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Measurements of the resonance lines of ^{6}Li and ^{7}Li by Doppler-free frequency-modulation spectroscopy

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Using Doppler-free frequency-modulation spectroscopy, we have measured most of the hyperfine components and the centers of gravity of the D_1 line of ^{7}Li and the D_2 line of ^{6}Li. By using previously measured fine- and hyperfine-structure splittings, we have also determined centers of gravity for the D_2 line of ^{7}Li and the D_1 line of ^{6}Li. Our present results are more accurate than previously reported values by a factor of more than 100. The wave numbers are ^{7}Li D_2: 14 903.983 468 (14) cm^{-1}; ^{7}Li D_1: 14 903.648 130 (14) cm^{-1}; ^{6}Li D_2: 14 903.632 116(18) cm^{-1}; ^{6}Li D_1: 14 903.296 792 (23) cm^{-1}. Values for the isotope shifts of the resonance lines of 0.351 338 (21) cm^{-1} for the D_1 line and 0.351 352 (15) cm^{-1} for the D_2 line have been derived. All uncertainties represent a 95% level of confidence.

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INTRODUCTION

In recent years there has been an increasing interest in precise theoretical calculation of binding energies and other properties of the ground and low-lying excited states of neutral lithium [1–4]. Calculated nonrelativistic energies have improved to the point where significant tests of theoretical three-electron QED corrections are becoming possible. For the lithium resonance lines, the accuracy of theoretical results is now within one order of magnitude of the accuracy of experimental observations. Precise new measurements will soon be required to test the rapidly developing theoretical methods.

As the strongest lines in the spectrum of neutral lithium, the resonance lines are of practical importance in spectrochemistry and solar [5] and stellar [6–8] astronomy. We have undertaken measurements for these lines as part of a larger effort to improve and summarize the knowledge of the spectra of ^{6}Li and ^{7}Li. This effort will result in more accurate wave numbers and energy levels and in more complete measurements of the isotope shifts. Although there have been many accurate measurements of fine- and hyperfine-structure intervals in ^{7}Li and somewhat fewer in ^{6}Li, until recently, the last measurement of the wave numbers of the resonance lines had been performed almost a half century ago.

The most accurate conventional measurements of the ^{7}Li resonance lines were reported in 1948 by Meissner, Mundie, and Stelson [9]. They were obtained with a lithium atomic beam excited by an electron gun and Fabry-Pérot interferometry. Because the interferometer was in air, it was necessary to apply an air dispersion formula to obtain the vacuum wave numbers. That formula was improved in 1953 [10], and applied to these lines as reported by Johansson in 1959 [11], who also reported other infrared data. The most accurate conventional measurement of the isotope shift was reported by Hughes in 1955 [12]. Previous wave-number measurements were reviewed in Refs. [9,11], and previous isotope shift measurements in Ref. [12].

With the advent of microwave, double resonance, and laser techniques, many results have been reported for the fine and hyperfine splittings for the resonance lines in both ^{6}Li and ^{7}Li, as well as the isotope shifts. The most accurate of these were summarized in 1990 [13]. However, the wave numbers of the lines themselves have not been remeasured except for two recent reports that did not substantially improve the earlier values. In 1986 Fuchs and Rubahn [14] reported wave-number values, calibrated with respect to calculated frequencies in the *X-A* band of ^{7}Li$_2$, which were about 10 cm^{-1} from the accepted values. More recently Windholz and Umfer [15] reported results using a laser-excited atomic beam coupled with a wavemeter. Their values were in disagreement with Ref. [11] and claimed only a marginally smaller uncertainty.

We have used Doppler-free frequency-modulation spectroscopy of natural lithium to measure the wave numbers of the resonance lines in both ^{6}Li and ^{7}Li. The source was a lithium beam, and the wave numbers were determined with a vacuum Fabry-Pérot interferometer. The results are presented in this paper along with a conversion to wavelength at standard temperature and pressure. Our values disagree significantly with those of Windholz and Umfer [15].

EXPERIMENT

The resonance lines of Li were observed by using Doppler-free frequency-modulation (FM) spectroscopy. This technique is well suited to the problem because it is a high-sensitivity direct absorption method that works well for the low density samples and low laser powers required to obtain narrow lines. In addition it produces an antisymmetric line

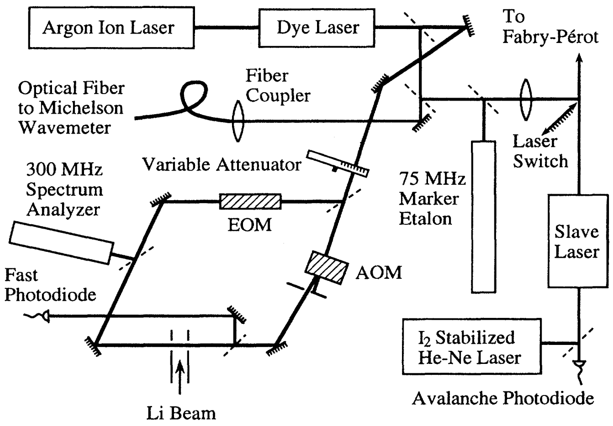

FIG. 1. Apparatus diagram for FM spectroscopy of the lithium resonance lines.

profile to which the exciting laser can easily be locked for measurement. The theory of FM spectroscopy and a full discussion of the line profiles obtained in this method have been given by Bjorklund *et al.* [16].

Our experimental apparatus, as set up at NIST, is shown schematically in Fig. 1. The ring dye laser, operated with the laser dye DCM, produces more than 250 mW of single-frequency light at 6707 Å with a linewidth of about 1 MHz as observed on an optical spectrum analyzer. The laser beam is split into pump and probe beams that counterpropagate coaxially through the lithium vapor. The beams are collimated and are approximately 2 mm in diameter. The frequency of the pump beam is shifted upward by 72 MHz and its intensity is sinusoidally modulated at about 80 kHz by an acousto-optic modulator (AOM). The frequency shift introduced by the AOM effectively isolates the dye laser from optical feedback due to the counterpropagating beams.

The probe beam passes through a traveling-wave electro-optic phase modulator (EOM) that creates symmetric sidebands around the laser center frequency. We adjust the rf drive power to the EOM to put 10% of the laser power in each sideband by observing the spectrum of the probe beam on a 300-MHz optical spectrum analyzer. During the course of the experiment sideband frequencies ranging from 6 to 120 MHz were used; however, all absolute measurements were made with a 20-MHz sideband interval. As the dye laser is scanned across a Li resonance, the sidebands probe the saturated absorption dip induced by the pump beam. The probe beam is focused on a fast photodiode and the resulting signal is processed by a low-noise current amplifier, a double-balanced mixer, and a lock-in amplifier to recover the antisymmetric FM signal. The phase of the local oscillator signal to the mixer is adjusted by a variable delay line to obtain an FM profile proportional to the dispersion of the spectral feature (S_2 in Fig. 3 of Ref. [16]). This profile has a steep zero crossing regardless of the relative sizes of the linewidth and sideband spacing and is therefore most suitable for locking the laser to the spectral line.

Unlike the heavier alkali metals, lithium vapor cannot be observed in a simple glass cell because the metal rapidly attacks the glass at the temperatures required to obtain sufficient vapor pressure. We have therefore produced lithium vapor in a weakly collimated atomic beam as illustrated in Fig. 2. Lithium metal is heated in a stainless steel oven chamber with typical operating temperatures of 450–520 °C. Lithium vapor drifts through the stainless steel chimney into the interaction region and condenses on the cool glass end of the apparatus, leaving the windows uncontaminated.

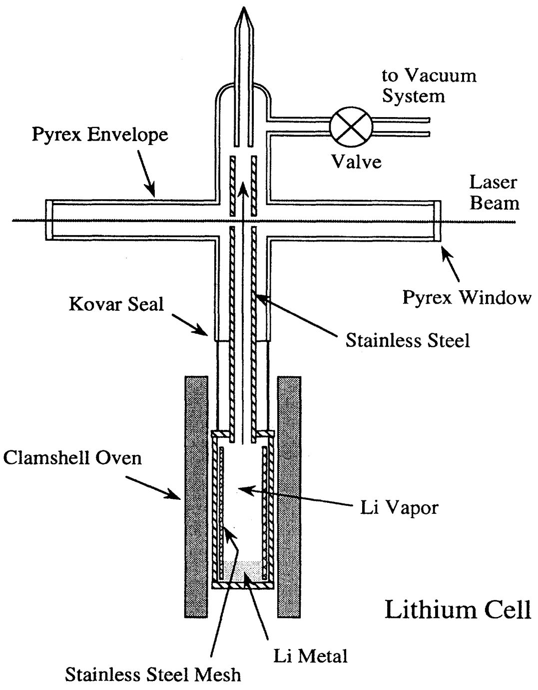

FIG. 2. Schematic of the natural lithium cell used for the experiment.

Using Doppler-limited absorption measurements we have characterized the Li vapor in the excitation region. The observed width of the Doppler profile was 600± 100 MHz. The calculated Doppler width for ^{7}Li at 773 K (500 °C) is about 3400 MHz. This suggests that the Li beam was substantially collimated by its passage through the stainless steel chimney and that the effective temperature for our observations was only about 25 ± 8 K. From the absorption depth the Li number density in the excitation region was determined to be $(1.3-3.9)\times10^{10}$ cm^{-3}. At this density the mean free path is in excess of 70 m. We therefore conclude that our observations were made in a collision-free environment.

Typical spectra are displayed in Figs. 3 and 4. We have made absolute measurements of the wave numbers of all well-defined components and crossover resonances in the observed spectra. Lines measured in this work are identified by alphabetic labels in the figures. The lithium transitions responsible for each observed line are also indicated in the figures.

Measurements for each line were made by servolocking the dye laser to the zero crossing of the FM signal. The wave number of the laser was then measured with a high-precision Fabry-Pérot wavemeter developed at NIST [17]. This instrument, which can be configured with Invar spacers of various lengths, analyzes spatially dispersed Fabry-Pérot interference

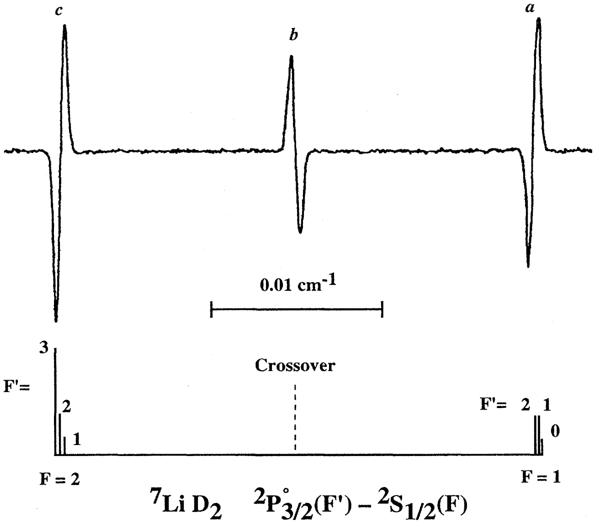

FIG. 3. Typical trace for the ^{7}Li D_2 line of natural lithium. Note the asymmetry of features *a* and *c* due to unresolved hyperfine structure.

fringes to determine the wave number of a tunable laser with respect to a He-Ne reference laser. To determine the integer order of interference the wavemeter needs initial values for the spacer length and tunable laser wave number. Each time the spacer is changed, auxiliary observations of well-known lines of uranium, thorium, and molecular iodine are used to determine its length within one-quarter of the wavelengths of interest. An initial value for the tunable laser wave number is provided by a traveling Michelson wavemeter. When configured with a 218-mm spacer, the Fabry-Pérot wavemeter has an accuracy of a few parts in 10^9. The reference laser for the wavemeter was stabilized to the *g* component of the $^{127}I_2$ transition $R(127)$ 11-5. Our laser follows the same design as that used by Jennings *et al.*, who determined the frequency of this line to be 473 612 340.492 (74) MHz [18].

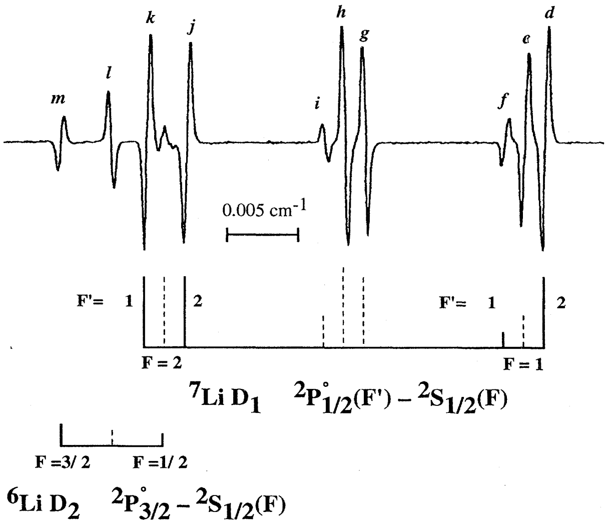

FIG. 4. Typical trace for the ^{7}Li D_1 and ^{6}Li D_2 lines of natural lithium.

Our results are derived principally from measurements made with a Fabry-Pérot spacer 218 mm in length. Every line was measured at least seven times with this spacer on different days with independent alignments of the apparatus to minimize the effect of any systematic errors in the measurements. Each measurement was the average of about 45 individual determinations from the Fabry-Pérot wavemeter taken over a period of about 1 min. The standard deviation of such a set of determinations was typically 5×10^{-6} cm^{-1} (0.15 MHz). Variation between independent measurements taken on different days was significantly larger.

Three systematic corrections have been applied to the data. First, the raw data were reduced by 2.1×10^{-6} cm^{-1} for ^{7}Li and 2.5×10^{-6} cm^{-1} for ^{6}Li to correct for photon recoil shift, which is not negligible for these light atoms. Second, the results were reduced by an additional 1.2×10^{-6} cm^{-1} to correct for the nonstandard temperature [18] and intracavity power [19] of our reference laser. Third, the results were corrected for the phase dispersion of the aluminum reflective coatings in our interferometer. To determine this correction we configured the Fabry-Pérot wavemeter with Invar spacers of 7, 11.4, and 218 mm lengths. Results from observations with the shorter spacers were used to derive the correction for change of phase on reflection as described in Ref. [20]. The correction applied, which was determined from the fully resolved spectral features only, amounted to $+0.001\,57(20)$ of an order, corresponding to a wave-number correction of $+3.6(5)\times10^{-5}$ cm^{-1}.

We have not attempted to correct the measurements for any shifts in the zero crossing that may be due to overlapping wings of neighboring transitions or crossover resonances. Computer modeling indicates that these shifts do not exceed 3×10^{-6} cm^{-1}, substantially smaller than our statistical uncertainty. The direction of the shift varies from line to line and its size depends significantly on the linewidth and local oscillator phase which can vary from measurement to measurement of the same line.

RESULTS

Hyperfine structure and crossover wave numbers

The vacuum wave numbers and uncertainties of the measured ^{6}Li and ^{7}Li resonance line hyperfine-structure components and crossovers are given in Table I. They are keyed to the features illustrated in Figs. 3 and 4. Because of the interference of some ^{6}Li D_2 transitions with ^{7}Li D_1 components, not all ^{6}Li D_2 components could be measured. No individual components of the ^{6}Li D_1 line were resolved sufficiently for measurement. We have made no attempt to work with the unresolved profiles that we observed since the signal-to-noise (S/N) ratio was relatively low for this weakest of the Li D lines.

The wave numbers shown in Table I are averages of many individual measurements using three spacers of length 7, 11.5, and 218 mm. For each line we determined the standard deviation of the result as the quadrature sum of the standard error of the mean, the estimated uncertainty attributable to baseline shifts from neighboring lines or electronic offsets (3.0×10^{-6} cm^{-1}), the uncertainty of the phase correction (5×10^{-6} cm^{-1}), the uncertainty of the reference laser wave

TABLE I. Measured wave numbers of hyperfine-structure components and crossover resonances in the Li D lines. F_L and F_U are the total angular momentum of the lower and upper states.

Line	Feature ID[a]	Classification	F_L	F_U	Wave number (cm^{-1})[b]
^{7}Li D_2	a	$2s\ ^2S_{1/2}$-$2p\ ^2P_{3/2}$	1	0,1,2	14 904.000 382(17)[c]
	b	crossover			14 903.986 875(21)[c]
	c	$2s\ ^2S_{1/2}$-$2p\ ^2P_{3/2}$	2	1,2,3	14 903.973 421(19)[c]
^{7}Li D_1	d	$2s\ ^2S_{1/2}$-$2p\ ^2P_{1/2}$	1	2	14 903.666 030(18)
	e	crossover			14 903.664 489(16)
	f	$2s\ ^2S_{1/2}$-$2p\ ^2P_{1/2}$	1	1	14 903.662 961(18)
	g	crossover			14 903.652 636(20)
	h	crossover			14 903.651 099(22)
	i	crossover			14 903.649 569(22)
	j	$2s\ ^2S_{1/2}$-$2p\ ^2P_{1/2}$	2	2	14 903.639 228(24)
	k	$2s\ ^2S_{1/2}$-$2p\ ^2P_{1/2}$	2	1	14 903.636 162(21)
^{6}Li D_2	l	crossover			14 903.633 385(22)
	m	$2s\ ^2S_{1/2}$-$2p\ ^2P_{3/2}$	3/2	1/2,3/2,5/2	14 903.629 580(25)

[a] Letter that identifies this spectral feature in Fig. 3 or 4.

[b] The reported uncertainty represents a 95% level of confidence.

[c] This measured wave number, although highly reproducible, is shifted from the true center of gravity due to unresolved hyperfine structure. See text.

number (2.3×10^{-6} cm^{-1}), and the uncertainty of the reference laser power and temperature correction (2.8×10^{-6} cm^{-1}). The reported uncertainty is twice the standard deviation, providing a 95% confidence interval.

Not all hyperfine-structure (hfs) components could be resolved due to the small intervals separating many of the components and crossovers and to the observed linewidth. The width was determined to be about 12 MHz by expanding the sideband spacing and measuring the full width at half maximum (FWHM) of the resulting undisturbed feature. In doing so, the local oscillator phase was changed to observe an FM signal proportional to the absorption rather than the dispersion of the spectral line (S_1 in Fig. 3 of Ref. [16]). The natural linewidth of the transition is about 6 MHz [21], and the dye laser width is about 1 MHz. We assume that the remainder of the width is due to power broadening.

Reduction of hfs to D_1 and D_2 line center-of-gravity wave numbers

To convert the component measurements to centers of gravity (CG) of the D_1 and D_2 lines we used previously measured fine- and hyperfine-structure intervals. Measured by optical–radio-frequency double resonance, level crossing, and atomic beam magnetic resonance methods, the intervals have been reported with extremely high accuracy [22–24]. Arimondo, Inguscio, and Violino [25] have published a critical review of these earlier measurements. The splittings are summarized in Table II, where the original values and uncertainties given in MHz have also been converted to cm^{-1}. An expression converting component or crossover to CG wave numbers was derived for each feature.

While reducing the measurements we checked our accuracy by comparing measured with known intervals. We found excellent agreement (better than 0.2 MHz) for all intervals except for the measurement of the ^{7}Li $2s\ ^2S_{1/2}$ hfs interval using features a, b, and c. Apparently the three unresolved hyperfine-structure components in each feature blended to give a resulting FM signal which was shifted to an undetermined degree. Some indication of this was observed because features a and (especially) c in Fig. 3 were more asymmetric than any of the fully resolved components in Fig. 4. Attempts to model these unresolved features did not resolve the 5-MHz disagreement between our measurements and more accurate previous determinations of this in-

TABLE II. Fine- and hyperfine-structure intervals used in analyzing the measurements.

Interval	Frequency (MHz)	Wave number[a] (cm^{-1})	Reference
^{7}Li $^2S_{1/2}$ $F=1-2$	803.504 086 6 (10)	0.026 802 011 36 (3)	[22]
^{7}Li $^2P_{1/2}$ $F=1-2$	91.828 (50)	0.003 063 1 (17)	[23]
^{6}Li $^2S_{1/2}$ $F=1/2-3/2$	228.205 259 0 (30)	0.007 612 108 07 (10)	[22]
^{7}Li $^2\mathrm{P}_{3/2}$-$^2\mathrm{P}_{1/2}$	10 053.184 (58)	0.335 338 1 (19)	[23]
^{6}Li $^2\mathrm{P}_{3/2}$-$^2\mathrm{P}_{1/2}$	10 052.76 (22)	0.335 324 (7)	[24]

[a] Converted from frequency, which was the reported quantity in the references.

TABLE III. Center-of-gravity wave numbers (cm^{-1}) of the lithium resonance lines.

Line	This work[a]	Johansson[b] (Ref. [11])	Windholz and Umfer (Ref. [15])
^{7}Li D_2	14 903.983 468 (14)	14 903.990 (3)	14 903.974 (2)
^{7}Li D_1	14 903.648 130 (14)	14 903.654 (3)	14 903.638 (2)
^{6}Li D_2	14 903.632 116 (18)		14 903.622 (2)
^{6}Li D_1	14 903.296 792 (23)		14 903.287 (2)

[a] The reported uncertainties represent a 95% level of confidence.

[b] Measurements of Meissner, Mundie, and Stelson [9] adjusted by Johansson using an improved air dispersion formula [10].

terval. As a result we decided to rely on our measurements for the ^{7}Li D_1 and ^{6}Li D_2 lines, and to obtain the CG values for the other two lines from known fine-structure splittings.

The CG value for ^{7}Li D_1 was obtained by averaging 58 independent values derived from measurements of features $d-k$, while the ^{6}Li D_2 CG was derived from 14 measurements of features l and m. The quoted uncertainty is twice the result of adding in quadrature the standard error of the mean and the uncertainties in the phase correction and reference laser frequency. To obtain the CG for ^{7}Li D_2 we used the ^{7}Li ^2P fine-structure splitting measured by Orth, Ackermann, and Otten [23]. The ^{6}Li D_1 CG was calculated by using the ^{6}Li ^2P fine-structure splitting measured by Brog, Eck, and Wieder [24].

We considered the possibility that Stark effect might shift the levels. A recent study of the Stark effect on the D_1 line of both isotopes [26] concluded that the scalar Stark shift was 4.62 kHz/(kV/cm)^2. A shift on the order of our uncertainties (420 kHz) would require an electric field of 10 kV/cm. The only external field source in our experiment was the oven, which was 10 cm from the volume in which the lithium vapor was interacting with the laser beam. We conclude that the measurements are not perturbed by Stark effect due to external electric fields.

The wave numbers and uncertainties of the D_1 and D_2 line centers of gravity, obtained as described above, are given in Table III, where they are compared with previous results. Our uncertainties represent a 95% confidence level. In frequency units the uncertainties range from 0.42 to 0.69 MHz. Our results are approximately 0.006 cm^{-1} lower than the values of Johansson [11] and about 0.010 cm^{-1} higher than those of Windholz and Umfer [15] and well outside the combined uncertainties in each case.

Air wavelengths for standard temperature and pressure were calculated using the three-term index-of-refraction formula of Peck and Reeder [27]. Twelve significant figures were carried because, at our level of accuracy, the index of refraction was slightly different for the four lines. The values of $(n-1)\times 10^8$ ranged from 27 607.712 for ^{7}Li D_2 to 27 607.680 for ^{6}Li D_1. The wavelengths, uncertainties, and comparisons with Refs. [11,15] are shown in Table IV. It should be noted that in Ref. [15] different wavelength values are given in the Abstract and in Table 2. We have compared our values with those in Table 2 since the Abstract values are clearly in error.

Although the exact origin of the discrepancy between our measurements and those of Windholz and Umfer [15] is not clear, several factors that may contribute to the disagreement can be identified. First, the measurements of Ref. [15] were made in air. To convert the results to vacuum wave numbers, the Cauchy formula [28] for the index of refraction of air was used. The ratio of the index of refraction at the reference laser wavelength to that at the lithium wavelengths for standard air is given in Ref. [15] as 1.000 000 383 84, which differs by five parts in 10^8 from the more accurate value 1.000 000 436 703 derived from the formula of Peck and Reeder [27]. Second, the conversion from air to vacuum was made for standard air, not air at the actual temperature and pressure in the laboratory when the measurements were made. This introduces an additional error of unknown size. Third, the frequency of the reference laser used in Ref. [15] may not have been accurately known. The frequency reported by Bay, Luther, and White [29] that was used in Ref. [15] differs by more than 520 MHz from the center fre-

TABLE IV. Center-of-gravity wavelengths (Å) of the lithium resonance lines in air at standard temperature and pressure.

Line	This work[a]	Johansson[b] (Ref. [11])	Windholz and Umfer (Ref. [15])
^{7}Li D_2	6707.763 741 (6)	6707.761 (1)	6707.7716 (5)
^{7}Li D_1	6707.914 670 (6)	6707.912 (1)	6707.9226 (5)
^{6}Li D_2	6707.921 877 (8)		6707.9298 (5)
^{6}Li D_1	6708.072 807 (10)		6708.0807 (5)

[a] Converted from the wave number by using the three-term air dispersion formula of Peck and Reeder [27]. The uncertainties given include no contribution from the air dispersion formula, the uncertainty of which is probably of comparable size.

[b] Measurements of Meissner, Mundie, and Stelson [9] adjusted by Johansson using an improved air dispersion formula [10].

TABLE V. Comparison of present and previous determinations of the transition isotope shift of the lithium resonance lines.

Frequency (MHz)	Wave Number[a] (cm^{-1})	Reference
10 533.3 (5)	0.351 352 (15)	this work (D_2 line)
10 539.9 (12)	0.351 573 (40)	[15] (D_2 line)
10 532.9 (6)	0.351 338 (21)	this work (D_1 line)
10 534.3 (3)	0.351 386 (10)	[15] (D_1 line)
10 532 (5)		[31]
10 534.8 (20)		[14]
10 520 (20)		[32]

[a] The reported uncertainty for this work represents a 95% level of confidence.

quency found by Niebauer *et al.* [30] for a commercial polarization-stabilized He-Ne laser. This large difference is probably a consequence of different isotopic composition of the neon in the laser gain tubes. Reference [15] gives no evidence that the frequency of Bay, Luther, and White [29] is actually applicable to the laser used in the experiment.

Isotope shift

Values of the transition isotope shift can be obtained for both the D_1 and D_2 lines. It is well to remember that the isotope shift values depend not only on our measurements but also on the fine-structure splittings used to obtain the wave numbers we did not measure. Table V contains the results and comparisons with previous measurements. The uncertainties given are based on random sources of error only, since possible systematic errors in the reference laser wave number and the phase dispersion correction cancel in the calculation of the shifts.

Our isotope shifts are in good agreement with those of Refs. [14,31] but outside the combined uncertainties when compared to the values of Ref. [15]. Unlike Ref. [15], our results do not show a significant difference between the isotope shifts for the D_1 and D_2 lines. We note that the ^{7}Li fine-structure interval of Ref. [15], which is completely contained in the spectral region measured to obtain the D_2 line isotope shift, is about 3.4 MHz larger than other much more accurate measurements of this interval [23]. This suggests that the isotope shift for the D_2 line may well be in error by significantly more than the reported 1.2-MHz uncertainty. Although our results for the D_1 and D_2 lines differ by only about 0.000 014 (13) cm^{-1} (one standard deviation level uncertainty), we have not averaged the results since theory suggests that the difference in the isotope shifts for the two D lines should be on the order of 1 MHz [33,34].

CONCLUSION

We have made the most accurate measurements to date of the centers of gravity of the ^{7}Li D_1 and ^{6}Li D_2 transitions. Our 95% confidence interval is more than 100 times smaller than the estimated uncertainty of Ref. [15]. By combining our measurements with well-known splittings we have calculated the centers of gravity for ^{7}Li D_2 and ^{6}Li D_1 with similar accuracy. This has led to more precise wavelengths for all four lines and to a new determination of the isotope shifts of the resonance lines.

Note added in proof. Subsequent to the submission of this manuscript we were advised by Dr. Windholz that a revision of the results of Ref. [15] is being prepared for publication.

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