Stark-shift measurement of the $(6s)^2\,{}^1S_0 \rightarrow (6s6p)^1P_1$ barium transition

J. Li and W. A. van Wijngaarden
Department of Physics, York University, Toronto, Ontario, Canada M3J 1P3
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The Stark shift of the $(6s)^2\,{}^1S_0 \rightarrow (6s6p)^1P_1$ transition in barium was measured using a laser to excite an atomic beam as it traversed a uniform electric field and a field-free region. The laser frequency was scanned across the transition while fluorescence produced by the radiative decay of the excited state was detected. The frequency scan was calibrated using an acousto-optic modulator which frequency shifted the laser by a specified amount. The Stark-shift rate was found to be -28.532 ± 0.060 kHz/(kV/cm)^2. The hyperfine splitting of the ^{135,137}Ba $F=\frac{1}{2}$ level relative to ^{138}Ba of the $(6s6p)^1P_1$ state was determined to be -549.47 ± 0.12 MHz, where the minus sign indicates that the ^{138}Ba transition is at a lower frequency.

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I. INTRODUCTION

Precision measurements of Stark shifts provide information about polarizabilities of atomic states that are important for describing a number of properties, including charge-exchange cross sections, van der Waals constants, and dielectric constants [1]. Polarizabilities have been determined using a number of methods that are nicely reviewed by Miller and Bederson [2,3]. In recent years, laser spectroscopic techniques have measured Stark shifts for transitions in alkali-metal atoms with unprecedented accuracy [4–11]. In addition, some theoretical calculations have been made [12–16]. This paper reports the measurement of the Stark shift of the $(6s)^2\,{}^1S_0 \rightarrow (6s6p)^1P_1$ transition in barium. The result is significantly more precise than any we have found in the literature for this transition.

Barium has naturally occurring isotopes having atomic mass units of 138 (71.7%), 137 (11.3%), 136 (7.8%), 135 (6.6%), 134 (2.4%), 132 (0.1%), and 130 (0.1%). The nuclear spin for the even isotopes is zero and for the odd isotopes is $\frac{3}{2}$. This experiment measured the Stark shift experienced by ^{138}Ba, since this isotope produced the largest fluorescent signal and did not have any hyperfine structure. The response of an atom to an electric field E is given by the Hamiltonian

$$H=-\left\{\alpha_0+\alpha_2\frac{3m^2-J(J+1)}{J(2J-1)}\right\}\frac{E^2}{2}\ . \tag{1}$$

Here α_0 and α_2 are the scalar and tensor polarizabilities, respectively. J is the electronic angular momentum and m is its azimuthal component. The electric-field shifts the frequency for the transition between the $(6s)^2\,{}^1S_0$ and the $(6s6p)^1P_1$ states by the amount

$$\Delta\nu=KE^2\ , \tag{2}$$

where the Stark-shift rate K is

$$K=-\tfrac{1}{2}\{\alpha_0(^1P_1)-2\alpha_2(^1P_1)-\alpha_0(^1S_0)\}\ . \tag{3}$$

Here, m has been set to zero since the laser is linearly polarized along the quantization axis that is specified by the electric field and therefore only populates the $m=0$ sublevel of the $^1P_1$ state.

The Stark shift of the $(6s)^2\,{}^1S_0 \rightarrow (6s6p)^1P_1$ transition was first measured by Kopfermann and co-workers [17]. The absorption spectrum of barium was studied by passing light from a lamp through an atomic beam that traversed an electric field. The transmitted light was filtered by a Fabry-Pérot interferometer and detected using a photographic plate. Spectra were taken at various field strengths, using both linear and circular polarized light. Values of $\alpha_0(^1P_1)-\alpha_0(^1S_0)=36.8\pm1.0$ kHz/(kV/cm)^2 and $\alpha_2(^1P_1)=-9.9\pm0.7$ kHz/(kV/cm)^2 were measured, giving a Stark-shift rate $K=-28.3\pm1.2$ kHz/(kV/cm)^2. The scalar polarizability of the ground state has been determined by Schwartz *et al.*, who measured the deflection of an atomic beam by an inhomogeneous electric field. A result of $\alpha_0((6s)^2\,{}^1S_0)=66.6\pm5.4$ kHz/(kV/cm)^2 was obtained [18]. This agrees with the theoretical value of 67.8 kHz/(kV/cm)^2 computed using many-body theory [19]. The tensor polarizability of the $(6s6p)^1P_1$ state has been measured using level-crossing and quantum-beat spectroscopy to be -10.72 ± 0.10 and -10.79 ± 0.29 kHz/(kV/cm)^2, respectively [20,21].

Our experiment used a ring dye laser to excite an atomic beam when it passed through a uniform electric field and a field-free region. The laser frequency was tuned across the transition, while fluorescence produced by the radiative decay of the excited state was monitored by two detectors. The frequency scan was calibrated by using an acousto-optic modulator to frequency shift part of the laser beam. The frequency-shifted and -unshifted laser beams were superimposed and excited the atoms in the field-free region. Each atomic transition therefore generated two peaks in the fluorescence spectrum that were separated by the modulation frequency. This method of frequency calibration has been used to precisely measure hyperfine and isotope shifts [22]. It requires simpler and cheaper equipment and yields more accurate results than

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using an interferometer to monitor the change in laser frequency. In this paper, Sec. II describes the apparatus. The data analysis and conclusions are given in Sec. III.

II. APPARATUS

The apparatus is illustrated in Fig. 1. An atomic beam was generated by heating the barium metal close to its melting point of 725 °C. Atoms were collimated by a series of slits and baffles, producing a beam that has a divergence of about 2.5 mrad. The entire system was housed in a vacuum chamber that was pumped by a diffusion pump and a liquid-nitrogen trap to a pressure of 1×10^{-7} torr.

The laser light was supplied by a ring dye laser (Coherent 699). This was pumped by an Ar^+ laser that produced 6 W of power at 514 nm. The frequency of the dye laser was electronically stabilized, giving a manufacturer-quoted linewidth of 0.5 MHz. The dye laser generated several hundred milliwatts of light at 553.7 nm using Rhodamine 110 laser dye. Part of the dye laser beam was frequency shifted by an acousto-optic (AO) modulator (Brimrose TEF 27-10). The modulation signal ν_{AO} was supplied by a frequency synthesizer (Hewlett-Packard 8647A) with an accuracy of one part in 10^6, and then amplified. The amplifier bandwidth limited the modulation frequency to 300.000 MHz.

Atoms in the field-free region were excited by both the frequency-shifted and -unshifted laser beams. The two lasers were superimposed using two 0.1-cm wide slits located 100 cm apart on either side of the vacuum chamber. The beams could be collimated to within a quarter of the slit width, permitting an alignment accuracy of 0.25 mrad. Atoms in the field region were only excited by the laser that was not frequency shifted. The lasers intersected the atomic beam orthogonally to eliminate first-order Doppler shifts.

The electric field was generated by applying a voltage across two stainless-steel plates. The plates are 0.5 in. thick and have a diameter of 3 in. The surfaces were ground smooth to better than 1×10^{-4} in. The plate spacing was found using machinists blocks whose thickness had an uncertainty of less than 1×10^{-5} in. The measurement accuracy was tested by separating the plates using 0.4000-in. spacers. It was then found that blocks having a maximum thickness of 0.3999 in. could slide between the plates. The test was repeated using different sets of precision blocks and the measured plate spacing always agreed with the spacer size to within 0.0001 in. For the final plate configuration (with the 0.4000-in. spacers removed) the distance separating the plates was determined to be 0.4001 in., with an uncertainty conservatively estimated to be 0.0002 in. The electric field between the plates was numerically modeled, and its uniformity at the intersection of the laser and atomic beams was found to be better than one part in 10^5.

Plate voltages of 0–50 kV were provided by a power supply (Glassman EH50R02.0) that had a root-mean-square ripple of less than 0.03%. The voltage was continuously monitored using a voltage divider (Julie Labs KV-50/01), which reduced the voltage by a factor of 5000 with an accuracy of 0.01%. The reduced voltage was measured by a voltmeter (HP34401A) having an uncertainty of less than 0.002%. The voltage was observed to be stable to within two parts in 10^5 during the time for a scan of the laser frequency across the transition.

Fluorescence resulting from the radiative decay of the $(6s6p)^1P_1\rightarrow(6s)^2\,^1S_0$ state was detected using two photomultipliers (Hamamatsu R928) labeled PM in Fig. 1. PM1 detected fluorescence from the field-free region emitted along the vertical ($\hat{z}$) axis, while PM2 observed light emitted in the horizontal ($\hat{y}$) direction. The laser beams were linearly polarized along an axis perpendicular to the direction of detected fluorescence for both the field and field-free regions. This configuration has been shown to maximize the fluorescence signal generated by ^{138}Ba relative to ^{135,137}Ba [23,24]. Hence, the laser beams traversing the field-free region were horizontally polarized, while the laser beam passing through the electric field was vertically polarized.

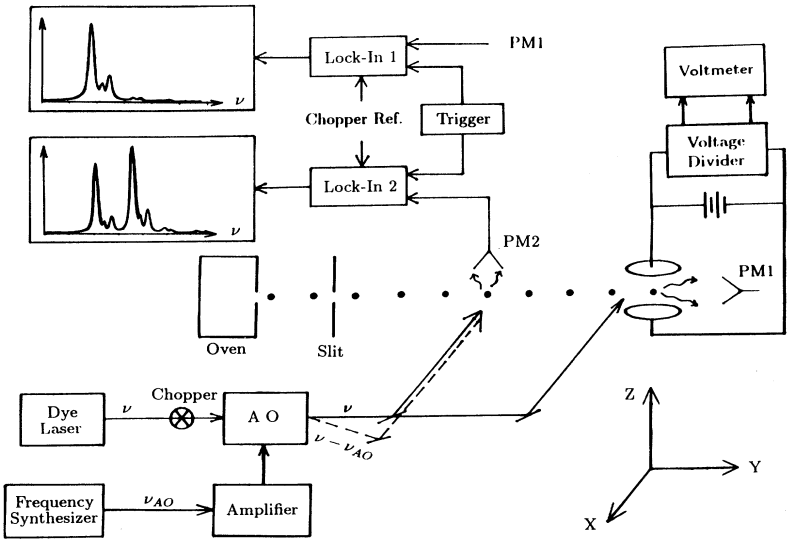

FIG. 1. Apparatus. Details are described in the text.

The photomultiplier signals were processed by separate lock-in amplifiers [Stanford Research (SR) 850]. The reference signal was supplied by a chopper (SR540) that modulated the laser beam with a maximum possible frequency of 2 kHz. The demodulated signals were recorded by both lock-in amplifiers when they were simultaneously triggered by a single generator at a rate of 256 Hz.

III. ANALYSIS AND CONCLUSIONS

A typical wavelength scan consisting of about 7500 data points is shown in Fig. 2. Figure 2(a) displays data produced by exciting atoms in the field-free region using the frequency-shifted and -unshifted laser beams. Figure 2(b) was obtained by exciting atoms passing between the electric-field plates by the dye laser beam that was not frequency shifted. The laser power was attenuated to a few milliwatts to reduce power broadening. The peak positions varied up to 0.1% from scan to scan due to variations in the laser scan speed. The frequency of each scan

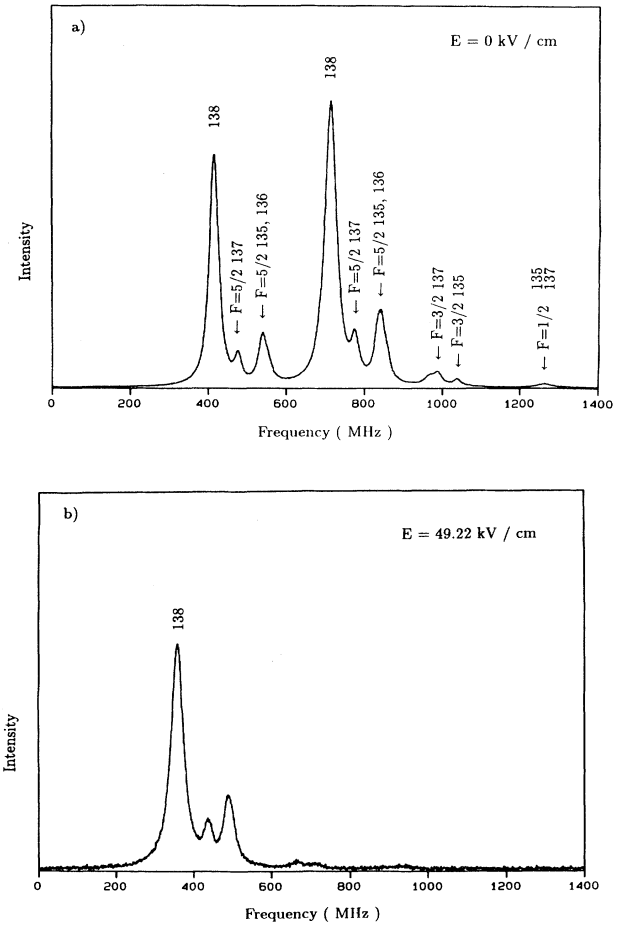

FIG. 2. Sample data. (a) shows the fluorescent signal observed in the field-free region. The first ^{138}Ba peak was produced using the dye laser that is not frequency shifted, while the second ^{138}Ba peak was excited using the part of the laser beam that is shifted 300.000 MHz. (b) shows the signal generated when the frequency-unshifted laser beam excited atoms as they passed through an electric field.

was therefore separately calibrated as follows. First, the lock-in amplifiers were used to fit a Gaussian function to the peaks to determine their center positions. Next, the frequency interval per data point was determined by dividing the acousto-optic frequency of $\nu_{AO}=300.000$ MHz by the number of points separating the two ^{138}Ba peaks in the signal observed in the field-free region. The average frequency interval represented by a single data point was 0.1845 MHz. Frequency intervals separating any two peaks could then be found.

The barium spectrum shown in Fig. 2(a) consists of a number of overlapping lines. Hence, the line center differs slightly from the frequency where the peak intensity is maximum. This so-called frequency pulling effect was studied by modeling the spectrum by the function

$$F(\nu)=\sum_i \left\{ \frac{A_i}{1+\left[\frac{\nu-\nu_i}{\Gamma/2}\right]^2} + \frac{B_i}{1+\left[\frac{\nu-\nu_{AO}-\nu_i}{\Gamma/2}\right]^2} \right\} . \tag{4}$$

Here, i is summed over the various hyperfine and isotope transitions. Values of the line centers ν_i relative to ^{138}Ba were taken from Ref. [24]. A_i and B_i are the observed peak amplitudes of the signals generated by the frequency-unshifted and -shifted laser beams, respectively. The linewidth (full width at half maximum intensity) Γ was set to the observed value of 35 MHz. This is larger than the natural linewidth of 19 MHz due to power broadening. The maximum intensity positions of the two ^{138}Ba peaks differed from the actual line centers by less than 0.1 MHz, while their separation frequency differed from the acousto-optic frequency of 300.000 MHz by less than 0.05 MHz. A similar analysis was used to study the Stark-shifted spectrum shown in Fig. 2(b). Isotopes such as ^{137}Ba that have a hyperfine structure were observed to have a smaller Stark shift than ^{138}Ba. The overlap of the $F=\frac{5}{2}$ ^{137}Ba and ^{138}Ba peaks decreased as the electric field increased, reducing the frequency pulling effect. This affected the largest observed Stark shift of 69.16 MHz by less than 0.1%. These conclusions were unchanged when the line shape was modeled using a Gaussian instead of a Lorentzian function.

The calibration procedure was checked by measuring the hyperfine splitting of the $F=\frac{1}{2}$ ^{135,137}Ba ^{138}Ba peak relative to ^{138}Ba of the $(6s6p)^1P_1$ state. Data from 590 separate wavelength scans were averaged, yielding a value of -549.47 MHz with a standard deviation of 0.12 MHz. The hyperfine splitting has been measured by Nowicki *et al.* to be 549.9 ± 0.6 MHz [24]. They locked two separate lasers to the transitions observed in different isotopes. The lasers beams were then superimposed on a detector and the beat frequency was measured.

The average frequency shifts are plotted versus the square of the electric field in Fig. 3 [25]. The error bars of the data points corresponding to zero and 50 kV have the smallest uncertainties, since 500 of the 590 scans were taken at these voltages. A least-squares fit of the function $y=KE^2+y_0$ to the data yielded

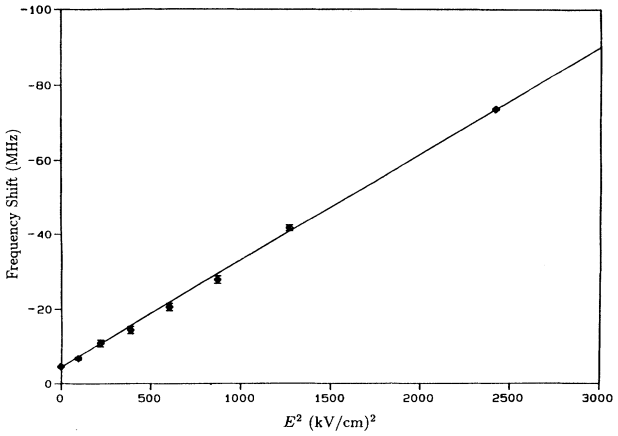

FIG. 3. Frequency shift versus electric field squared. The uncertainties of the Stark shifts measured at the highest and lowest electric fields are substantially smaller than the diameter of the data point.

$$K = -28.532 \pm 0.060 \text{ kHz}/(\text{kV/cm})^2 .$$

The uncertainty of this result is due to statistical variation of the data caused by jitter of the dye laser frequency. Other factors such as the frequency pulling effect and uncertainties in the determination of the electric field were negligible in comparison. The frequency shift at zero field y_0 was -5.09 MHz. This offset is believed to arise due to a small difference of the intersection angle of the laser and atomic beams in the field and field-free regions. For a barium beam generated at an oven temperature of 1000 K, the atoms have a speed of about 4×10^4 cm/sec, giving rise to a Doppler width of 750 MHz. Hence, a 7-mrad alignment difference accounts for the observed offset.

The result for K is consistent with the earlier value obtained by Kopfermann, although it is much more accurate. Substituting it along with measurements of $\alpha_0(^1S_0)$ and $\alpha_2(^1P_1)$ into Eq. (3) yields the scalar polarizability of the excited state $\alpha_0((6s6p)^1P_1) = 102.2 \pm 5.8$ kHz/(kV/cm)^2. The accuracy of this result is limited by the uncertainty of the ground-state polarizability.

The experiment can be improved by using a higher electric field to produce a larger Stark shift. The uncertainty in the determination of the line centers can also be reduced by using an atomic beam consisting of only the ^{138}Ba isotope. It should then be possible to determine the Stark-shift rate with an uncertainty of less than 0.1%.

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