

# Hyperfine splittings and isotope shifts of $(6s)^2\ ^1S_0 \rightarrow (6s6p)\ ^1P_1$ transition in barium

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**Abstract:** The hyperfine and isotope shifts of the  $(6s)^2\ ^1S_0 \rightarrow (6s6p)\ ^1P_1$  transition in barium were measured using a ring dye laser to excite an atomic beam. The laser frequency was scanned across the transition while fluorescence produced by the radiative decay of the excited state was detected. The frequency was calibrated using an acousto-optic modulator to frequency shift part of the laser beam. The spectrum therefore consisted of pairs of peaks separated by the modulation frequency. The shifts (MHz) relative to  $^{138}\text{Ba}$  are  $^{137}\text{Ba}\ F = 5/2, 63.43 \pm 0.25$ ;  $^{135}\text{Ba}\ F = 5/2, 120.55 \pm 0.25$ ;  $^{136}\text{Ba}, 128.02 \pm 0.39$ ;  $^{137}\text{Ba}\ F = 3/2, 274.56 \pm 0.09$ ;  $^{135}\text{Ba}, 323.44 \pm 0.19$ ; and  $^{135,137}\text{Ba}\ F = 1/2, 549.47 \pm 0.12$  where the transition in  $^{138}\text{Ba}$  occurs at the lower frequency. The magnetic dipole  $a$  and electric quadrupole  $b$  hyperfine coupling constants (MHz) of the  $(6s6p)\ ^1P_1$  state were determined to be  $a_{135} = -98.16 \pm 0.14$ ,  $b_{135} = 34.01 \pm 0.22$ ,  $a_{137} = -109.50 \pm 0.13$ , and  $b_{137} = 50.09 \pm 0.21$ .

**Résumé :** Les déplacements hyperfins et isotopiques de la transition  $(6s)^2\ ^1S_0 \rightarrow (6s6p)\ ^1P_1$  dans le baryum ont été mesurés en utilisant un laser annulaire à colorant pour exciter un faisceau atomique. La fréquence laser balayait la région de la transition pendant que la fluorescence produite par la décroissance de l'état excité était détectée. La fréquence était calibrée en utilisant un modulateur électro-acoustique pour décaler la fréquence d'une partie du faisceau laser. Le spectre était par conséquent constitué de paires de pics séparés par la fréquence de modulation. Les déplacements, relativement à  $^{138}\text{Ba}$ , sont (en MHz):  $^{137}\text{Ba}\ F = 5/2, 63,43 \pm 0,25$ ;  $^{135}\text{Ba}\ F = 5/2, 120,55 \pm 0,25$ ;  $^{136}\text{Ba}, 128,02 \pm 0,39$ ;  $^{137}\text{Ba}\ F = 3/2, 274,56 \pm 0,09$ ;  $^{135}\text{Ba}, 323,44 \pm 0,19$ ; and  $^{135,137}\text{Ba}\ F = 1/2, 549,47 \pm 0,12$  où la transition dans  $^{138}\text{Ba}$  se trouve à la fréquence inférieure. Les valeurs déterminées pour les constantes de couplage hyperfin de dipôle magnétique  $a$  et de quadrupôle électrique  $b$ , dans l'état  $(6s6p)\ ^1P_1$ , sont (en MHz):  $a_{135} = -98,16 \pm 0,14$ ,  $b_{135} = 34,01 \pm 0,22$ ,  $a_{137} = -109,50 \pm 0,13$ , et  $b_{137} = 50,09 \pm 0,21$ .

[Traduit par la rédaction]

## 1. Introduction

Measurements of hyperfine and isotope shifts of atomic transitions are important for studying nuclear structure [1]. A number of experiments have investigated barium [2–7], which has naturally occurring isotopes with atomic mass units 138 (71.7%), 137 (11.3%), 136 (7.8%), 135 (6.6%), 134 (2.4%), and 132 (0.1%). The nuclear spin for the even isotopes is zero and for the odd isotopes is 3/2. The  $(6s)^2\ ^1S_0 \rightarrow (6s6p)\ ^1P_1$  transition has been studied using a narrow linewidth laser to excite an atomic beam [2,3]. This permitted the determination of the hyperfine and isotope shifts of the  $^1P_1$  state with a resolution of 0.5 MHz. This paper reports results for the same state having uncertainties between 0.09 and 0.39 MHz.

Nowicki et al. [2] used an atomic beam consisting of  $^{138}\text{Ba}$  and a second beam composed of another barium isotope. The two atomic beams were excited using two separate narrow linewidth lasers. The laser beams intersected the atomic beams orthogonally to eliminate the first-order Doppler shift. Each dye-laser frequency was locked to the atomic transition by maximizing the fluorescence produced

by the radiative decay of the excited state back to the ground state. The frequency difference of the two laser beams was determined by focussing the two laser beams onto a fast photodiode that measured the beat frequency. The isotope shifts and hyperfine splittings were determined with an uncertainty of 0.5 MHz.

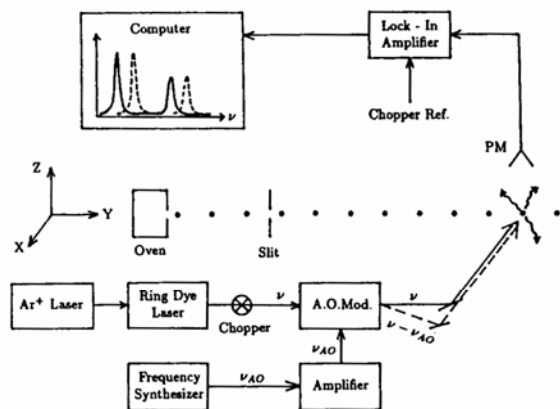
Baird et al. [3] used a narrow linewidth laser to excite an atomic beam consisting of natural barium. The laser frequency was scanned across the  $(6s)^2\ ^1S_0 \rightarrow (6s6p)\ ^1P_1$  resonance while fluorescence was detected by a photomultiplier. The change in laser frequency was monitored by passing part of the laser beam through a confocal Fabry–Perot etalon. A maximum in laser transmission occurred whenever the frequency changed by an amount equal to the cavity's free spectral range. The cavity was stabilized using a HeNe laser whose wavelength was locked to an iodine transition. The results had uncertainties between 0.6 and 1.3 MHz.

The present experiment used a single laser to excite an atomic beam. Part of the dye laser was frequency-shifted using an acousto-optic modulator. The frequency-shifted and unshifted laser beams were superimposed and the laser frequency was scanned across the resonance. Each transition therefore generated two peaks in the fluorescence spectrum that were separated by the modulation frequency, permitting the frequency to be calibrated. This method has been shown to be useful for measuring Stark, hyperfine, and isotope shifts [8–12]. The results have been found to be in agreement with

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Fig. 1. Apparatus. See text for discussion.



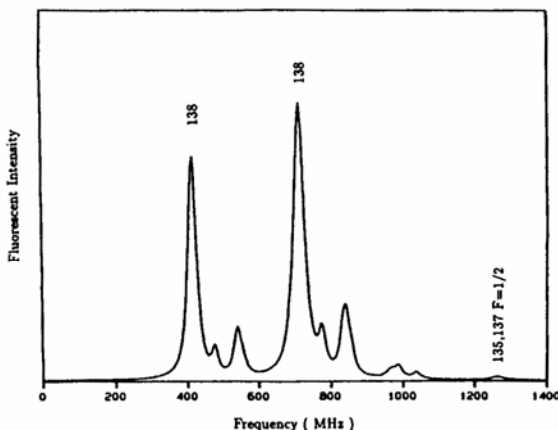
data obtained by other groups using different methods such as quantum beat spectroscopy and the optical double-resonance method [9–11]. For the Yb  $(6s)^2 \ ^1S_0 \rightarrow (6s6p) \ ^3P_1$  transition [8], the hyperfine and isotope shifts were consistent with the most accurate data produced by four previous experiments, each of which used a Fabry–Perot interferometer to calibrate the frequency. In this paper, the apparatus is described in Sect. 2. The data analysis and results are presented in Sect. 3.

## 2. Apparatus

The apparatus is illustrated in Fig. 1. It is described elsewhere and will therefore be only briefly discussed [12–13]. An atomic beam was produced using an oven that heated the barium metal close to its melting point of 725°C. The atoms were collimated by a series of slits producing a beam having a divergence of about 2 mrad. The oven and slits were enclosed by a vacuum chamber that was pumped by diffusion pumps and a liquid nitrogen trap to a pressure of about  $1 \times 10^{-7}$  Torr (1 Torr = 133.3 Pa).

The laser light was generated by a ring dye laser (Coherent 699) that was pumped by argon ion laser that produced 6 W of light at 514 nm. The dye-laser frequency was electronically stabilized giving a manufacturer quoted linewidth of 0.5 MHz. It produced several hundred milliwatts of light at 553.6 nm using rhodamine 110 laser dye. Part of the dye-laser beam was frequency-shifted using an acousto-optic (A.O.) modulator (Brimrose TEF 27-10). The modulator consisted of a tellurium oxide crystal that had a diffraction efficiency of over 50% for shifting light at 590 nm by 220–320 MHz. The modulation frequency was supplied by a frequency synthesizer (Hewlett Packard 8647A) with an accuracy of one part in  $10^6$ , and then amplified. A frequency of 300.000 MHz was used, which equalled the amplifier bandwidth.

The frequency-shifted and unshifted laser beams were superimposed using 0.1 cm wide slits located 100 cm apart on either side of the vacuum chamber. The lasers intersected the atomic beam orthogonally to eliminate first-order Doppler shifts. Power broadening was minimized by attenuating the laser beams to a power of a few milliwatts using neutral density filters. The laser polarization could be selected to be parallel to either the  $z$  (vertical) or  $y$  (horizontal) directions using appropriately oriented linear polarizers.

Fig. 2. Excitation of  $(6s)^2 \ ^1S_0 \rightarrow (6s6p) \ ^1P_1$  barium transition by frequency-shifted and unshifted laser beams.

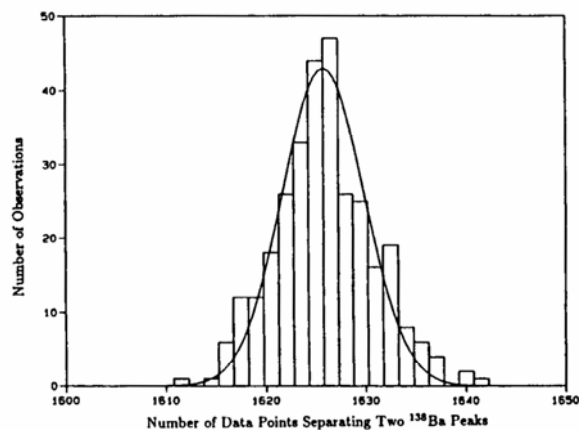
Fluorescence resulting from the radiative decay of the  $^1P_1$  state back to the ground state was detected by a photomultiplier (Hamamatsu R928) labelled PM in Fig. 1. The region where fluorescence was observed was carefully shielded to reduce laser scatter to a negligible amount. The photomultiplier signal was sent to a lock-in amplifier (Stanford Research 850) whose reference was supplied by a chopper that modulated the laser beam. The demodulated lock-in signal was recorded 256 times per second as the laser frequency was scanned across the resonance. A typical wavelength scan consisted of about 7500 data points, which were stored on a disk. The lock-in amplifier fitted a Gaussian function to each peak to determine its maximum-intensity position.

## 3. Data analysis and results

The laser frequency was calibrated using data as shown in Fig. 2. Here, the atoms were excited by two laser beams whose frequency differed by the acousto-optic modulation frequency of 300.000 MHz. Both laser beams were linearly polarized along the horizontal ( $y$ ) direction. The number of data points separating the two  $^{138}\text{Ba}$  peaks was found for 309 separate wavelength scans. This number varied slightly from scan to scan due to changes in the laser scan speed. The linearity of the scan was tested in a previous experiment in which the frequency-shifted and unshifted laser beams excited the Yb  $(6s)^2 \ ^1S_0 \rightarrow (6s6p) \ ^3P_1$  transition. A spectrum consisting of eight pairs of fluorescent peaks, generated by the seven stable Yb isotopes, was obtained in a 7 GHz scan of the laser frequency across the resonance. The frequency was then calibrated at different times during the scan and no evidence of a scanning nonlinearity was observed. Figure 3 displays a histogram showing the number of times a particular interval separating the two  $^{138}\text{Ba}$  peaks generated by the frequency-shifted and unshifted laser beams, was observed. The average separation was found to be  $1625.78 \pm 0.30$  points. Hence, a single data point corresponded to a frequency interval of  $0.184\,527 \pm 0.000\,034$  MHz.

Figure 4 shows data taken with a single laser beam. One hundred and fifty scans were taken with the laser polarized along the horizontal  $y$  direction as in Fig. 4a. An additional

**Fig. 3.** Frequency calibration. The number of data points separating the two  $^{138}\text{Ba}$  peaks appearing in Fig. 2 was determined for 309 separate laser scans. The Gaussian function is centered about the average value of  $1625.78 \pm 0.30$  data points, which corresponds to the acousto-optic modulation frequency of 300.000 MHz.

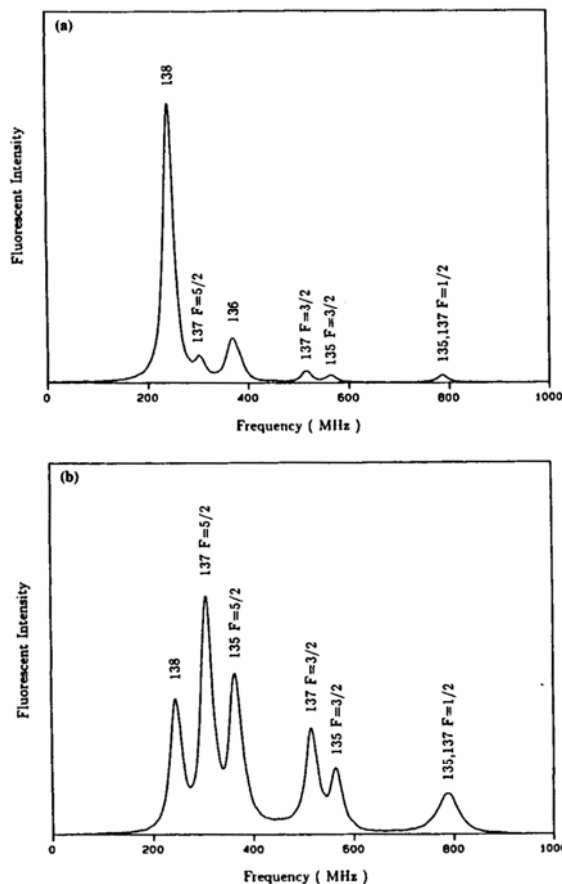


121 scans were recorded as in Fig. 4b where the laser was polarized parallel to the  $z$  axis. The  $^{136,138}\text{Ba}$  peaks were larger in Fig. 4a than those generated by  $^{135,137}\text{Ba}$  since the even isotopes do not have a nuclear spin. Hence,  $^{136,138}\text{Ba}$  act as classical electric dipole oscillators radiating primarily in the  $z$  direction, which was perpendicular to both the laser polarization and propagation directions.

The hyperfine splittings and isotope shifts can be obtained by fitting all of the peaks in the spectrum simultaneously. This has been done by Baird et al. [3] who fitted 18 data scans. They restricted the ratios of the hyperfine coupling constants of the  $^{135}\text{Ba}$  and  $^{137}\text{Ba}$  isotopes to equal those found in an earlier experiment [14]. It is claimed that the line centers could be determined with uncertainties between 0.7 and 1.2 MHz even though the frequency interval separating data points was 5 MHz. Each peak was modelled using at least three Lorentzian-like functions that were convolved with each other and contained several free parameters. It is claimed that this accounts for effects such as laser-frequency jitter, residual Doppler widths, and the natural linewidth. Additional parameters to account for optical pumping effects were used when fitting each of the hyperfine components generated by  $^{135,137}\text{Ba}$ . A detailed account showing precisely how the various fitting parameters affected the line center frequencies was not given. In contrast, Nowicki et al. [2] found that the spectrum was fitted satisfactorily using a pure Lorentzian lineshape.

This experiment obtains the hyperfine and isotope shifts by first determining the frequency intervals separating the various peak maxima. These results were then corrected for the overlap of nearby peaks that can cause the line center to be shifted slightly away from the peak maximum. These so-called frequency-pulling corrections will be discussed later. This method of data analysis is far simpler than that given in ref. 3 and reveals the effects of overlap of the various peaks. Furthermore, it is not at all clear that fitting the data using

**Fig. 4.** Excitation of barium using vertical and horizontal polarized light. The laser light was polarized along the  $y$  direction for Fig. 4a and parallel to the  $z$  axis for Fig. 4b. The peak labelled 136 in Fig. 4a contains a significant component due to the  $^{135}\text{Ba}$   $F = 3/2$  level. This causes the peak maximum to be  $1.3 \pm 0.39$  MHz closer to the  $^{138}\text{Ba}$  transition than if  $^{135}\text{Ba}$  were not present as is discussed in the text.



functions having many free parameters yields more accurate results.

The shifts of the various peak maxima relative to  $^{138}\text{Ba}$  are listed in Table 1. The results for  $^{136}\text{Ba}$ ,  $^{137}\text{Ba}$   $F = 3/2$ ,  $^{135}\text{Ba}$   $F = 3/2$ , and  $^{135,137}\text{Ba}$   $F = 1/2$  were determined using data as shown in Fig. 4a. The uncertainties were obtained by summing the standard deviation of the observed shifts about their average value along with the uncertainty arising from the frequency calibration. The positions of the  $^{137}\text{Ba}$   $F = 5/2$  and  $^{135}\text{Ba}$   $F = 5/2$  maxima shown in Fig. 4b were measured relative to  $^{135,137}\text{Ba}$   $F = 1/2$  since the  $^{138}\text{Ba}$  peak was significantly distorted by nearby peaks. The shifts relative to  $^{138}\text{Ba}$  were obtained using the  $549.47 \pm 0.12$  MHz separation of the  $^{138}\text{Ba}$  and  $^{135,137}\text{Ba}$   $F = 1/2$  peaks. Hence, the results for  $^{137}\text{Ba}$   $F = 5/2$  and  $^{135}\text{Ba}$   $F = 5/2$  have a larger uncertainty compared with the other data since they were found by subtracting two measured frequency intervals.

The difference between the line centers and the locations

**Table 1.** Ba  $(6s)^2\ ^1S_0 \rightarrow (6s6p)\ ^1P_1$  transition frequencies relative to  $^{138}\text{Ba}$  where the  $^{138}\text{Ba}$  transition occurs at the lower frequency.

$\Delta\nu(\text{MHz})$						
Isotope	$F$	Maxima diff.	Frequency-pulling correction	Final result	Ref. 2	Ref. 3
137	5/2	63.43±0.25	<0.05	63.43±0.25	62.8±0.6	63.0±0.6
135	5/2	120.75±0.24	-0.2±0.06	120.55±0.25	121.6±0.6	120.1±0.7
136		126.72±0.05	1.3±0.39	128.02±0.39	128.9±0.5	127.5±1.3
137	3/2	274.56±0.09	<0.05	274.56±0.09	275.1±0.6	274.5±0.9
135	3/2	322.94±0.12	0.5±0.15	323.44±0.19	326.6±0.6	324.2±0.8
135	1/2	549.47±0.12	<0.05	549.47±0.12	547.2±0.6	543.5±1.0
137	1/2				551.5±0.6	550.2±1.1

of the peak maxima were determined by fitting the spectra shown in Figs. 4a and 4b to the function

$$F(\nu) = \sum_i \frac{A_i}{1 + \left(\frac{\nu - \nu_i}{\Gamma/2}\right)^2} \quad (1)$$

Here,  $i$  is summed over the various isotope and hyperfine transitions. The optimum values of  $A_i$ ,  $\Gamma$ , and  $\nu_i$  were obtained using a least-squares fitting routine. The best-fit value of  $\Gamma$  was found to be about 25 MHz, which was power broadened from the natural linewidth of 19 MHz. Next, the frequencies where the peaks had maximum intensity were found and used to determine the frequency-pulling corrections listed in Table 1. The corrections for the  $^{137}\text{Ba}$   $F = 5/2$  and  $^{135}\text{Ba}$   $F = 5/2$  peaks were found by fitting data as shown in Fig. 4b. The remaining peaks were similarly studied using data displayed in Fig. 4a. The  $\pm 30\%$  uncertainty of these corrections was conservatively estimated by changing  $A_i$  by  $\pm 20\%$  from their best-fit values and varying  $\Gamma$  between 19 and 30 MHz.

The final results for the hyperfine splittings and isotope shifts, listed in Table 1, were obtained by adding the frequency-pulling corrections to the frequency intervals separating the various peak maxima. The respective uncertainties were summed in quadrature to give the error bars of the final results. The data are consistent with those obtained previously except for the shift of the  $^{135}\text{Ba}$   $F = 3/2$  level, which differs from the value obtained in ref. 2 but agrees with that found in ref. 3. Our value for the  $^{135,137}\text{Ba}$   $F = 1/2$  peak is consistent with the results of  $549.9 \pm 0.6$  and  $547.7 \pm 1.1$  MHz found in refs. 2 and 3, respectively, which were computed by weighting the shifts given in Table 1 for the  $F = 1/2$  level using the relative isotopic abundance of  $^{135,137}\text{Ba}$ .

The implications of the experimental results on the isotopic structure is now presented in Table 2 following an analysis given by Nowicki et al. [2]. The center-of-gravity isotope shift  $\Delta\nu$  of a transition at frequency  $\nu$ , can be expressed as the sum of the normal mass shift  $\Delta\nu_{\text{NM}}$ , the specific mass shift  $\Delta\nu_{\text{SM}}$ , and the so-called field shift  $\Delta\nu_{\text{F}}$  [15].

$$\Delta\nu = \Delta\nu_{\text{NM}} + \Delta\nu_{\text{SM}} + \Delta\nu_{\text{F}} \quad (2)$$

$\Delta\nu_{\text{NM}}$  is due to the change in the electronic reduced mass. Its

**Table 2.** Isotope shifts and change in nuclear radial moment  $\langle r^2 \rangle$ .

Atomic no.	$\Delta\nu$ (MHz)	$\Delta\nu_{\text{F}}$ (MHz)	$\delta\langle r^2 \rangle$ ( $10^{-2} \text{ fm}^2$ )
138	0.0		
137	215.15±0.16	231	-4.9
136	128.02±0.39	159	-3.4
135	259.29±0.17	307	-6.5

value for an isotope of barium with atomic number  $A$  relative to  $^{138}\text{Ba}$ , is given by

$$\Delta\nu_{\text{NM}} = \frac{m_e}{m_p} \frac{A - 138}{138A} \nu \quad (3)$$

where  $m_e$  is the electron mass and  $m_p$  is the proton mass.  $\Delta\nu_{\text{SM}}$  arises from the correlation of the momenta of the atomic electrons and has been estimated to be  $(0 \pm 1)\Delta\nu_{\text{NM}}$  for barium [2]. Finally,  $\Delta\nu_{\text{F}}$  is caused by a change of the nuclear-charge distribution and may be expressed as

$$\Delta\nu_{\text{F}} = F\delta\langle r^2 \rangle \quad (4)$$

where  $F$  is an electronic factor that has been calculated to be  $-4.7 \times 10^3 \text{ MHz fm}^{-2}$  [16] and  $\langle r^2 \rangle$  is the expectation value of the nuclear-charge moment squared. The negative sign of  $\delta\langle r^2 \rangle$  in Table 2 indicates that the neutron deficient isotopes of barium have smaller rms charge radii than  $^{138}\text{Ba}$ .

The hyperfine energies of  $^{135,137}\text{Ba}$  are found from the Hamiltonian

$$H = ah\mathbf{I} \cdot \mathbf{J} + bh \frac{[3(\mathbf{I} \cdot \mathbf{J})^2 + \frac{3}{2}\mathbf{I} \cdot \mathbf{J} - (\mathbf{I} \cdot \mathbf{I})(\mathbf{J} \cdot \mathbf{J})]}{2\mathbf{I}(\mathbf{I} - 1)\mathbf{J}(\mathbf{J} - 1)} \quad (5)$$

Here,  $h$  is Planck's constant,  $\mathbf{J}$  is the electronic angular momentum,  $\mathbf{I}$  is the nuclear spin,  $a$  and  $b$  are the magnetic-dipole and electric-quadrupole coupling constants, respectively. The energy splittings between the hyperfine levels of  $^{135,137}\text{Ba}$  are

$$E_{F=5/2} - E_{F=3/2} = \left(\frac{5}{2}a + \frac{5}{4}b\right)h \quad (6)$$

$$E_{F=3/2} - E_{F=1/2} = \left(\frac{3}{2}a + \frac{9}{4}b\right)h$$

**Table 3.** Magnetic dipole  $a$  and electric quadrupole  $b$  hyperfine coupling constants.

	<sup>135</sup> Ba		<sup>137</sup> Ba	
	$a(\text{MHz})$	$b(\text{MHz})$	$a(\text{MHz})$	$b(\text{MHz})$
Ref. 2	$-98.3 \pm 0.5$	$32.5 \pm 0.4$	$-109.8 \pm 0.5$	$49.7 \pm 0.5$
Ref. 3	$-97.76 \pm 0.14$	$32.29 \pm 0.47$		
This work	$-98.16 \pm 0.14$	$34.01 \pm 0.22$	$-109.50 \pm 0.13$	$50.09 \pm 0.21$

The left-hand sides of (6) were evaluated using data obtained in the present experiment for the shifts of the  $F = 5/2$  and  $3/2$  levels and the value for the  $F = 1/2$  level shift was measured by Nowicki et al. [2]. The resulting values for  $a$  and  $b$  are shown in Table 3. The magnetic-dipole constants found in ref. 2 and the present experiment agree closely. Reference 3 quotes a value for  $a_{135}$ , having an uncertainty of 0.14%, that is surprisingly more accurate than any of their measured frequency shifts listed in Table 1 from which it was presumably computed. The values obtained for the quadrupole moment of <sup>137</sup>Ba in ref. 2 and this experiment are consistent. However, our result for the quadrupole moment of <sup>135</sup>Ba is somewhat smaller than that found previously. This discrepancy is to be expected given the difference in the observed shift of the <sup>135</sup>Ba  $F = 3/2$  level mentioned earlier.

The hyperfine constants found for the <sup>135,137</sup>Ba isotopes can be checked using measurements of the nuclear magnetic-dipole  $\mu$  and electric-quadrupole moments  $Q$  as shown below [15].

$$\frac{a_{135}}{a_{137}} = \frac{\mu_{135}}{\mu_{137}} = \frac{0.83229}{0.93107} = 0.8939 \quad (7)$$

$$\frac{b_{135}}{b_{137}} = \frac{Q_{135}}{Q_{137}} = 0.651 \pm 0.001 \quad (8)$$

Equation (7) was evaluated using data obtained by a nuclear magnetic-resonance experiment [17,18] while the right-hand side of (8) has been determined using the optical double-resonance method [14]. The corresponding values obtained using the hyperfine coupling constants obtained in this experiment are  $a_{135}/a_{137} = 0.896 \pm 0.002$  and  $b_{135}/b_{137} = 0.679 \pm 0.007$ . It is not clear why the two values for  $b_{135}/b_{137}$  differ, although ref. 14 studied the  $(6s6p) {}^3P_1$  state while the present experiment examined the  $(6s6p) {}^1P_1$  state.

In conclusion, this experiment has yielded data for the isotope and hyperfine shifts of the barium  $(6s) {}^1S_0 \rightarrow (6s6p) {}^1P_1$  transition that have smaller uncertainties. The experiment can be improved by using isotopically enriched barium to reduce the overlap of various peaks. This should permit all of the isotope shifts to be determined with an accuracy of 0.1 MHz. Future refinements may include using an acousto-optic modulator capable of frequency shifting a laser by an amount equal to the various isotope shifts. One isotope of an atomic beam could then be excited by the unshifted laser while another isotope is excited by the frequency-shifted laser further along the atomic beam path. The laser frequency would then be scanned across the resonance. The isotope shift is then determined by measuring the acousto-optic frequency such that

the atoms in both regions are simultaneously in resonance. This procedure<sup>1</sup> is inherently insensitive to nonlinearities in the laser scan [19]. Hence, modulated laser beams are likely to play an increasingly important role in the precision measurement of isotope shifts and hyperfine splittings.

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<sup>1</sup> This method was not used in the present experiment as funds were not available to purchase the required acousto-optic modulator. It has been used to measure Stark shifts in cesium.