Measurement of the ^{133}Cs $6p\,^2P_{1/2}$ state hyperfine structure

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(Received 25 November 1996; revised manuscript received 19 February 1997)

We employ crossed-beam laser spectroscopy to measure the hyperfine structure of the $6p\ ^2P_{1/2}$ state of atomic ^{133}Cs ($I=\frac{7}{2}$). A frequency-stabilized Ti:sapphire laser with 1-GHz sidebands excites a thermal atomic cesium beam through the $6s\ ^2S_{1/2}\ F=3,4\rightarrow 6p\ ^2P_{1/2}\ F'=3,4$ resonance transitions at 895 nm. The hyperfine splitting of the excited state is determined from the observed fluorescence spectra. Our result for the $6p\ ^2P_{1/2}\ F'=3$ to $F'=4$ hyperfine splitting is 1167.54(32) MHz, yielding a magnetic dipole coefficient of $A=291.89(8)$ MHz. In this Brief Report we also compare our results with previous measurements and theory. [S1050-2947(97)00907-4]

PACS number(s): 32.10.Fn, 32.80.Ys

The low-lying electronic states of Cs are being studied in our laboratory because of their importance to the interpretation of atomic parity nonconservation (PNC) measurements [1]. High accuracy calculations near the nucleus are of interest to the study of PNC in atoms because the weak force produces a short-range electron-nucleon interaction and is sensitive to the electronic wave function in this region. One of the most stringent tests of atomic structure calculations in the vicinity of the nucleus is the comparison between theoretical and experimental determinations of hyperfine structure [2]. In particular, hyperfine structure is sensitive to relativistic, core polarization, and core correlation effects which are sources of difficulty in accurate calculations for multielectron systems. Experimental measurements of the hyperfine structure in alkali-metal atoms are reviewed in Ref. [3].

In the approximation that J is a good quantum number, the hyperfine interaction shifts the energy of the atomic fine structure levels by an amount given by

$$W=hA(\mathbf{I}\cdot\mathbf{J})+hB\frac{6(\mathbf{I}\cdot\mathbf{J})^2+3(\mathbf{I}\cdot\mathbf{J})-2(\mathbf{I}\cdot\mathbf{I})(\mathbf{J}\cdot\mathbf{J})}{2I(2I-1)2J(2J-1)} \quad (1)$$

for states where $J=L\pm\frac{1}{2}$. A and B are, respectively, the coefficients of the magnetic dipole and electric quadrupole contributions to the hyperfine structure. Measurements of A and B are most conveniently compared to relativistic atomic structure calculations through the effective operator formalism developed by Sandars and Beck where A and B are related to various radial matrix elements [4,5]. We report in this paper a precision measurement of the hyperfine splitting in the ^{133}Cs $6p\ ^2P_{1/2}$ fine structure state ($I=\frac{7}{2}$). For cases where $J=\frac{1}{2}$, as for the measurement reported here, the coefficient B is zero [3] and our results yield a determination of the magnetic dipole contribution A.

The hyperfine splitting of the ^{133}Cs $6p\ ^2P_{1/2}$ state has been previously measured by a number of authors [6–12] using interferometric and optical double resonance techniques. Figure 1 shows the previous measurements along with the results we are reporting here. Our measurements were performed using a cesium thermal beam apparatus, similar to that of Ref. [13], which we constructed as an optical frequency diagnostic for another experiment [14–16]. Our beam apparatus was designed to maintain high precision for laser scans over large ($\approx$10 GHz) frequency intervals and remain essentially drift-free over long time periods.

In our measurement, single-frequency laser radiation from a Coherent 899-21 Ti:sapphire laser tuned near the $6s\ ^2S_{1/2}\rightarrow 6p\ ^2P_{1/2}$ resonance transition was phase modulated with a resonant-cavity-type $LiTaO_3$ electro-optic modulator (EOM) at a radio frequency (rf) near that of the $6p\ ^2P_{1/2}\ F'=3$ to $F'=4$ hyperfine interval. The apparatus is shown schematically in Fig. 2. The phase-modulated light was linearly polarized and directed to intersect a collimated beam of cesium atoms at right angles in an ultrahigh-vacuum vertical thermal beam tube. Fluorescence from the atomic beam was detected with two photodiodes mounted on opposite sides of the interaction volume, and sensitivity was improved by chopping the laser light and using phase-sensitive detection of the fluorescence via the lock-in technique. The output of the lock-in amplifier and an additional light power signal were passed to a 17-ms gated integrator operating under computer control. The computer was responsible for scanning the laser, controlling the integrator, and recording the integrator output at each laser frequency. Each scan of approximately 500 MHz width consisted of 4000 channels, each corresponding to a discrete laser frequency. A total light

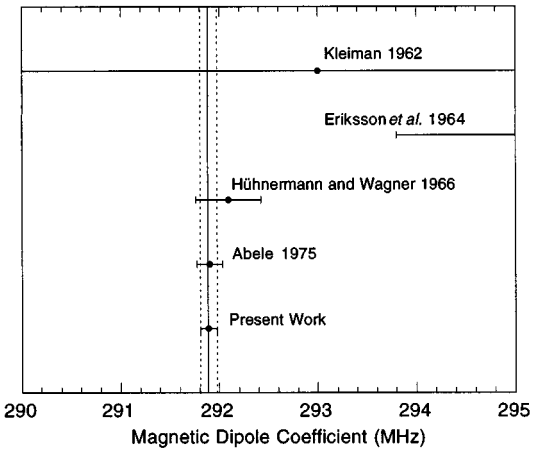


FIG. 1. Graphical comparison between previous measurements and our present result. Kleiman 293.0(3.7) MHz; Eriksson, Johansson, and Norlén, 304(11) MHz.

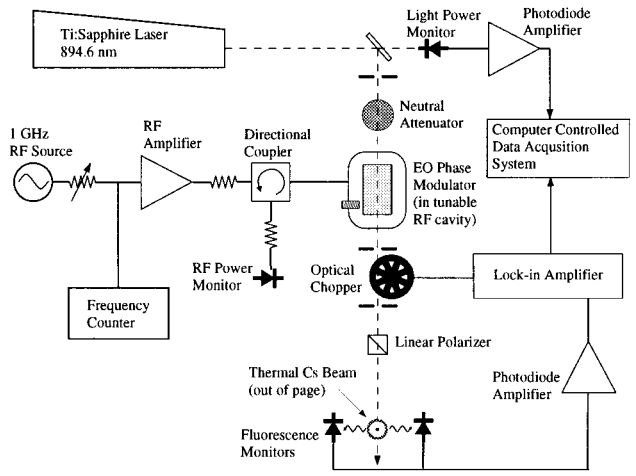


FIG. 2. Schematic diagram of the experimental apparatus.

power density of 190 μW/cm^2 was incident upon the atomic beam, with $\leqslant 60$ μW/cm^2 in each of the relevant frequency sidebands. Resolution was limited by the finite collimation of the atomic beam which gave rise to a fluorescence line shape 34 MHz wide at half maximum.

The energy splitting $\Delta\nu_{43}=\nu_4-\nu_3$ between the $F'=3$ and $F'=4$ levels of the $6p\ ^2P_{1/2}$ state was determined by measuring the separation of the $F'=4$ fluorescence peak and a second $F'=3$ fluorescence peak generated by the first-order rf sideband as the laser frequency was scanned through the $6s\ ^2S_{1/2}\rightarrow 6p\ ^2P_{1/2}$ resonance. Figure 3 shows the emitted fluorescence as a function of the laser scan parameter for large detunings where both the ground-state and excited-state hyperfine structures can be seen. Data scans were recorded over the narrow detuning range that includes the $F'=4$ peak and the $F'=3$ first-order sideband peak for numerous rf modulation frequencies. Our rf signal-generator–amplifier–EOM combination had an effective tuning range of $\nu_{\rm rf}=920-1080$ MHz, therefore we measured the $F'=4$ peak to

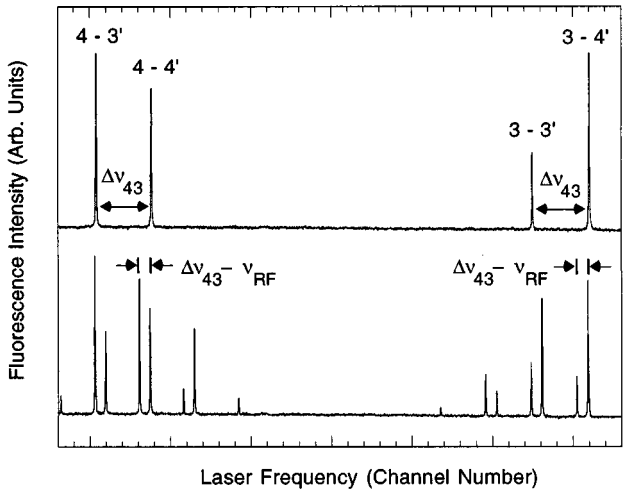
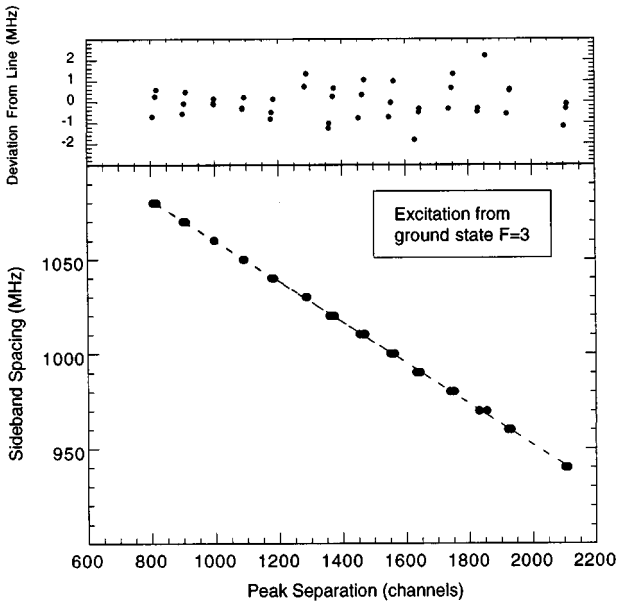


FIG. 3. Shown is a wide scan of detected fluorescence as a function of laser detuning for the $6s\ ^2S_{1/2}\rightarrow 6p\ ^2P_{1/2}\ F'$ transitions. The upper trace shows the hyperfine structure of both the ground and excited states without sidebands. The lower trace shows a similar scan with a rf sideband frequency of 940 GHz.



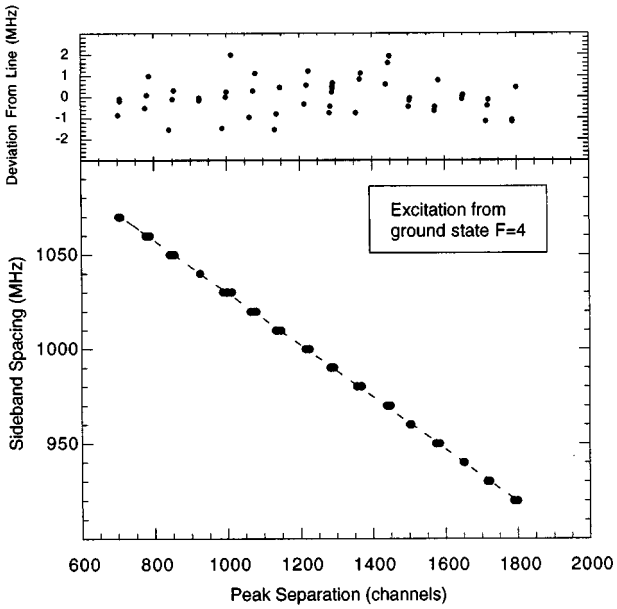


FIG. 4. Separation between $F'=4$ (zeroth-order sideband) to $F'=3$ (first-order sideband) laser fluorescence peaks as a function of rf sideband frequency with the residuals to a least-squares linear fit to the data.

$F'=3$ first-order sideband interval ($\Delta\nu_{43}-\nu_{\rm rf}$) over a rf tuning range from about 90 to 250 MHz. We then extrapolated to find the rf value at which the peak-to-sideband separation vanishes, which exactly corresponds to the hyperfine interval $\Delta\nu_{43}$. Two sets of measurements were performed which correspond, respectively, to excitation from each of the $6s\ ^2S_{1/2}\ F=3$ and $F=4$ ground-state levels, and permitted two sets of slightly different laser operating conditions. Each laser scan was fit to an appropriate sum of line profiles to determine the distance between the line centers. In Fig. 4 we show a plot of the sideband frequency as a function of the distance between the $F'=4$ peak center and the $F'=3$ sideband peak center. For excitation from the $F=3$ hyperfine ground state, we acquired 44 laser scans at 14 different rf

values, and a linear fit to the peak separation yielded a $\Delta\nu_{43}-\nu_{\mathrm{rf}}=0$ intercept of 1167.49(45) MHz. When exciting from the $F=4$ ground-state level, 52 observations were recorded at 16 frequencies, and an intercept of 1167.59(46) MHz was obtained. The uncertainties represent the statistical precision in determining the intercepts from a linear least-squares fit to the sideband frequency. From the weighted average of these intercepts, we determine the hyperfine splitting of the $6p\ ^2P_{1/2}$ state to be 1167.54(32) MHz.

By far, the most significant contribution to our experimental uncertainty comes from random errors which are reflected by the statistical uncertainty in determining the intercepts. These random errors have their origin in the determination of the peak positions and are dominated by the finite fluorescence linewidths with small contributions from random laser and rf source jitter. In addition, there are systematic effects which might shift $\Delta\nu_{43}$ towards greater or lesser values, but these are insignificant compared to the statistical uncertainty as discussed below. The systematic effects include any laser scan nonlinearity which is repeatable from scan to scan, Doppler shifts, magnetic field effects, and light-intensity-dependent effects.

We have investigated the laser scan nonlinearity for wide (5–10 GHz) tuning ranges using the ''picket fence'' of sideband peaks present in the atomic-beam fluorescence spectrum using the hyperfine structure of both the $6s\ ^2S_{1/2}\rightarrow 6p\ ^2P_{1/2}\ D_1$ and $6s\ ^2S_{1/2}\rightarrow 6p\ ^2P_{3/2}\ D_2$ resonance transitions as a frequency map. The smaller splitting and the high precision to which it is known makes the $6p\ ^2P_{3/2}\ F'=2,3,4,5$ hyperfine structure ideal for this purpose [2]. We find that the maximum departure of the laser frequency from that which is expected from a linear scan is typically $\leqslant 0.05\%$ of the scan width for a wide range of operating conditions; however, the functional form of the departure from linearity is not reproducible. During data collection, it was also observed that the entire spectrum can be shifted to the red or blue within the scan window using the laser manufacturer's frequency offset control with no observable change in the peak spacing. We therefore estimate that, for the narrow frequency interval (150 MHz) used in the present measurement, the contribution to the random error in peak separation from scan to scan due to laser tuning nonlinearities is approximately 0.075 MHz, and it is our opinion that any systematic shift due to nonlinearities is concealed far beneath the total random error.

Misalignment of the laser–atomic-beam intersection can give rise to small Doppler shifts which alter the value of $\Delta\nu_{43}$. Since the various optical sidebands produced in the EOM spatially overlap in the output beam, the leading contribution arises only from the fact that the peaks in the fluorescence spectrum are at slightly differing optical frequencies. The resulting shift would be proportional to the hyperfine splitting and the projection of the atomic beam velocity along the laser direction. Using the most probable velocity of the Maxwellian distribution at the cesium oven temperature of 400 K, and an angular misalignment of $\pm 2.5°$ corresponding to the maximum possible in the apparatus, one would expect a maximum possible shift of 37 Hz, which is clearly negligible. The small transverse temperature of the atomic beam (measured $\sim$3 K) gives rise only to the observed thermal linewidth of 34 MHz, but no shift due to the cylindrical symmetry.

We have investigated optical pumping as a possible source of systematic error which might depend on power. At a fixed EOM drive frequency, we measured the peak separation over a range of laser powers from 10 to 1000 μW and found no dependence on laser power. This result originates from our choice of experimental parameters which limit the possibility of differential line-shape asymmetries caused by optical pumping in two ways. First, the diameter of the laser beam, the resonant laser power, and the interaction cross section in combination limit a typical atom to the possibility of interacting with only one photon during a transit time through the laser beam. Second, linear polarization was chosen for the excitation process because it results in a symmetric magnetic sublevel distribution in cases where a single atom has the opportunity to experience more than one excitation-emission cycle. The residual magnetic field in the interaction region of the apparatus was measured with a Hall effect Gaussmeter to be 0.4 G, oriented parallel to the atomic-beam direction. The magnetic effects are thus limited to a broadening of the resonance fluorescence as the unresolved magnetic levels begin to move apart. We also simulated these effects by diagonalizing the atomic Hamiltonian in fields of up to 30 G and calculating the resulting line profiles. Because of the equal displacements and populations of opposing magnetic sublevels, fitting the simulated line shapes revealed no systematic shifts. (We also used the simulated broadened line profiles to determine the effects of a magnetic field on the uncertainty in locating the line centers, and found that this is also negligible below about 10 G.) The second-order energy shifts due to the magnetic field were also computed for the levels $F'=3$ and $F'=4$, and found to be an insignificant 5-Hz correction to the hyperfine splitting for the measured field.

Other corrections to the splitting which scale with the intensity of the laser radiation were also considered. The minute amount of power broadening is not significant, nor is the light shift of the hyperfine levels due to coupling through the laser electric field which we calculate to be less than 50 Hz for each of the levels. In summary, we do not expect the total systematic shift in the observed value of the hyperfine interval to exceed 200 Hz, or about 0.06% of the stated statistical uncertainty due to random errors.

We have also studied the residuals of our fitted line shapes for the presence of asymmetries and the effects these might have on our ability to measure peak separations. We extracted from our data the residual differential line shapes (RDL) for each of the laser scans. Within a set of laser scans at fixed rf, the RDLs were not statistically significant, therefore we calculated an average residual differential line shape (ARDL) that included all scans. The maximum excursions of the ARDL were less than 2% of the overall peak height and asymmetric features were less than 0.5% of the peak height. We calculated the effect of the ARDL on our ability to determine peak separations to be less than 4% of the statistical uncertainty in the separation. This analysis demonstrates the insignificance of residual differential asymmetries in our data irrespective of their origins.

The primary motivation for this work is to provide a precise test of the atomic theory necessary for the interpretation of PNC experiments in cesium. Our result for the $6p\ ^2P_{1/2}$ hyperfine splitting yields a magnetic dipole coefficient of $A = 291.89(8)$ MHz in good agreement with the measurement of $A = 291.90(12)$ by Abele [12]. The theoretical results of Ref. [17] determine a value for $A = 292.67$ MHz through a relativistic all-order many-body perturbation theory approach. This theoretical result differs from our measurement by approximately ten times our experimental uncertainty. Our result also agrees well with previous results and represents an improvement in the best previous measurement.

We would like to thank H. G. Berry and A. E. Livingston for helpful discussions. We gratefully acknowledge support from National Science Foundation Grant No. PHY95-04889.

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