Stark shift measurement of the $(4s)^2\ ^1S_0 \rightarrow (4s4p)\ ^3P_1$ calcium transition

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The Stark shift of the $(4s)^2\ ^1S_0 \rightarrow (4s4p)\ ^3P_1$ transition in calcium was measured using a laser to excite an atomic beam as it traversed through 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 to frequency shift part of the laser beam. The Stark shift rate was found to be -12.314 ± 0.041 kHz/(kV/cm)^2, where the minus sign indicates the transition frequency decreases when the electric field is applied.

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A number of experimental groups have determined values of Stark shifts and/or polarizabilities for various calcium states with an accuracy of a few percent [1–4]. Polarizabilities of atomic states are important for describing charge-exchange cross sections, van der Waals constants, and dielectric constants [5–7]. Accurate experimental data have also spurred on the development of atomic theory. In calcium, much work has been done computing the ground-state polarizability $\alpha_0(^1S_0)$. The various methods and their results are summarized by the papers by Glass [8] and Sadlej and Urban [9]. This paper reports a measurement of the Stark shift rate of the $(4s)^2\ ^1S_0 \rightarrow (4s4p)\ ^3P_1$ transition. The result has an uncertainty 3 parts in 10^3, which is significantly more accurate than any Stark shift found in the literature for a calcium transition. This particular transition has also been used to study hyperfine and isotope shifts of calcium isotopes [10] and recently has been of interest in atom interferometry [11].

In this experiment, only ^{40}Ca was excited, which has an abundance of 96.9% and a nuclear spin of zero. The response of this isotope to an electric field E is given by the Hamiltonian

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

where 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 Stark shift for the transition between the $(4s)^2\ ^1S_0$ and the $(4s4p)\ ^3P_1$ states is given by

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

where the Stark shift rate K is

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

Here, m was set to zero since the laser is linearly polarized parallel to the electric field E and therefore only populates the $m=0$ sublevel.

The apparatus is illustrated in Fig. 1. It has been described in detail elsewhere and is therefore only briefly discussed [12,13]. An atomic beam of calcium was generated using an oven and a series of slits producing a beam having a diver-

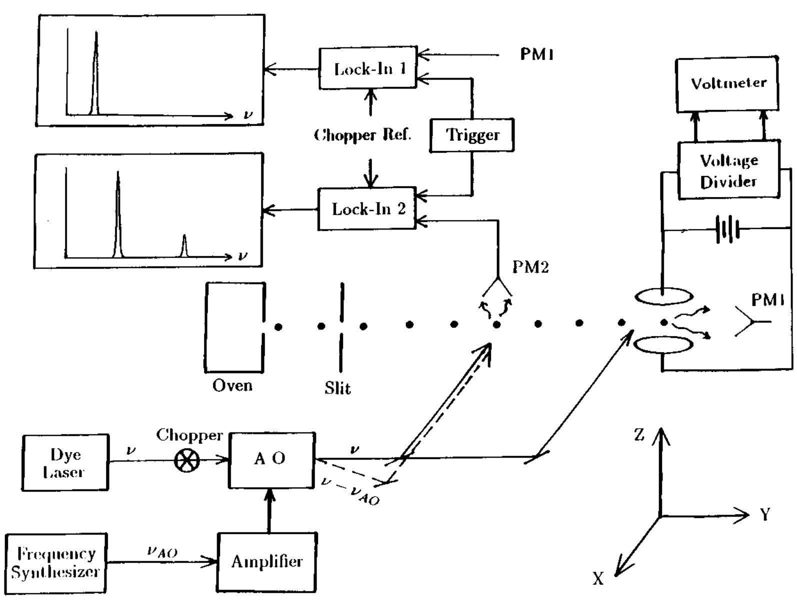

FIG. 1. Apparatus. Details are described in the text.

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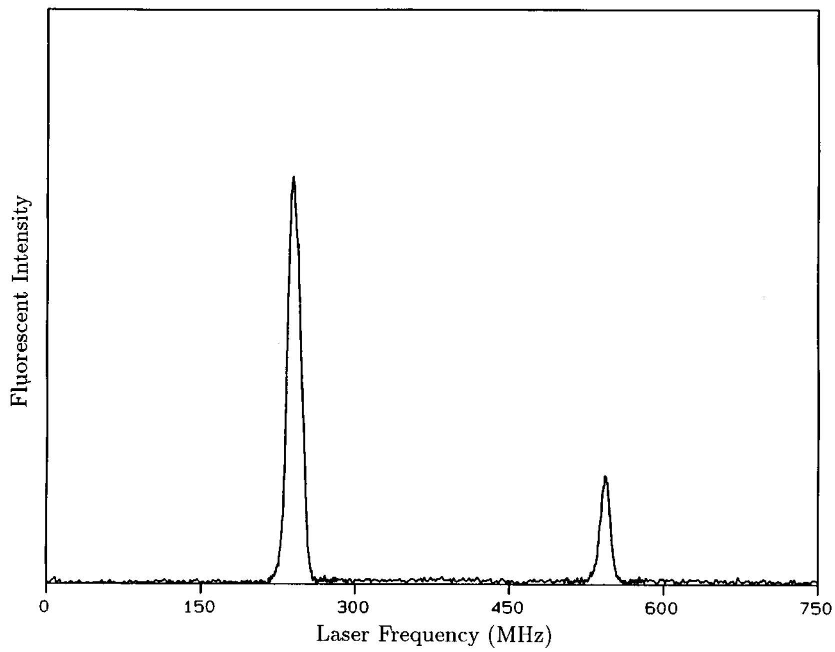

FIG. 2. Sample data. The fluorescent signal observed in the field-free region is shown. The first peak was produced when atoms were excited by the laser at frequency ν. The second peak was generated using the laser that was shifted 300 000 MHz by the acousto-optic modulator.

gence of about 2 mrad. The oven was contained in a vacuum chamber that was pumped to a pressure of 1×10^{-7} torr. Laser light at a wavelength of 657.46 nm was supplied by a ring dye laser (Coherent 699) that was pumped by an argon ion laser. Part of the dye laser beam was frequency shifted by an acousto-optic (AO) modulator. The modulation signal ν_{AO} was supplied by a frequency synthesizer with an accuracy of 1 part in 10^6, and then amplified. In our experiments, ν_{AO} was set to 300 000 MHz, which equals the amplifier bandwidth. The laser beams at frequencies ν and $\nu-\nu_{\mathrm{AO}}$ were superimposed and excited the atoms in a region free of electric fields. The laser beam at frequency ν also excited atoms as they passed through a uniform electric field. The lasers intersected the atomic beam orthogonally to eliminate first-order Doppler shifts. The electric field was generated by applying voltages of up to 50 kV across two stainless steel plates having a diameter of 3 in. A voltmeter measured the output of a voltage divider that reduced the high voltage by a factor of 5000 with an accuracy of 0.01%. The plate spacing was found to be 0.4001 ± 0.0002 in. using precision machinist blocks.

Fluorescence, produced by the radiative decay of the excited $(4s4p)\ ^3P_1$ state back to the ground state, was detected by two photomultipliers (PM1 and PM2) as the laser frequency was tuned across the resonance transition. The photomultiplier signals were processed by separate lock-in

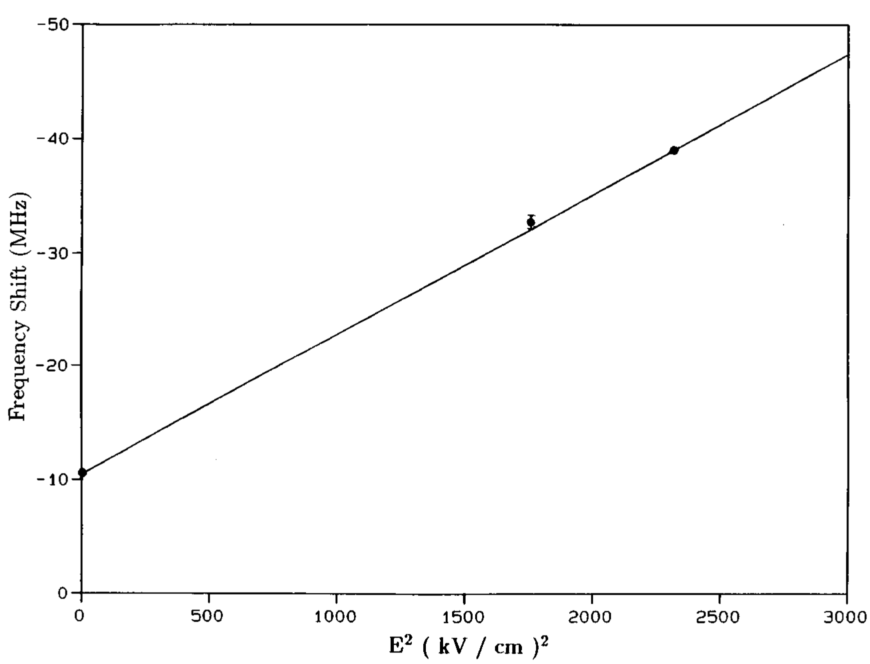

FIG. 3. Frequency shift vs electric field squared. The data point at 0 kV/cm represents the average shift found in 215 separate frequency scans while the points at 41.9 and 48.1 kV/cm were obtained from 18 and 209 separate scans, respectively. For the data taken at 0 and 48.1 kV/cm, the standard deviation of the frequency shifts about their average value is less than that indicated by the size of the data points.

amplifiers. The reference signal was supplied by a chopper that modulated the laser beam with a 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.

A typical wavelength scan consisting of about 5000 data points is shown in Fig. 2. It displays data produced in the field-free region by the laser beams at frequencies ν and $\nu-\nu_{AO}$. The frequency of each scan was calibrated as follows. First, the lock-in amplifiers fit a Gaussian function to the peaks to determine the position of the line centers. The frequency interval per data point was then found by dividing the acousto-optic frequency of 300 000 MHz by the number of points separating the two peaks observed in the field-free region. The average frequency interval represented by a single data point was 0.1576 MHz. Finally, the frequency interval separating the peaks excited by the laser at frequency ν in the field and field-free regions was found.

The results of 442 separate wavelengths scans taken at three different electric fields are summarized in Fig. 3. The results were unaffected when the voltage polarity was reversed. A least-squares fit of the function $y=KE^2+y_0$ to the data yielded

$$K=-12.314\pm 0.041 \text{ kHz/(kV/cm)}^2.$$

The frequency shift at zero field y_0 was found to be -10.48 MHz. This offset has been found to arise from a small difference of the intersection angle of the laser and atomic beams in the field and field-free regions [12]. The result for K can be used along with the ground-state polarizability $\alpha_0(^1S_0)=41.9\pm4.2$ kHz/(kV/cm)^2 determined in a beam deflection experiment [1] to obtain

$$\alpha_0(^3P_1)-2\alpha_2(^3P_1)=66.5\pm 4.2 \text{ kHz/(kV/cm)}^2.$$

A value of $\alpha_0(^3P_1)$ can be found once the excited-state tensor polarizability is known. $\alpha_2(^3P_1)$ can be obtained using various methods including quantum beat or level crossing spectroscopy as has been done for other alkaline states [14,15]. In conclusion, this experiment has reported the most accurate Stark shift measured to date for a calcium transition. The result provides a test of future alkaline atomic models that may be used to make theoretical computations of excited-states polarizabilities.

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