s_{1/2} \rightarrow 5p_{1/2}$ transition at 405 nm in potassium to be $\delta\nu^{A,A'} = 456.1 \pm 0.8$ MHz, implying a residual isotope shift to be $\delta\nu_{RIS} = -52.7 \pm 0.8$ MHz. From reasonable extrapolations of data and calculations in the literature for the $4s_{1/2} \rightarrow 4p_{1/2}$ transition, we have deduced the specific mass shift of the 405 nm transition $\delta\nu_{SMS}^{4S1/2\rightarrow 5P1/2} = -40 \pm 5$ MHz. This is considerably more negative than the literature value for the 770 nm transition $\delta\nu_{SMS}^{4s\rightarrow 5p1/2} = -19.9 \pm 5.1$ MHz [6]. Our result and those from the literature are summarized in Table 2.

Calculations in the literature of the specific mass shift of the $4p_{1/2}$ state involve cancellations between similar-sized terms, producing a result ≈20% the size of the largest contributions. (Examples include Table 7 of Ref. [6] and Table III of [7].) A small change in the degree of cancellation could account for the $5p_{1/2}$ result. We hope our measurement will provide an additional constraint for future calculations, which could in turn lead to more accurate extraction of the potassium nuclear charge radii.

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