Isotope shifts of the $4s^2 {}^1S_0 \rightarrow 4s5p {}^1P_1$ transition and hyperfine splitting of the $4s5p {}^1P_1$ state in calcium

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(Received 23 December 2003; published 1 April 2004)

Using a technique based on production of ion Coulomb crystals, the isotope shifts of the $4s^2 {}^1S_0 \rightarrow 4s5p {}^1P_1$ transition for all naturally occurring isotopes of calcium as well as the hyperfine splitting of the $4s5p {}^1P_1$ state in ⁴³Ca have been measured. The field shift and specific mass shift coefficients as well as the hyperfine structure constants for ⁴³Ca have been derived from the data.

DOI: 10.1103/PhysRevA.69.042502

PACS number(s): 32.10.Fn, 42.62.Fi, 32.80.Pj

I. INTRODUCTION

Isotope shifts and hyperfine splitting of optical transitions provide valuable information about atomic electron configurations and not at least properties of the nuclei. While from isotope shifts, nuclear charge distributions can be deduced (see, e.g., Refs. [1,2]), from hyperfine splittings, nuclear spins, magnetic dipole moments, and electric quadrupole moments can be determined [3].

Although isotope shifts and hyperfine splittings already have been measured for a large number of transitions in neutral Ca (see Refs. [4–6] and references therein) and in singly charged Ca⁺ ions [7–9], the $4s^2 {}^{1}S_0 \rightarrow 4s5p {}^{1}P_1$ transition considered in the present paper has hitherto not been measured. In contrast to previous experiments, the present measurements use ion Coulomb crystals as means to obtain the spectroscopic data.

II. EXPERIMENTAL PROCEDURE

In the experiments, we have used a frequency doubled cw laser system tuned to the $4s^2 {}^1S_0 \rightarrow 4s5p {}^1P_1$ transition at a wavelength of $\lambda \approx 272$ nm. The degree of excitation at a particular wavelength is determined from the ion production rate when excited neutral Ca atoms absorb yet another λ \approx 272 nm photon directly from the 4s5p ${}^{1}P_{1}$ state or from the metastable $4s3d^{-1}D_0$ state, populated through spontaneous emission as depicted in Fig. 1(a) [10]. As shown in the sketch of the experimental setup in Fig. 2(a), the spectroscopic laser beam at $\lambda \approx 272$ nm crosses a well-collimated effusive thermal beam of calcium atoms at right angles in the center of a linear Paul trap [11]. This geometry is chosen in order to obtain small Doppler shifts of the transition under study as well as to have a large capture efficiency of the ions produced. Since the atomic beam is derived from an oven containing natural metallic calcium, all the isotopes ${}^{40}Ca (96.9\%), {}^{42}Ca (0.647\%), {}^{43}Ca (0.135\%),$ 44 Ca (2.086%), 46 Ca (0.004%), and 48 Ca (0.187%) are present. The detection of the ions produced within a given exposure period is done by first trapping them independently of the specific isotope, and then converting almost all ions into ${}^{40}Ca^+$ through near-resonant electron transfer collisions with atoms in the atomic beam containing 96.9% ${}^{40}Ca$ atoms. Finally, the number of trapped ions is counted with a near-100% efficiency by monitoring the fluorescence from the ${}^{40}Ca^+$ ions when laser cooled into a Coulomb crystal [12,13]. Below, the key parts of the applied technique are discussed in detail.

A. Isotope and hyperfine selective ion production

The laser system used for the spectroscopy consists of a tunable, single-mode ring cw dye laser operated at 544 nm with an output power of a few hundreds mW which is frequency doubled to 272 nm by a β -barium borate crystal placed in an external cavity. The output power at the desired $4s^2 {}^1S_0 \rightarrow 4s5p {}^1P_1$ transition wavelength is typically about \sim 10 mW, and the laser linewidth is below 1 MHz. This linewidth is much smaller than the full width at half maximum (FWHM) Doppler width of ~50 MHz due to the transverse velocity spread of the thermal atomic beam originating from an oven at a temperature of $\sim 600^{\circ}$ C. Hence, since the natural linewidth of the $4s^2 {}^{1}S_0 \rightarrow 4s5p {}^{1}P_1$ transition according to the NIST database [14] is known to be $\Gamma \approx 2 \text{ MHz}$ (>50% uncertainty), the Doppler effect is the resolution limiting factor in the experiments. With isotope shifts typically of the order of GHz and hyperfine splittings of about ~ 100 MHz of ⁴³Ca, not only can one selectively excite individual even isotopes (no nuclear spin), but also the three



FIG. 1. Simplified energy level diagrams of Ca (a) and Ca⁺ (b) of relevance to the experiments. The straight arrows indicate laser driving transitions, while the wavy arrow symbolizes spontaneous emission. In (a), the solid line beneath the hatched region indicates the ionization limit.

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FIG. 2. (a) Schematics of the experimental setup with an inset showing a side view of the imaging system. For the optical elements in the figure, the following notation has been used: M, mirror; DM, dichroic mirror; PBS, polarization beam splitter; and $\lambda/2$, half-wave-plate. (b) The linear Paul trap electrodes with applied rf voltages. The *z* axis defines the trap axis. The middle sections are 5.4 mm long and the end pieces are 20 mm each. The diameter of the electrodes is 8.0 mm and the minimum distance to the trap axis is 3.5 mm.

hyperfine components of the 4s5p ${}^{1}P_{1}$ state in the case of 43 Ca. As mentioned above, ions are produced from excited calcium atoms either by absorbing yet another $\lambda \approx 272$ nm photon directly from the 4s5p ${}^{1}P_{1}$ state, or from the metastable 4s3d ${}^{1}D_{0}$ state populated through spontaneous decay. By exposing the atomic beam to the photoionization laser beam with a certain frequency for a fixed period of time (in our experiment controlled by a mechanical light shutter), the ion production will reflect the excitation degree of the 4s5p ${}^{1}P_{1}$ state of calcium atoms at this frequency. Counting the number of produced ions as a function of laser frequency thus reveals the excitation spectrum.

B. The ion detection scheme

The central element of the ion detection is the linear Paul trap situated in a vacuum chamber which is operated at a pressure of $\sim 10^{-10}$ Torr. As sketched in Fig. 2(b), the trap consists of four sectioned cylindrical electrodes placed in a quadrupole configuration. Confinement of ions in the *xy* plane is obtained by an effective radial harmonical potential created by applying a rf-potential of amplitude U_{RF} and frequency Ω_{RF} to each of the electrodes in such a way that the voltage on the diagonally opposite electrode is 180° out of phase. Along the trap axis, the *z*-axis, confinement originates from a positive dc voltage U_{EC} applied to all the eight end-



FIG. 3. Image of a pure $^{40}\mathrm{Ca^+}$ Coulomb crystal consisting of ${\sim}10\ 000$ ions.

pieces. With the trap dimensions used [see Fig. 2(b)], for $U_{RF} \sim 200 \text{ V}$, $\Omega = 2\pi \times 3.9 \text{ MHz}$ and $U_{EC} \sim 5 \text{ V}$, the effective trap depth becomes $\sim 1 \text{ eV}$. This ensures that ions produced in the trapping region are efficiently trapped despite the thermal velocities of the neutral atoms.

When ions of a certain calcium isotope, e.g., ⁴⁰Ca⁺, are produced, they can be Doppler laser-cooled by driving the 4s $S_{1/2} \rightarrow 4p P_{1/2}$ and the $3d D_{3/2} \rightarrow 4p P_{1/2}$ transitions by laser light around 397 nm and 866 nm, respectively, as depicted in Fig. 1(b). As the ions are cooled to sufficiently low temperatures (~ 10 mK), they arrange in a spatial ordered structure, often referred to as an ion Coulomb crystal. In our setup, these ion Coulomb crystals are imaged onto an image intensified Charge-coupled device camera by collecting fluorescence from the Doppler cooled ions. A typical picture of a Coulomb crystal containing $\sim 10\ 000\ ^{40}$ Ca⁺ ions is shown in Fig. 3. Here, one clearly observes the elliptical projection of the crystal, which for our trapping potential is spheroidal with the trap axis (z axis) as the axis of rotational symmetry. Single-component Coulomb crystals in our trap have spatially uniform ion densities when viewed on length scales larger than the typical distance between neighboring ions (see Ref. [15]). The specific density is set by the trapping parameters. Hence, by determining the crystal volumes, e.g., by measuring the main axis of the projected ellipse, we can determine the total number of trapped ions.

Generally, when the frequency of the spectroscopy laser is tuned between the resonances the various isotopes, two or more singly charged calcium isotope ions will be produced and trapped. Such multispecies ion ensembles can be cooled into Coulomb crystals as well, with the lighter isotopes more tightly bound towards the trap axis than the heavier due to the dynamical confinement in the xy plane [15]. In Fig. 4(a), an image of the fluorescence from laser cooled ⁴⁰Ca⁺ ions in such a crystal is shown. The crystal consists mainly of ⁴⁰Ca⁺ and ⁴⁴Ca⁺ ions, where the nonfluorescing ⁴⁴Ca⁺ ions are cooled only sympathetically through the Coulomb interaction with the directly laser cooled ${}^{40}Ca^+$ ions [16,17]. The number of ions in the multicomponent crystal could principally be determined by imaging the fluorescence from the individual isotopes. This is, however, technically very demanding, since each isotope ion would need its own set of laser frequencies due to isotope shifts of the transitions shown in Fig. 1(b). As an alternative route to quantify the total ion production, we choose to expose the multicomponent crystals similar to the one in Fig. 4(a) to the thermal



FIG. 4. Image of a ${}^{40}Ca^+$ (fluorescing)– ${}^{44}Ca^+$ (nonfluorescing) two-component crystal of ~1500 ions before (a) and after (b) exposure to the atomic beam which leads to the electron transfer as described in the text. The white line indicates the outer boundary of the crystal.

atomic beam of calcium until nearly all the ions (\sim 96.9%) by near-resonant electron transfer collisions of the type

$${}^{A}\mathrm{Ca}^{+} + {}^{40}\mathrm{Ca} \rightarrow {}^{A}\mathrm{Ca} + {}^{40}\mathrm{Ca}^{+} \tag{1}$$

have been converted into ${}^{40}Ca^+$ ions, and subsequently we measure the size of the now nearly pure ${}^{40}Ca^+$ crystal. Since the energy difference between the two sides of Eq. (1) is just the isotope shift of the electronic levels, which is much smaller than the thermal energy of the atoms in the beam, no significant energy barrier exists for the process. Furthermore, since the kinetic energy of the produced ${}^{40}Ca^+$ ions is more than an order of magnitude smaller than the trap potential depth, essentially 100% of the ions produced through Eq. (1) stay trapped. In Fig. 4(b), the two-component crystal presented in Fig. 4(a) has been converted via the electron transfer process into a nearly pure ${}^{40}Ca^+$ crystal.

III. SPECTROSCOPIC RESULTS

A. The 4s5p $^{1}P_{1}$ excitation spectrum

Figure 5 shows the relative ion production rate or, equivalently, the relative rate at which calcium atoms are being excited to the ${}^{1}P_{1}$ state, as a function of the detuning of the spectroscopy laser. These relative rates were obtained from knowledge of the volumes of the created Coulomb crystals, the laser exposure times, and the intensities of the spectroscopy laser. The spectrum contains resonances of all the naturally abundant isotopes, including the one corresponding to ⁴⁶Ca which has a natural abundance of only 0.004%. The results presented in Fig. 5 have been obtained from three partially overlapping frequency scans. The three data series were measured at slightly different oven temperatures: $T_{oven} = 612 \,^{\circ}\mathrm{C} \,(\Delta), \qquad T_{oven} = 630 \,^{\circ}\mathrm{C} \,(\blacksquare),$ and Toven =602 °C (\Box). The relative rates of the Δ data and the \blacksquare data have been normalized, so they share the same fitted maximum value and position at the ⁴²Ca resonance peak. In the



FIG. 5. Semilogarithmic plot of the production rate of Ca⁺ ions as a function of the frequency detuning of the spectroscopy laser from the ⁴⁰Ca resonance. The data points presented are the results of three frequency scans made at slightly different temperatures $[(\Delta) T_{oven}=612^{\circ}\text{C}, (\blacksquare) T_{oven}=630^{\circ}\text{C}, \text{ and } (\Box) T_{oven}=602^{\circ}\text{C}]$. The full line indicates a Voigt profile fit to the ⁴⁰Ca resonance within the frequency range ~[-1300;700] MHz.

same way the data indicated by \blacksquare and \Box have been normalized to the three ⁴³Ca hyperfine peaks.

The temperature was during each experiment kept stable to better than ±2°C corresponding to a maximum uncertainty in the atomic density of about $\pm 7\%$ during the whole data accumulation time of ~ 40 min. Since the time to measure a single resonance is only a very small fraction of this time, any systematic errors in the resonance profiles due to fluctuating oven temperatures can be neglected. The Doppler broadening of the resonances, which only depends slightly on the atomic mass, is best found by fitting the ⁴⁰Ca resonance data to a Voigt profile. By doing this one obtains a total FWHM of \sim 54±5 MHz dominated by the Doppler broadening. At the laser intensity which we apply $(\sim 100 \text{ mW/cm}^2)$, we have assumed that neither the $4s^2$ ${}^{1}S_{0} \rightarrow 4s5p \; {}^{1}P_{1}$ transition nor the following photoionization process is saturated. Consequently, the ion production rate is expected to be proportional to the laser intensity squared. We accounted for fluctuations in the spectroscopic laser intensity during the data accumulation time by using this assumption. That this assumption is rather good is supported by noting that the relative measured resonance peak values in Fig. 5 are in good agreement with the natural abundances of the calcium isotopes, which are ${}^{40}Ca$ (96.9%), ${}^{42}Ca$ (0.647%), 44 Ca (2.086%), 46 Ca (0.004%), ⁴³Ca (0.135%), and ⁴⁸Ca (0.187%).

B. Isotope shifts

Before deriving the isotope shifts from data like the ones presented in Fig. 5, the uncertainty of the individual data points has to be estimated and taken into account. The uncertainty is essentially due to three effects. First, due to intensity fluctuations of the laser intensity there will be a fluctuation in the number of ions produced. Since for neighboring data points around a resonance this intensity fluctuation is maximally a few percent, the expected square

TABLE I. Isotope shifts of the transition $4s^2 {}^{1}S_0 \rightarrow 4s5p {}^{1}P_1$ in calcium derived from the experimental data like the ones presented in Fig. 5. All shifts are with respect to the 40 Ca resonance. The errors stated represent one standard deviation originating from the statistical errors in the experiments. In addition, the shifts are subject to an overall linear scaling uncertainty of 1% due to our frequency scan calibration (see text). The isotope shift for 43 Ca is the center of gravity of the hyperfine components.

Mass (A)	Shift (MHz)
42	967±9
43	1455 ± 9
44	1879 ± 14
46	2746 ± 16
48	$3528\!\pm\!16$

dependence on the intensity of the ion production leads to an estimated uncertainty of ~5%. Second, in determination of the Coulomb crystal volumes, both systematic and random uncertainties due to the measurements of the main axis of the elliptical projections of the crystals occur. Both these errors are of about ~5%, but since the systematic errors are equal for points symmetrically positioned around a resonance, it will have a small effect on the determination of the resonance frequencies. Hence, in our analysis, we have only accounted for the random errors of 5%. Finally, due to the finite number of ions produced within one measurement, we have included an uncertainty of the square root of the estimated number of ions. If all these uncertainties are accounted for in a weighted least-squares Gaussian fit, the uncertainty on the resonance position becomes lower than 2 MHz.

A more critical error arises when we compare similar scans. Here we find that the measured resonance peak positions are associated with much larger uncertainties than the ones from the Gaussian fits. This additional error originates from local frequency drift of the spectroscopy laser during a whole scan which typically lasts about 40 min. From a series of scans, we have found that this laser drift error leads to a rms uncertainty in the resonance frequencies of about 9 MHz. This last error source is the dominant one for the estimated errors of the isotope shifts given in Table I. In this table, the isotope shift for ⁴³Ca is found as the center of gravity of the three hyperfine components of ⁴³Ca which will be discussed in Sec. III C. In addition to these statistical errors, there is also a systematic uncertainty of $\pm 1\%$ arising from the calibration of the spectroscopy laser frequency scan to an optical spectrum analyzer with a known free-spectral range.

For completeness, we have estimated that we might have introduced an unimportant error of a few hundred kHz due to the fact that we have determined the resonance frequencies from single Gaussian fits instead as from a more realistic multipeak Voigt profile fit.

The isotope shift for a given transition is usually described as a sum of the mass and the field shift in the following way [1]:



FIG. 6. Plot of the values $[A'40/(A'-40)]\delta \nu_{RIS}^{40A'}$ derived from the measurements as a function of $[A'40/(A'-40)]\delta \langle r^2 \rangle^{40A'}$ for the $4s^2 {}^{1}S_0 \rightarrow 4s5p {}^{1}P_1$ transition in Ca. The dashed line is a weighted linear regression fit to determine the coefficients M_{SMS} and F from the relation given in Eq. (4).

$$\delta\nu^{AA'} = M \frac{A' - A}{AA'} + F \delta\langle r^2 \rangle^{AA'}, \qquad (2)$$

where *M* is the mass shift coefficient, *A* and *A'* denote the atomic masses of the two isotopes, *F* is the field shift coefficient, and $\delta \langle r^2 \rangle^{AA'}$ is the difference in mean square nuclear charge radii between the isotopes.

The mass shift is usually written as a sum of the normal mass shift (NMS) and the specific mass shift (SMS), which means that we can write the mass shift coefficient as $M = M_{NMS} + M_{SMS}$. Here the NMS coefficient is given by the simple expression $M_{NMS} = \nu_0 m_e/m_u$, where ν_0 is the transition frequency, m_e is the electron mass, and m_u is the atomic mass unit. The NMS originates from the reduced mass correction for the electron, while the SMS comes from the change in the correlated motion of all the electrons (see, e.g., Ref. [18]). Subtraction of the NMS from the total isotope shift gives the residual isotope shift (RIS),

$$\delta\nu_{RIS}^{AA'} = M_{SMS}(A' - A)/AA' + F\delta\langle r^2 \rangle^{AA'}.$$
 (3)

Rewriting Eq. (3) by multiplication by the factor AA'/(A' - A) leads to

$$\frac{A'A}{A'-A}\delta\nu_{RIS}^{AA'} = M_{SMS} + F\left(\frac{A'A}{A'-A}\delta\langle r^2\rangle^{AA'}\right),\tag{4}$$

which shows that M_{SMS} and F can be determined from a linear fit when the $\delta \langle r^2 \rangle^{AA'}$'s are known. Using values of the $\delta \langle r^2 \rangle^{AA'}$'s for calcium from Ref. [2], in Fig. 6, we have plotted the left-hand side of Eq. (4) using the values of Table II as a function of $[A'A/(A'-A)] \delta \langle r^2 \rangle^{AA'}$ for the fixed value of A=40. The SMS coefficient and field shift coefficient, obtained by weighted linear regression fit to the data points in Fig. 6, are listed in Table II.

The field shift coefficient for the $4s^2 {}^{1}S_0 \rightarrow 4s5p {}^{1}P_1$ transition (Table II) is, within the stated error, almost equal to the experimentally determined field shift coefficient for the $4s^2 {}^{1}S_0 \rightarrow 4s4p {}^{1}P_1$ transition of $F=-175.8\pm1.2$ MHz/fm² reported in Ref. [5]. This can be attributed to the fact that a *p* electron has negligible overlap with the nucleus compared to

ISOTOPE SHIFTS OF THE $4s^2 {}^1S_0 \rightarrow 4s5p {}^1P_1$ TRANSITION...

TABLE II. The specific mass shift M_{SMS} and field shift *F* coefficients for the $4s^2 {}^1S_0 \rightarrow 4s5p {}^1P_1$ transitions of calcium derived from the the linear fit presented in Fig. 6. The first stated uncertainty estimate is the one standard deviation obtained from the linear regression to the data of Fig. 6. The systematic error in the isotope shifts due to the frequency scan calibration is included as the second uncertainty estimate. For completeness the NMS coefficient is M_{NMS} =604.3 GHz amu.

M_{SMS} (GHz amu)	$F (\mathrm{MHz/fm^2})$
$243{\pm}3{\pm}9$	$-179{\pm}39{\pm}2$

an *s* electron, so the main contribution to the field shifts is expected in both cases to come from the *s* electrons.

C. ⁴³Ca hyperfine splitting

The nuclear spin of I=7/2 for ⁴³Ca leads to three hyperfine levels of the ¹P₁ state with total spins F=5/2,7/2, and 9/2, respectively. The hyperfine structure (hfs) constants and isotope shift of ⁴³Ca are determined by fitting to the normal Casimir formula [3]

$$\Delta E_F = \Delta \nu_{cg} + \frac{A}{2}C + \frac{B}{4} \frac{\frac{3}{2}C(C+1) - 2I(I+1)J(J+1)}{(2I-1)(2J-1)IJ}, \quad (5)$$

where C = F(F+1) - I(I+1) - J(J+1), $\Delta \nu_{cg}$ is the isotope shift of the center of gravity of the hfs, and *A* and *B* are the magnetic dipole and electric quadrupole coupling constants, respectively. Several scans across the three hyperfine resonances were made to increase the level of confidence of the *A* and *B* constants. The hfs constants derived from these scans are summarized in Table III, while the center of gravity for ⁴³Ca has been given in Table I. The small value of the *B* constant indicates that the magnetic dipole coupling has the most prominent contribution to lifting the degeneracy of the 4s5p ${}^{1}P_{1}$ level for ${}^{43}Ca$. Compared with the

TABLE III. hfs constants for the 4s5p $^{1}P_{1}$ state in 43 Ca. The first stated uncertainties originate from statistical errors in determining the resonance positions, while the second account for the systematic errors due to frequency calibration uncertainty.

A (MHz)	B (MHz)
$39.8 \pm 0.8 \pm 0.4$	$-0.3 \pm 3 \pm 0.03$

work of Ref. [5], where the hfs constants for the $4s4p \ ^1P_1$ state in 43 Ca have been measured to be $A = -15.54 \pm 0.03$ MHz and $B = -3.48 \pm 0.13$ MHz, the $4s5p \ ^1P_1$ state has an opposite sign for the A constant and the B constant is the same order of magnitude or smaller.

IV. CONCLUSION

With a technique based on determining the number of ions produced, collected, and cooled into Coulomb crystals through resonant two-photon ionization, the isotope shifts of the $4s^2 S_0 \rightarrow 4s5p \ ^1P_1$ transition for all naturally occurring isotopes of calcium as well as the hyperfine splitting of the $4s5p^{-1}P_1$ state in ⁴³Ca have been measured. The field shift and specific mass shift coefficients as well as the hyperfine structure constants for ⁴³Ca have been derived from the data. Though in the presented experimental scheme, near-resonant charge exchange collisions were used to gain information about the ion number, it is not a requirement for the technique for these to be used. Actually, the technique should be applicable to many atomic species as long as some laser cooled ions are simultaneously trapped and can sympathetically cool the species of interest into a Coulomb crystal [17,19,20]. From the spatial organization of the observable laser cooled species, one can easily deduce the number of the, e.g., nonfluorescing ions of interest.

ACKNOWLEDGMENTS

This work was supported by the Danish National Research Foundation and the Carlsberg Foundation.

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