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Precision measurement of Stark shifts for $6P_{3/2} \rightarrow nS_{1/2}$ $n = 10-13$ transitions in cesium

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The Stark shifts of four transitions in cesium were measured using two atomic beams. One traverses through a uniform electric field and the other a field-free region. The atoms were excited using a ring dye laser, with the laser beam interacting with the atoms in the electric-field region frequency shifted by an acousto-optic modulator. The Stark shift was determined by measuring the electric field for which the atoms in both beams are simultaneously in resonance. The results for the $6P_{3/2} \rightarrow (10-13)S_{1/2}$ transitions are $-59.36(14)$, $-154.68(13)$, $-356.57(29)$, and $-745.43(61)$ MHz $(\mathrm{kV/cm})^2$, respectively.

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I. INTRODUCTION

Recent advances in many-body theory have yielded improved atomic wave functions of alkali-metal atoms such as cesium [1,2]. These are essential for a number of experiments, including tests of parity violation [3]. Atomic wave functions are needed to compute radiative lifetimes and transition probabilities that can be compared to experimental results. Unfortunately, the aforementioned quantities are difficult to measure with accuracies of better than 1%. Recently, Stark shifts have been measured for transitions in lithium and cesium with uncertainties less than 0.1% [4,5]. The work to date has studied transitions from the ground to first excited states. This work reports measurements of the Stark shifts of the cesium $6P_{3/2} \rightarrow (10-13)S_{1/3}$ transitions. The results are over 50 times more accurate than those found previously and test our understanding of these excited states.

The experimental method is as follows. Two atomic beams are used. One passes through a field-free region while the other passes through a uniform electric field E which shifts the transition frequency an amount KE^2 where K is the Stark shift rate for the transition. The atoms are excited by a ring dye laser having a frequency ν. The laser beam exciting the atoms in the field is shifted in frequency using an accousto-optic modulator by an amount ν_{AO}. The laser is then scanned across the resonance, which is monitored by detecting fluorescence produced by the radiative decay of excited atoms. The Stark shift is found by measuring the electric field for which both beams are simultaneously in resonance. At this field, the acousto-optic frequency shift cancels the Stark shift:

$$h\nu_{\mathrm{AO}} = KE^2 , \tag{1}$$

and K is found by measuring the acousto-optic frequency and the electric field.

The simplest method for measuring Stark shifts is to use a single atomic beam or a cell. The advantage of simultaneously detecting two signals is that any fluctuations in either the laser frequency or intensity will affect the fluorescence from both beams and hence not affect the Stark shift measurement. Furthermore, fluorescence signals obtained using atomic beams are much narrower than those obtained using cells where atoms collide with each other and the cell walls, causing Doppler broadening and/or shifts in the resonance frequency. Another major improvement is the use of an acousto-optic modulator to shift the laser frequency, as was done by Watts *et al.* [6]. This permits more accurate measurement of frequency shifts than is possible using Fabry-Pérot interferometers.

II. EXPERIMENTAL PROCEDURE

The experimental setup is shown in Fig. 1. Two cesium atomic atomic beams are produced by an oven heated to a temperature of about 200 °C. The oven is enclosed by a liquid-nitrogen-cooled baffle to reduce scatter of the atomic beams. The atoms emerge from two opposite directions and are collimated by slits such that each beam has a divergence angle of 2.5 mrad. The entire system is housed in a vacuum chamber that is pumped by a diffusion pump to a pressure of 1×10^{-6} torr.

The $F=4$ hyperfine level of the cesium $6S_{1/2}$ ground state is excited to the $F=5$ hyperfine level of the $6P_{3/2}$

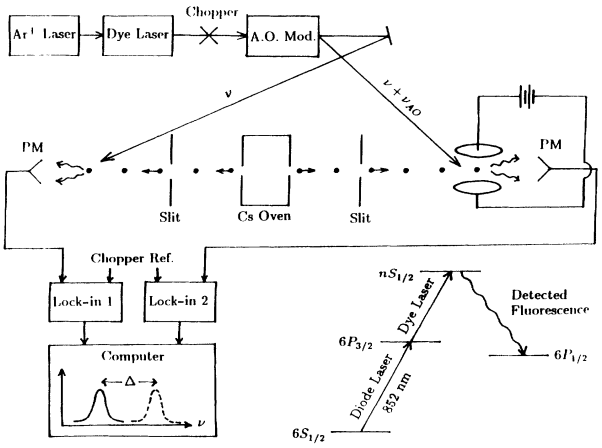

FIG. 1. Schematic of experimental setup. Details are described in the text.

state by a diode laser. The diode (Northern Telecom LT50A-03a) generates a maximum of 50 mW of power at 852 nm. The laser is tuned by adjusting its current and temperature. Fluorescence produced by the radiative decay of the $6P_{3/2}$ state back to the ground state is detected using an infrared camera. The laser frequency is tuned to optimize the fluorescence resulting from one of the atomic beams by adjusting the current.

A ring dye laser (Coherent 699) excites the $F=5$ level of the $6P_{3/2}$ state to the $F=4$ level of the $(10-13)S_{1/2}$ state. The laser frequency is electronically stabilized giving a manufacturer quoted linewidth of 0.5 MHz. Part of the dye laser is split off from the laser beam and is shifted in frequency by an acousto-optic modulator (Brimrose TEF 27-10). The modulator consists of a tellurium oxide crystal that has a diffraction efficiency of over 50% for shifting light at 590 nm by 220–320 MHz. The modulation frequency is supplied by a frequency synthesizer (Marconi 2022D), which is then amplified. The shifted and unshifted dye laser beams pass through 1-mm slits located on either side of the vacuum chamber. This permits alignment of the laser beams with an accuracy of 0.2 mrad.

The electric field was generated by applying a voltage across two stainless-steel plates which were ground smooth to better than 1×10^{-4} in. The plates are 0.5 in. thick and 5 in. in diameter. The plate spacing was determined using precision blocks whose thickness had an uncertainty of less than 1×10^{-5} in. To test the accuracy of our measurements, the plates were separated by 1.0000-in. spacers. It was then found that blocks having a maximum thickness of 0.9998 in. could slide between the plates. This test was repeated with different sets of blocks and spacers. The measured plate spacing always agreed with the spacer size to within 0.0002 in., which was therefore taken to be the measurement accuracy. For the final plate configuration (with the 1.0000-in. spacers removed), the plate spacing was measured to be 0.9998 in. at the position where the atomic beam passed and 0.9995 in. near the plate edge. The total uncertainty was conservatively estimated by adding the measurement uncertainty and half of the measured bending in quadrature, yielding a plate spacing of 0.9998 ± 0.00025 in. The electric field between the plates has been numerically modeled and was found to be uniform to better than one part in 10^5 at the intersection of the laser and atomic beams.

The voltage was supplied by a Fluke model 408A power supply that has a root mean square ripple measured to be less than 0.03%. The voltages were measured using a voltage divider (Julie Labs model KV-VB-10) that reduced the voltages by a factor of 1000, with a manufacturer quoted accuracy of better than 0.015% and an HP34401A voltmeter whose accuracy exceeds 0.005%. In our experiments, the voltage was measured before and after each data run. The two readings were found to agree to better than one part in 10^5.

Fluorescence resulting from the decay of the $nS_{1/2}\rightarrow6P_{1/2}$ state was detected using photomultipliers (Hamamatsu R928) labeled PM in Fig. 1. Two interference filters were used to eliminate scattered light at the wavelengths of the dye and diode lasers. The photomultiplier signals were each sent to a lock-in amplifier (Stanford Research Systems 850) whose reference was supplied by a chopper that modulated the dye laser beam at a frequency of 645 Hz. Both lock-in amplifiers recorded data when simultaneously triggered externally by a signal generator at a rate of 100 Hz while the laser scanned across the resonance. These data were then transferred to a computer for analysis.

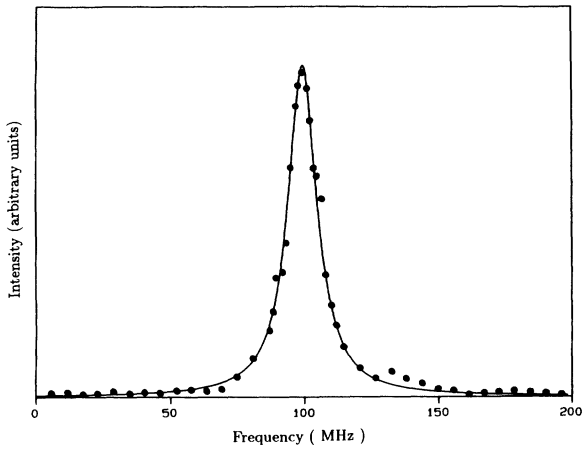

FIG. 2. Example of Lorentzian fit to data. The observed FWHM linewidth of 13 MHz is power broadened from the natural linewidth of 5 MHz. This broadening did not affect the determination of the Stark shift as is discussed in the text.

Each lock-in signal was fit to a Lorentzian function using a nonlinear least-squares fit, as shown in Fig. 2. The residual noise was caused by the frequency instability of the diode laser and randomly distorted the line shape from one data run to the next. However, this did not affect the determination of the difference between the line centers, Δ, which was found to be reproducible to better than one-tenth of the observed 13-MHz linewidth [full width at half maximum (FWHM)]. Figure 3 shows a plot of Δ versus the square of the electric field. These data were fit to a line $\Delta=A+BE^2$ to determine the electric field for which $\Delta=0$. Data collected in a few hours were

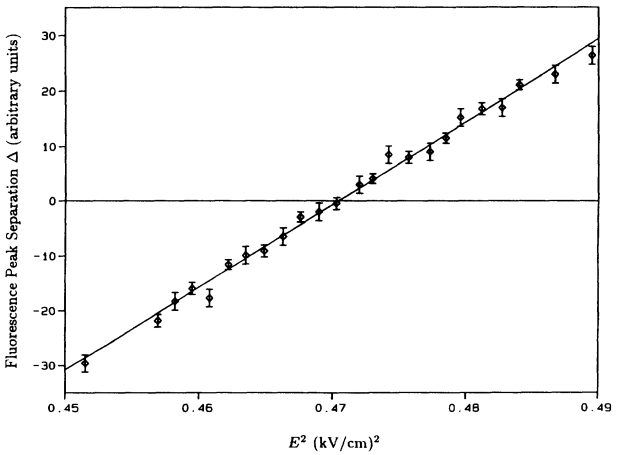

FIG. 3. Fluorescence peak separation Δ versus electric field squared. These data were taken for the $13S_{1/2}$ state using an acousto-optic frequency shift of 350 MHz, as described in the text.

TABLE I. Stark shift rate for the $6P_{3/2} \rightarrow 11S_{1/2}$ transition. The uncertainty due to statistics and alignment of laser and atomic beams is given in parentheses.

Data	Shift rate [MHz/(kV/cm)^2]
$\nu_{AO}=150$ *MHz*	−154.85(23)
$\nu_{AO}=250$ *MHz*	−154.60(12)
$\nu_{AO}=350$ *MHz*	−154.71(9)
Positive polarity	−154.68(11)
Negative polarity	−154.69(11)
Day 1	−154.64(11)
Day 2	−154.77(18)
Average of all data	−154.68(9)

sufficient to determine this field with a statistical uncertainty of about 0.05%. Finally, the Stark shift was found using Eq. (1).

Data were taken for several different acousto-optic frequency shifts and at both positive and negative voltages. The measurements were also repeated on different days. No dependence of the Stark shift rate on the acousto-optic frequency or voltage polarity was found, as is illustrated in Table I. The independence of the result on polarity indicates that possible stray electric fields due to charging of surfaces or due to contact potentials are not present. The lack of dependence on acousto-optic frequency indicates that the shift is indeed quadratic. The results were also checked to be independent of dye-laser power which was varied by a factor of 100 using neutral density filters. The line shape was broadened at the higher powers but not frequency shifted. Each dye-laser beam was attenuated to a power of about 400 μW, to reduce power broadening while data were collected.

III. RESULTS

The uncertainty of the final results arises due to a number of effects listed in Table II. One uncertainty results from a possible first-order Doppler shift that can arise if the laser and atomic beams are not orthogonal to each other. These shifts would affect our experiment only if the two fluorescent signals experienced different Doppler shifts. This was tested by measuring the signals when both atomic beams experienced no electric field. The two Lorentzian signals should exactly overlap in the case of identical alignment. A consistently small difference of 0.5 MHz was found, indicating that the intersection angles of the two laser beams with the two atomic beams differ by about 1 mrad. For a Stark shift that is determined using an acousto-optic shift of 350 MHz, this affects the final result by one part in 700. The offset between the two signals obtained at zero electric field was measured to an accuracy of about 30%. The so-called beam alignment uncertainty listed in Table II takes into account this offset as well as the accuracy with which the laser beams could be positioned through the 1-mm alignment slits.

Table II also lists the contributions of uncertainties in plate spacing and voltage measurement to the shift rate. The total uncertainty is taken as the square root of the sum of squares of the various uncertainties. The uncertainty of the $10S_{1/2}$ shift rate is larger than the others since data were only taken with an acousto-optical shift of 150 MHz. Larger shifts could not be used since the required voltages exceeded the capability of the power supply.

The polarizability of the $nS_{1/2}$ state can be found from the shift rate, using

$$K=-\tfrac{1}{2}[\alpha_0(nS_{1/2})-\alpha_0(6P_{3/2}) -\alpha_2(6P_{3/2})(m_J^2-5/4)] , \quad (2)$$

where α_0 and α_2 are the scalar and tensor polarizabilities, and m_J is the azimuthal quantum number of the $6P_{3/2}$ state. The shift is dominated by the large $nS_{1/2}$ polarizability; and the small (approximately 0.1%) $6P_{3/2}$ contributions can be subtracted out using the values $\alpha_0(6P_{3/2})=0.407$ and $\alpha_2(6P_{3/2})=-0.065$ MHz/kV/cm^2 computed by Zhou and Norcross [7]. The latter value is in excellent agreement with the experimental result of $-0.0652(4)$ MHz/kV/cm^2 [5]. It was assumed that the circularly polarized diode laser predominantly excites $m_J=3/2$; however, the effect of $\alpha_2(6P_{3/2})$ is entirely negligible compared to the uncertainty of the measured shift rate. The results for $\alpha_0(nS_{1/2})$ are listed in Table III and are consistent with but substantially more precise than those obtained by Fredriksson and Svanberg [8]. The improved accuracy results from the use of a diode laser instead of a lamp to excite the $6P_{3/2}$ state, as well as more accurate measurements of electric fields and frequency shifts.

Table III also compares the results to those computed in Ref. [8] using a Coulomb approximation. The agreement is very good, although the present data are consid-

TABLE II. Error budget of Stark shift rates for $6P_{3/2} \rightarrow nS_{1/2}$ transitions.

n	Statistics (%)	Beam alignment (%)	Plate spacing (%)	Voltage (%)	Total uncertainty (%)	Shift rate MHz/(kV/cm)^2
10	0.10	0.20	0.05	0.03	0.24	−59.36(14)
11	0.02	0.06	0.05	0.03	0.08	−154.68(13)
12	0.02	0.05	0.05	0.03	0.08	−356.57(29)
13	0.03	0.05	0.05	0.03	0.08	−745.43(61)

TABLE III. $\alpha_0(nS_{1/2})$ $[a_0^3]$. The uncertainty is given in parentheses.

n	Previous work [8]	Present work	Theoretical [8]
10	123(6)	119.06(28)	118
11	322(16)	309.70(26)	309
12	720(45)	713.48(58)	709
13	1650(170)	1491.20(122)	1490

erably more accurate, indicating more elaborate theoretical work is desirable. It is interesting to note that each $nS_{1/2}$ polarizability is predominantly due to the two closest P states. Hence, these measurements can be viewed as measurements of the r matrix element connecting the $S_{1/2}$ and its neighboring P states. As such, the measurements give precise information about the wave functions and matrix elements of these highly excited states.

In conclusion, we wish to emphasize that excited-state polarizabilities can be measured to accuracies of better than 0.1%. The method of monitoring two atomic beams eliminates a large number of potential systematic effects. We therefore believe that this is a useful method for studying excited states of any atom accessible to lasers.

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[1] V. A Dzuba, V. V. Flambaum, and O. P. Sushkov, Phys. Lett. A **140**, 493 (1989).

[2] S. A. Blundell, W. R. Johnson, and J. Sapirstein, Phys. Rev. A **43**, 3407 (1991).

[3] M. C. Noecker, B. P. Masterson, and C. E. Wieman, Phys. Rev. Lett. **61**, 310 (1988).

[4] L. R. Hunter, D. Krause, K. E. Miller, D. J. Berkeland, and M. G. Boshier, Opt. Commun. **94**, 210 (1992); L. R. Hunter, D. Krause, Jr., D. J. Berkeland, and M. G. Boshier, Phys. Rev. A **44**, 6140 (1991).

[5] C. E. Tanner and C. Wieman, Phys. Rev. A **38**, 162 (1988).

[6] R. N. Watts, S. L. Gilbert, and C. E. Wieman, Phys. Rev. A **27**, 2769 (1983).

[7] H. L. Zhou and D. W. Norcross, Phys. Rev. A **40**, 5048 (1989).

[8] K. Fredriksson and S. Svanberg, Z. Phys. A **281**, 189 (1977).