Precise measurement of the Stark shift of the lithium $D1$ line

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The dc Stark shift of the lithium $D1$ line has been observed using two visible diode lasers and an atomic beam. A heterodyne signal obtained from the two lasers permits a well-calibrated, high-precision measurement to be made. The scalar shift is found to be 4.6216(21) kHz/(kV/cm)^2. We believe this result to be the most precise Stark shift measurement reported thus far.

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Multiconfiguration Hartree-Fock (MCHF) and many-body perturbation-theory (MBPT) calculations are now yielding levels of precision that were unthinkable only a decade ago [1–3]. In addition to the importance of this achievement to atomic physics, the success of these models has allowed parity violation experiments [4] to place increasingly tight constraints on various particle theories as well [5]. These calculations have been particularly successful in light atomic systems. Indeed, the most recent calculations have achieved agreement with the known energy levels of lithium at the 0.01% level. Unfortunately, there have been no experimental measurements that adequately test the behavior of the predicted atomic wave functions at large values of r. Oscillator strengths and atomic lifetimes, the traditional tests of the large-r behavior of the wave functions, are quite difficult to measure with a precision of much better than 1%. In response to this need for meaningful tests of the wave function away from the nucleus we have developed a new technique for measuring atomic Stark shifts that yields unprecedented levels of precision. Stark shifts not only test the large-r behavior of the atomic wave functions, but also provide a precision test of the methods used to sum the infinite series so often encountered in atomic perturbation theory. These summation techniques also play a crucial role in the prediction of atomic parity violation and of various Stark-induced interference processes.

Conceptually, our new method of measurement is quite simple. An atomic beam is used to provide narrow atomic resonance lines. Two lasers are locked onto different hyperfine transitions of the resonance line of interest in two separate regions. In one of these regions a well-calibrated electric field may be applied. Light from the two lasers is mixed on a fast photodiode and the difference frequency between the two lasers is monitored. When a field is applied to one of the two regions the atomic resonance shifts, dragging the laser frequency along with it. By measuring the change in the beat frequency when the field is turned from on to off the Stark shift is determined.

Because of its importance as a test of MCHF and MBPT theories we have chosen to demonstrate our new method for measuring Stark shifts on the $D1$ line of lithium. The basic apparatus is sketched in Fig. 1. The lithium atoms emerge from a 1-mm-diam hole in a stainless-steel oven, maintained at 500 °C. The atomic beam is defined by a 4-mm-wide 3-mm-high aperture 1 m downstream from the source. Approximately midway between the source and detection regions a baffle has been placed to allow differential pumping. The pressure in the experimental region is typically 5×10^{-7} Torr. The atomic beam passes through two interaction regions, each with four viewing ports that permit optical access. The second interaction region has a pair of high-voltage electrodes suspended within it. The two interaction regions are separated by a metal baffle so that the application of an electric field in the "field region" will not create any field

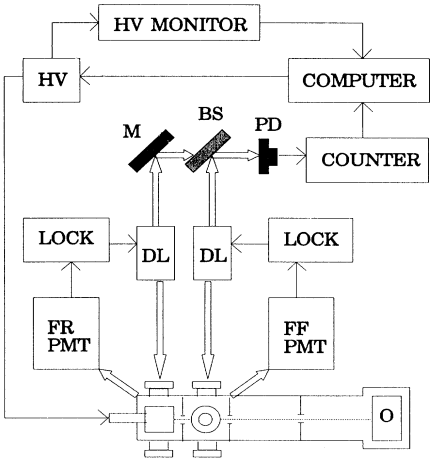

FIG. 1. Schematic of the apparatus; DL, diode laser; LOCK, laser locking circuitry; FR (FF) PMT, photomultiplier tubes viewing the field region (field-free region); COUNTER, high-frequency counter; HV, high-voltage supplies and controllers; O, lithium oven; open arrows, 671-nm light paths, solid arrows, electrical connections; dashed line, lithium beam; M, mirror; BS, beam splitter; PD, photodiode.

in the "field-free" region.

The electrodes are made of a transparent indium-tin-oxide coated glass that is 85% transmitting at 671 nm. The 1.5-in. square electrodes are spaced 3.8665(8) mm apart and will support a voltage difference of about 14 kV before allowing any significant discharge. Four 80% reflecting aluminum pads (diameter of about 2 mm, thickness of about 15 nm) have been deposited symmetrically upon a ring 7 mm from each electrode center. The optical cavities formed by these pads are used to measure the electrode spacing interferometrically using a ring dye laser and a wavemeter. Initially, an additional pair of somewhat larger aluminum pads were deposited at the electrode center where the field must be well known. After measuring the spacings of all five pads the central pad was removed with KOH to allow unobscured light collection. Over the course of data acquisition the outer pads were measured five additional times in order to assess the spacing stability. Numerical solutions to Laplace's equation for our geometry indicate that over the central interaction volume the field is reduced from that of infinite parallel plates by less than 1 ppb.

Two single-mode external cavity diode lasers generates the required 671-nm light [6]. Two NEC model NDL3200 laser diodes have been antireflection coated and placed in a cavity defined by a Littrow-mounted grating. A 1-mm-thick 85% reflecting étalon is required within each cavity to ensure single-mode operation. The beams from these lasers are brought in through the horizontal ports on the atomic beam apparatus to intersect the lithium beam at 90°. Photomultipliers placed below the interaction regions view the atomic fluorescence. A typical scan of the ^{6}Li D1 resonance lines is shown in Fig. 2. The observed 15-MHz linewidth (FWHM) is consistent with the residual Doppler broadening anticipated for our beam geometry. Each laser is frequency modulated by applying a sinusoidal high voltage to a piezoelectric transducer (PZT) that dithers the cavity length. Typically the modulation frequencies are a few kilohertz and the amplitude of the frequency modulations are a few megahertz. Lock-in detection at the modulation frequencies results in error signals that are fed back to the PZT in order to lock the lasers on to the centers of the atomic resonance lines.

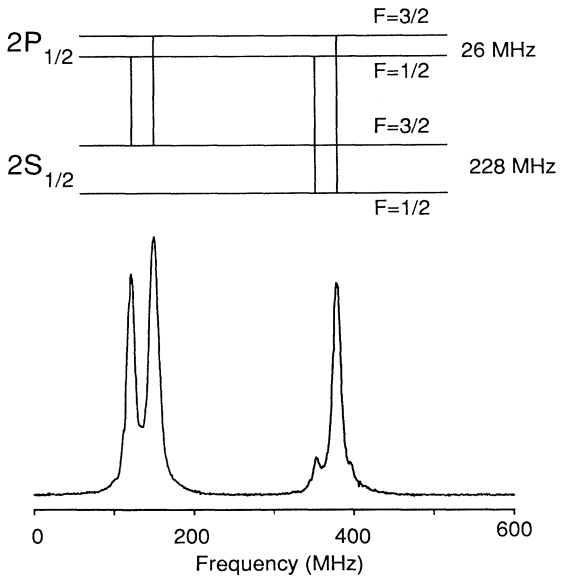

FIG. 2. Fluorescence spectrum of the ^{6}Li $2S_{1/2}$-$2P_{1/2}$ transition.

The light from the two lasers is also mixed on an avalanche photodiode (Mitsubishi PD 1002) with a 2-GHz bandwidth. The difference frequency is amplified and measured by a HP 5350B frequency counter. The results are integrated and then transmitted to our PC/AT data acquisition system.

Separate positive and negative high-voltage sources supply up to +7.5 and −7.5 kV to the two electrodes in the field region. Computer controlled relays turn the field on and off and reverse the polarity of the applied field. The voltages are monitored by two 30-MΩ high-voltage divider chains constructed from precision wire wound resistors. The voltage dividers have been calibrated to a precision of 60 ppm using a HP 3457A multimeter and a Fluke 8842A multimeter. The stability of the bridges is better than 20 ppm. The divided bridge voltages are read by the Fluke multimeter to a precision of 30 ppm and stored during data acquisition.

The data collection sequence is as follows. The lasers are locked on to a particular pair of hyperfine components and the voltage to be applied to the field plate is selected. The laser beat frequency is read ten times (about 6 sec) while the field is off. The field is turned on and there is a 6-s wait while the charging transients die down and the frequency servo resettles on the shifted atomic resonance frequency. Following another ten frequency measurements the field is again turned off and the process is repeated. A data point is constructed as the difference between the average of ten field-on beat frequency measurements and the average of the five field-off beat frequency measurements immediately preceding and following the field-on measurement. This procedure effectively removes the noise associated with any uniform drifts in the measured difference frequency. Typically, 50 to 100 such data points will be stored as a file before changing the experimental configuration (i.e., voltage, transition, intensity, modulation amplitude, or modulation frequency) and beginning again. Our total data sample consists of 133 files (about 70 hours of integration) taken in six separate data runs spread over about five months.

In the data analysis each point is weighted in inverse proportion to the square of the deviation of the constituent measurements. A weighted mean and standard error are then calculated for the ensemble of data points that make up the file. From the resulting average and the simultaneous voltage measurements, the Stark shifts [in kHz/(kV/cm)^2] is deduced. The measurements obtained in different files are averaged using a weight proportional to the inverse square of their standard error. The beat frequencies observed on different pairs of atomic transi-

tion vary from 20 MHz to nearly 900 MHz. The Stark induced shifts (typically about 5 MHz) are found to be independent of this initial frequency difference, as well as the polarization of the incident radiation (Table I). The observed shift is also found to be independent of the applied voltage, the laser intensity and the electric field polarity (see Table II). Also in Table II is a comparison of the data for which the beat frequency was either increasing or decreasing in magnitude when the field was applied. The good agreement between the different data acquisition configurations suggests that systematic effects associated with these variables are unimportant.

We conclude from our results that the scalar Stark shift for the $D1$ line is 4.6220(5)(20) kHz/(kV/cm)^2 for ^{6}Li and 4.6212(5)(20) kHz/(kV/cm)^2 for ^{7}Li, where the first uncertainty is statistical and the second reflects the systematic uncertainty associated with the field calibration which is dominated by our measurement of the electrode spacing. Averaging these results and combining the uncertainties in quadrature we obtain the value 4.6216(21) kHz/(kV/cm)^2 for the scalar Stark shift for this transition. With the usual definition of the scalar polarizability α_0 such that $\Delta E = -\frac{1}{2}\alpha_0 F^2$, where F is the applied field, our result constitutes a measurement of the difference between the scalar polarizabilities of the $2P_{1/2}$ and $2S_{1/2}$ states: $\alpha_0(2P_{1/2})-\alpha_0(2S_{1/2})=-5.512(2)\times10^{-24}$ cm^3. With the measured value [7] of $\alpha_0(2S_{1/2})=24.3(5)\times10^{-24}$ cm^3, the scalar polarizability of the $6P_{1/2}$ state can be inferred to be $\alpha_0(2P_{1/2})=18.8(5)\times10^{-24}$ cm^3.

We have calculated the Stark shift using second-order perturbation theory, deducing the relevant radial matrix elements from published oscillator strengths and energies [8] (Table III). Hydrogenic interpolations have been used for the high principal quantum numbers for which the energies and oscillator strengths are not available. Core contributions have been estimated to modify the shift by less than 0.1%. Continuum contribution have been neglected. Our measured shift is significantly smaller than that which is derived using the results of older theoretical models [9,10]. The agreement improves when experimental oscillator strengths are used where available [11–13]. If the two radial matrix elements presently available from the new MBPT calculation are substituted (Ref. [3]), the agreement improves still further. A fair comparison of our result with the new MCHF and MBPT theories will have to await a complete calculation of the effect. It is interesting to note that a 1% change in the $2S$-$2P$ oscillator strength results in a 5% change in the Stark shift. Our new result thus provides a formidable challenge to any calculation.

We have estimated the isotopic dependence of the Stark shift using experimental isotope shifts [14] and assuming that the radial matrix elements scale as the inverse of the reduced mass. The prediction that the Stark shift should be 0.6 Hz/(kV/cm)^2 larger in ^{6}Li than in ^{7}Li is consistent with the observed difference of 0.8(7) Hz/(kV/cm)^2.

TABLE I. $2S_{1/2}$-$2P_{1/2}$ scalar Stark shift [kHz/(kV/cm)^2] of the various hyperfine structure transitions. Vertical (V) polarization is parallel to E while horizontal (H) is perpendicular. Only statistical uncertainties have been included.

Isotope	Transition ($F\rightarrow F'$)	Shift V	Shift H
^{6}Li	1/2→3/2	4.6182(19)	4.6220(13)
	3/3→3/2	4.6213(14)	4.6221(7)
	3/2→1/2	4.6233(16)	4.6232(12)
^{7}Li	1→2	4.6226(16)	4.6222(46)
	1→1	4.6200(25)	4.6260(33)
	2→2	4.6212(9)	4.6170(48)
	2→1	4.6208(15)	4.6201(13)

TABLE II. Dependence of the observed Stark shift on the voltage difference between the electrodes, the intensity of the illuminating light (attenuation is by a factor of 15), the direction of the applied field, and whether the application of the field increased or decreased the magnitude of the frequency difference between the two lasers. Quoted uncertainties are purely statistical.

Selection criteria	Value	Stark shift [kHz/(kV/cm)^2]
Voltage	6 kV	4.6233(34)
Voltage	8 kV	4.6220(20)
Voltage	10 kV	4.6217(17)
Voltage	12 kV	4.6217(4)
Voltage	13.2 kV	4.6213(12)
Intensity	Attenuated	4.6209(7)
Intensity	Unattenuated	4.6219(5)
Field	Up	4.6218(5)
Field	Down	4.6215(5)
Frequency	Increasing	4.6216(7)
Frequency	Decreasing	4.6217(4)
None		4.6216(4)

TABLE III. The Stark shift deduced using various theoretical and experimental values of the lithium oscillator strengths. Energies of the various levels are taken from Ref. [8].

Model	Shift [kHz/(kV/cm)^2]
Theor.[a]	−5.42
Theor.[b]	−5.37
Expt.[c]	−4.73
Expt.[d]	−4.32
Expt. and theor.[e]	−4.69
Present result	−4.6216(21)

[a]Reference [9] with hydrogenic extrapolations for the Rydberg levels.
[b]As in footnote a with the f values of Ref. [10] supplemented where available.
[c]As in footnote a with Expt. values from Refs. [11] and [12] used where available.
[d]As in footnote c with with $2s$-$2p$ taken from Ref. [13].
[e]As in footnote c with ($2s$-$2p$) and ($2p$-$3s$) taken from Ref. [3].

The differential method used here has allowed the measurement a 4.5-MHz shift with a statistical uncertainty of 400 Hz. Most of the systematic difficulties encountered in locking a laser to an atomic line center (e.g., ac Stark effect, optical pumping, etc.) are unimportant when the signal is required to change with the application of an electric field. To the best of our knowledge the present result represents the most precise experimental determination of any Stark shift. The previous best measurement, done on the Cs $D2$ line achieved a precision of 0.2% [15]. It is interesting to note that the Cs Stark shift is about 25 times larger than that which we have now measured to 0.05% in Li.

We intend to apply this technique next to the Cesium $D1$ line. With a flatter and more stable electrode structure we hope to achieve a precision in this measurement of about 0.01%. This measurement may provide a resolution of the present discrepancy between theory [16] and experiment [17] as well as challenge the MCHF and MBPT calculations where they must be accurate in order to interpret the atomic parity violation experiments.

This method should also yield order-of-magnitude improvements in our knowledge of the scalar Stark shifts of rubidium and potassium. In atoms where the tensor shift is large enough to produce resolved pairs of magnetic substates, the tensor as well as scalar polarizabilities should be measurable. The method could also be easily inverted and used to precisely map electric fields.

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