Precision study of \(6p^2P_j \rightarrow 8s^2S_{1/2}\) relative transition matrix elements in atomic Cs

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A combined experimental and theoretical study of transition matrix elements of the \(6p^2P_j \rightarrow 8s^2S_{1/2}\) transition in atomic Cs is reported. Measurements of the polarization-dependent two-photon excitation spectrum associated with the transition were made in an approximately 200 cm\(^{-1}\) range on the low frequency side of the \(6s^2S_{1/2} \rightarrow 6p^2P_{1/2}\) resonance. The measurements depend parametrically on the relative transition matrix elements, but also are sensitive to far-off-resonance \(6s^2S_{1/2} \rightarrow np^2P_j \rightarrow 8s^2S_{1/2}\) transitions. In the past, this dependence has yielded a generalized sum rule, the value of which is dependent on sums of relative two-photon transition matrix elements. In the present case, best available determinations from other experiments are combined with theoretical matrix elements to extract the ratio of transition matrix elements for the \(6p^2P_j \rightarrow 8s^2S_{1/2}\) \((j = 1/2,3/2)\) transition. The resulting experimental value of 1.423(2) is in excellent agreement with the theoretical value, calculated using a relativistic all-order method, of 1.425(2).

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I. INTRODUCTION

Although some of the earliest experiments in atomic physics were measurements of atomic lifetimes and oscillator strengths [1, 2], precise determination of atomic transition matrix elements remains a demanding enterprise [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16]. Measurements made for non-alkali atoms, often motivated by the need for data in some other area of atomic physics research [3, 4, 5], have typically cited even lower precision. We have similarly been concerned with precise measurements in alkali atoms, and we have a continuing interest in the absolute transition matrix elements made for non-alkali atoms, often motivated by the need for data in some other area of atomic physics research [3, 4, 5]. Some perspective on this may be gained by noting that, in spite of the development of a wide array of sophisticated experimental techniques, measurements depending directly on atomic transition matrix elements seldom have achieved a precision better than \(\sim 0.5\%\) this being for the deeply studied alkali atoms \([3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16]\). Measurements made for non-alkali atoms, often motivated by the need for data in some other area of atomic physics research [3, 4, 5], have typically cited even lower precision. We have similarly been concerned with precise measurements in alkali atoms, and we have a continuing interest in the absolute transition matrix elements made for non-alkali atoms, often motivated by the need for data in some other area of atomic physics research [3, 4, 5].

In spite of the fact that many experimental [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16] and theoretical [27, 28, 29, 30, 31, 32] approaches have been applied to determination of atomic properties of atomic \(^{133}\text{Cs}\), including lifetimes or oscillator strengths and polarizabilities, only limited higher precision experimental data is available for many valence transition matrix elements in this atom. These studies have been motivated in part by a serious need for empirical data to extract more fundamental information from precise measurement of parity nonconservation (PNC) in this atom [33], especially in light of the discrepancy reported in [34] between values of the weak charge \(Q_w\) extracted from high precision atomic physics experiment [33] and the accepted Standard Model value. In order to clarify the situation, recent experimental efforts have concentrated on measurement of transition matrix elements associated with the \(6s - 6p\) and \(6s - 7p\) multiplet transitions [12, 13]. However, the \(6p - 7s\) and \(7p - 7s\) transitions, which are less experimentally accessible, make similarly important contributions. The availability of high-precision experimental data for any transitions between low-lying states of Cs is important for providing additional information regarding the accuracy of the theoretical calculations in Cs which is crucial for the accurate analysis of Cs PNC experiment.

In this paper we present results of our measurements of the spectral and polarization dependence of the \(6s^2S_{1/2} \rightarrow 6p^2P_j \rightarrow 8s^2S_{1/2}\) \((j = 1/2,3/2)\) two-color, two-photon transition in \(^{133}\text{Cs}\), where \(n = 6\) is the dominant term. In an earlier report [22] we described how such measurements could be interpreted in terms of a type of sum rule related to the scalar and vector transition probabilities. The sum rule is evaluated by fitting experimental polarization-dependent spectra to a generalized form containing, as fitting parameters, the relative two-photon transition matrix elements for the contributing transitions, allowing extraction of the ratio of the matrix elements.
elements in the dominant term. The contributions from far-off-resonance transitions are small but significant for heavy atoms such as Cs and are evaluated theoretically. In the present report we describe that approach as applied to the Cs $6s^2S_{1/2} \rightarrow 8s^2S_{1/2}$ two-photon transition. By combining the present measurements with precisely determined $6s^2S_{1/2} \rightarrow 6p^2P_j$ resonance transition matrix elements, the $6p^2P_j \rightarrow 8s^2S_{1/2}$ relative excited state matrix elements are determined. We point out that the same combined scheme can effectively be used to determine matrix elements for the $6s^2S_{1/2} \rightarrow 7p^2P_j \rightarrow 7, 8s^2S_{1/2}$ transitions which, because of their importance for analysis of the precision parity nonconservation measurements in Cs, remain interesting cases to investigate.

In the following sections, we briefly review our experimental approach, with particular attention to aspects new in the present study. This is followed by a description of our experimental results and data analysis. We then describe the application of results of relativistic many-body calculations of the off-resonance terms, which allows the extraction of the desired two-photon transition matrix element ratios. Finally, the relative $6p^2P_j \rightarrow 8s^2S_{1/2}$ relative transition matrix elements are determined and compared with theoretical calculations.

II. EXPERIMENTAL APPROACH

The general experimental approach is described in previous reports \[21, 22, 24, 25\], and so will only briefly be reviewed here. The basic experimental scheme \[21\] is illustrated in Figure 1, which contains a partial energy level diagram for the lowest few levels of atomic Cs. A block diagram of the experimental apparatus is shown in Figure 2. In the experiment, atoms in the $6s^2S_{1/2}$ ground level are excited by two-photon absorption to the $8s^2S_{1/2}$ final level. In principle, all the intermediate $np^2P_j$ levels, including continuum terms, contribute to the total excitation probability \[32\]. In practice, and depending on the precision of the measurements, significant contributions are limited to the first few nearest-to-resonance terms. The first step of the excitation scheme is accomplished with an Ar$^+$ laser pumped Ti:Sapphire laser (laser 1) tuned in a several-hundred cm$^{-1}$ energy range to the red of the $6s^2S_{1/2} \rightarrow 6p^2P_j$ transition, which has a hyperfine-weighted transition frequency of $\omega_{3/2} = 11732.31$ cm$^{-1}$ \([36, 38]\). With reference to Figure 1, the detuning from one-photon resonance is defined as $\Delta = \omega_1 - \omega_{3/2}$, where the laser 1 frequency is $\omega_1$. Although detunings of magnitude greater than 200 cm$^{-1}$ were investigated, useful data was obtained only for $|\Delta| < 200$ cm$^{-1}$. A Michelson-interferometer type wavemeter, which has a precision of $10^{-3}$ cm$^{-1}$, is used to determine $\omega_1$. The Ti:Sapphire laser is passively stabilized with a thin-thick etalon combination and has a short term line width on the order of a few MHz. Long-term drifts, on a time scale longer than a typical data run of a few minutes, are dominated mainly by thermal and mechanical noise, and have a negligible influence on the experimental results. The average power is approximately 200 mW. The laser 1 output is strongly linearly polarized, which is further purified by passing the beam through a Glan-Thompson prism polarizer. The beam is then passed through an electronically controlled liquid crystal retardation wave plate (LCR), which switches the linear polarization direction to one of two orthogonal linear polarization directions. The resulting variable polarization beam is then directed to the Cs sample cell. The second excitation step is driven by the linearly polarized output from an external cavity diode laser (ECDL, laser 2), which generates an average power of 8 mW in a short-term ($\sim 1$ s) bandwidth $\sim 1$ MHz. The laser has a frequency $\omega_2$ which nominally satisfies the two-photon resonance condition $\omega_1 + \omega_2 = \omega_0$, where $\omega_0$ is the frequency separation of hyperfine components of the $6s^2S_{1/2}$ and $8s^2S_{1/2}$ levels. The degeneracy-weighted hyperfine averaged value of $\omega_0 = 24317.17$ cm$^{-1}$ \[36\]. The laser 2 frequency is determined from the detuning according to $\omega_2 = \omega_0 - \omega_{3/2} - \Delta$. When $\Delta = 0$, the excited state resonance frequency is $\omega_2 = 12584.82$ cm$^{-1}$ \[37\]. The commercial ECDL is piezoelectrically scannable over a range of approximately 15 GHz around the two-photon resonance. The diode laser output beam is made to be nearly collinear with that from the Ti:Sapphire laser and the two beams are weakly focused with 0.5 m focal length lenses and overlapped in the central region of a sample cell assembly.

The Cs vapor cell is an evacuated and sealed Pyrex cell having a length of about 7.5 cm and a diameter of...
monitored by a computerized data acquisition and in-
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background light and the intense laser beams by a combi-
fluence light on the cathode of a red-light sensitive
power supply.

2.5 cm. Research grade windows are fused directly onto
the cell body, leaving about a 2.0 cm diameter undis-
torted central region for transmission of the laser beams.
The cell is prepared on an oil-free vacuum system, and
is evacuated to a base pressure of about 10^{-5} mbar. A
small amount of Cs metal is driven into the cell prior to
removing it from the vacuum system. The cell tempera-
ture is varied by placing it in a resistively heated oven,
which allows heating to a typical temperature of 375 K,
corresponding to a Cs vapor density of about 2.6 \times 10^{11}
cm^{-3}. The cell temperature is stabilized to about \pm 0.1
K using a thermocouple attached to the coolest part of
the cell; the thermocouple output is fed back to the oven
power supply.

Two-photon resonance signals are monitored by mea-
surement of the 7p \,^2P_{j} \rightarrow \,^6s \,^2S_{1/2} cascade fluorescence at 455 nm and 459 nm. The fluorescence is collected at
right angles to the laser beams by a short focal length
field lens (\sim 5 cm), which approximately collimates the
fluorescence light on the cathode of a red-light sensitive
photomultiplier tube (PMT). The tube is protected from
background light and the intense laser beams by a combina-
tion of colored glass and narrow band interference fil-
ters. Infrared transmitting colored glass filters mounted
on the entrances to the oven housing further reduce back-
ground light signals. The PMT output is amplified and
the photon counting rate measured with a commercial
100 MHz photon counter. Typical counting rates on two-
photon resonance are \sim 10^7 s^{-1}. We point out that the
operating temperature of 375 K turns out to be optimal for
this experiment. At lower temperatures the fluores-
cence signals are weak, while at elevated temperatures,
and correspondingly larger Cs atom density, the vapor
becomes optically thick to the 7p \,^2P_{j} \rightarrow \,^6s \,^2S_{1/2} signal
radiation. Then branching to the 7s \,^2S_{1/2} and 6d
^2D_{j} levels (see Figure 1) is enhanced, and signals in the
observed decay channel are correspondingly reduced.

The various instruments are globally controlled or
monitored by a computerized data acquisition and in-
strument control program. To illustrate the experimen-
tal protocol, consider a typical experimental run, where
laser 1 is set to a nominal frequency, which is measured
by the wavemeter, which passes this value to the main
control program. Sequentially, the photon-counting rate
is measured alternately for collinear and perpendicular
linear polarization directions of the two lasers. The
rettardance of the LCR is controlled with a specially writ-
ten instrument driver that communicates with the LCR
hardware via the computer parallel port. The frequency
of the ECDL (laser 2) is then shifted by piezoelectrically
scanning the ECDL cavity length; this is achieved by di-
rect communication between the ECDL controller and
the main experiment computer. The wavemeter reading
is recorded again and the data cycle is repeated. This ex-
perimental protocol is very effective for determining the
shape of the excitation line, and for assessing the blend-
ing of different hyperfine components of the excitation
line shape. An alternate data-taking protocol was also
employed. In this, once a proper setting of laser 2 was
determined which minimized the influence of hyperfine
blending (see discussion section) on the measured polar-
ization, the laser 2 frequency was not scanned. Data was
then accumulated by switching the laser polarization un-
til sufficient statistics were obtained.

For each data run, the experimental signal is deter-
mained for two different states of relative linear polariza-
tion of the excitation lasers. Although the absolute intens-
ities of these components depend on many experimental
factors, including the laser intensities, Cs density, and the
sensitivity of the detection electronics, the intensity ratio
is sensitive only to the relative polarization state of the
lasers and the two-photon matrix elements. The main
experimental observable for detailed analysis is then the
linear polarization degree defined as

\[ P_L = \frac{S_\| - S_\perp}{S_\| + S_\perp}, \]

where \( S_\| \) and \( S_\perp \) are the measured signal intensities
when the laser beams are linearly polarized collinearly
and perpendicularly, respectively.

The nuclear spin of \(^{133}\text{Cs}\) is \( I = 7/2 \), and so both the
6s \,^2S_{1/2} and 8s \,^2S_{1/2} electronic states have hyperfine
components of total angular momentum \( F = 3, 4 \). The
hyperfine splitting in the ground 6s \,^2S_{1/2} level is the
International Frequency Standard, which is approxi-
ately 9.192 GHz, while the hyperfine splitting in the 8s
^2S_{1/2} level is about one-tenth this value and is on the
order of 0.9 GHz. Each of these splittings is larger than
the one-photon Doppler width of several hundred MHz
associated with the separate 6s \,^2S_{1/2} \rightarrow \,^6p \,^2P_{j} and 6p \,^2P_{j}
\rightarrow \,8s \,^2S_{1/2} transitions, and so partial resolution of the
hyperfine splitting is expected even when the two exci-
tation laser beams are copropagating through the sample
cell. This is illustrated in Figure 3, which shows the
spectral and relative polarization dependence of the
two-photon excitation rate at \( \Delta = -80.37 \text{ cm}^{-1} \). The
main contribution to the spectral width is due to Doppler
Copropagating beams

\[ \Delta = -80.37 \text{ cm}^{-1} \]

**FIG. 3:** Experimental signal for frequency and polarization-dependent two-photon excitation at a detuning of \(-80.37 \text{ cm}^{-1}\) from resonance. All four hyperfine transitions are shown, are notated by \((F, F')\), and are labeled by the vertical lines above each component. The laser 2 offset is measured from a convenient starting point, denoted by the origin of the graph. Co-propagating beams.

broadening, which is approximately double that for a one-photon transition. On the other hand, for counterpropagating beams, nearly complete cancellation of the Doppler width is expected, with a residual two-photon Doppler width \(\sim 10 \text{ MHz}\). A typical such scan is shown in Figure 4, which corresponds to the \(F = 4 \rightarrow F' = 4\) transition with \(\Delta = -22.0 \text{ cm}^{-1}\). There it is seen that the spectral width of the line shape is approximately 40 MHz, which corresponds well to the combined residual Doppler width and the spectral width of the Ti:Sapphire laser. It is also seen that the excitation spectrum depends significantly on the relative polarization state of the two laser beams, giving in this case a large linear polarization degree of approximately 0.78.

In spite of the evidently good signal to noise ratio for the data in Figure 4, it proved difficult to achieve the desired reproducibility in the polarization measurements with the passively stabilized Ti:Sapphire laser. The main reason for this was that measurement of the two different polarization states were made sequentially, and fluctuations in the laser frequency on the switching time scale introduced unwanted noise in the extracted polarization values. The reason for this is that when the lasers are counterpropagating through the interaction region of the cell, the Doppler width of the two-photon transition is greatly reduced, and is on the order of the natural width of the final 8s level. In this case, in order to obtain reliable polarization measurements, the combined frequency drift of the two lasers would need to be smaller than a fraction of one MHz/s. Instead, we recorded the excitation spectrum for copropagating laser beams, for which the Doppler width is large (several hundred MHz), and for which the characteristic laser frequency drift rates of a few MHz/s are entirely acceptable. Note that variation of the polarization in the few MHz range is totally negligible. The small penalty to be paid for this is that the polarization values need to be determined in the wings of the partially blended lines. However, these determinations were made sufficiently far into the wings that the other hyperfine transition made nearly negligible contribution to the polarization value. In addition, the signals were significantly less noisy than with counterpropagating beams, making possible consistent and repeatable measurements of the polarization at each detuning. A typical higher resolution data run is presented in Figure 5, which corresponds to the \(F = 4 \rightarrow F' = 3, 4\) transitions at \(\Delta = -167.9 \text{ cm}^{-1}\).

Finally, we have modeled the residual effect of blending of the hyperfine lines on the measured polarization val-

**FIG. 4:** Experimental signal for frequency and polarization-dependent two-photon excitation at a detuning of \(-22.00 \text{ cm}^{-1}\) from resonance. The \(F = 4 \rightarrow F' = 4\) transition is shown. The laser 2 offset is measured from a convenient starting point, denoted by the origin of the graph. Counterpropagating laser beams.

**FIG. 5:** Experimental signal for frequency and polarization-dependent two-photon excitation at a detuning of \(-167.9 \text{ cm}^{-1}\) from resonance. The \(F = 3 \rightarrow F' = 4\) and \(F = 3 \rightarrow F' = 3\) transitions are shown. The laser 2 offset is measured from a convenient starting point, denoted by the origin of the graph. Copropagating laser beams.
ues. In the model, we approximate each hyperfine transition line shape to be Gaussian and having the measured width determined by the Doppler broadening of the two-photon transition. Using the known relative hyperfine transition probabilities gives a polarization-dependent correction that ranges from 0.0005 to 0.0013 (polarization values reported here can range from -1 to +1). Even though these corrections are well within the statistical error associated with each measured polarization, the corrections are systematic and so are made directly to the measured counting rates, prior to extracting $P_L$ values from the data through Eq. (1).

A. Analysis and Experimental Results

The variation of the linear polarization degree with detuning $\Delta$ of laser 1, and for each of the four electric-dipole allowed hyperfine transitions, may be readily calculated in terms of transition matrix elements (the general expression may be found in [32]). The dominant intermediate levels for the two-photon transitions are the 6p $^2P_j$ levels, with $j = 1/2$, 3/2, but other np $^2P_j$ transitions, including the p-continuum, also contribute to the total transition probability, and play an important role in the experiments reported here. For the transitions when $\Delta F = \pm 1$, absorption of two photons with collinear polarization directions is forbidden, and so the linear polarization degree is -1, independent of detuning. However, for the other transition pair, where $\Delta F = 0$, both fine structure multiplet components contribute, leading to strong spectral variations in the linear polarization degree. Theoretical expressions for the intensities are given by

$$I_\| = 4I_{o33} \left[ \frac{R}{\omega_1 - \omega_{3/2}} + \frac{1}{\omega_1 - \omega_{1/2}} + \frac{R}{\omega_2 - \omega_{3/2}} + \frac{1}{\omega_2 - \omega_{1/2}} + P \right]^2, \quad (2)$$

$$I_\| = I_{o33} \left[ \frac{R/2}{\omega_1 - \omega_{3/2}} - \frac{1}{\omega_1 - \omega_{1/2}} - \frac{R/2}{\omega_2 - \omega_{3/2}} + \frac{1}{\omega_2 - \omega_{1/2}} + Q \right]^2, \quad (3)$$

for the $F = 3 \rightarrow F' = 3$ transition and

$$I_\| = \frac{36}{15} I_{o44} \left[ \frac{R}{\omega_1 - \omega_{3/2}} + \frac{1}{\omega_1 - \omega_{1/2}} + \frac{R}{\omega_2 - \omega_{3/2}} + \frac{1}{\omega_2 - \omega_{1/2}} + P \right]^2, \quad (4)$$

$$I_\| = I_{o44} \left[ \frac{R/2}{\omega_1 - \omega_{3/2}} - \frac{1}{\omega_1 - \omega_{1/2}} - \frac{R/2}{\omega_2 - \omega_{3/2}} + \frac{1}{\omega_2 - \omega_{1/2}} + Q \right]^2, \quad (5)$$

for the $F = 4 \rightarrow F' = 4$ transition. In these expressions, the overall intensity for each hyperfine transition is proportional to $I_{o,F,F'}$, while $R$, $P$ and $Q$ are parameters that are proportional to ratios of reduced transition matrix elements [32]. In the present case,

$$R = \frac{<8s\parallel d\parallel 6p_{3/2}> <6p_{3/2}\parallel d\parallel 6s>}{<8s\parallel d\parallel 6p_{1/2}> <6p_{1/2}\parallel d\parallel 6s>}, \quad (6)$$

where $d$ is the electric-dipole operator. The quantities $P$ and $Q$ are given by

$$P = \sum_{n>jk} \frac{M_{njk}}{\omega_{nk} - \omega_{np_j}} \quad (7)$$

$$Q = \sum_{n>jk} \frac{(-1)^{j+1/2} M_{njk}}{j + 1/2 \omega_{nk} - \omega_{np_j}}, \quad (8)$$

where the matrix element ratio is given by

$$M_{mjk} = \frac{<8s\parallel d\parallel np_j><np_j\parallel d\parallel 6s>}{<8s\parallel d\parallel 6p_{1/2}> <6p_{1/2}\parallel d\parallel 6s>}. \quad (9)$$

The total angular momentum $j = 1/2, 3/2$, while $k = 1, 2$ labels the frequency of the absorbed photons.

An illustration of the variation of the polarization with detuning $\Delta$ is shown in Figure 6 for each of the four hyperfine transitions. In these plots we have taken $R = 2$, corresponding to no relativistic modification of the reduced transition dipole matrix elements, and $P = Q = 0$, which applies to the case when only the $n = 6$ intermediate levels are considered. Among the critical features of the plots are the resonance values of the linear polarization degree for each hyperfine transition. In the present

![Figure 6](image-url)
FIG. 7: Measured linear polarization degree of the two-photon excitation rate as a function of detuning for the F = 3 → F’ = 3 and F = 4 → F’ = 4 transitions. The solid curves represent the model curves for a matrix element ratio R = 2, and neglecting far-off-resonance transitions (P=Q=0).

P_{L} = 0.882 for the F = 4 → F’ = 4 transition, and P_{L} = 0.811 for the F = 3 → F’ = 3 transition. In addition, there is a detuning from resonance where the polarization is P_{L} = -1.0, corresponding to the cases when the intensity S_{0} = 0 for each hyperfine transition. The location of this point generally depends on the four main terms in Eq. (2) - (8), and on the values for P and Q.

Polarization measurements have been made in an approximately 200 cm^{-1} range of detunings to the low frequency side of the atomic 6s^{2}S_{1/2} → 6p^{2}P_{j} resonance transition. These measurements, which have a typical one standard-deviation uncertainty of 0.002, are presented in Figure 7. The solid curves in this figure are expanded versions of those in Figure 6. Note that the error bars on the experimental data points are negligible on the scale of the figure. It can be seen from the curves that there is significant discrepancy between the measurements and the theoretical curves (with P = 0, Q = 0, and R = 2) for the F = 4 → F’ = 4 transition, for which we have taken the most extensive data. At the largest detuning, this amounts to about 20 standard deviations. The departure has two main contributions, the being the fact that nonresonant transitions with n > 6 make a significant contribution through nonzero values of P and Q, and because the value of R generally departs from the nonrelativistic value of R = 2. In previous work, the three parameters P, Q and R were considered as constants, which is quite well justified by their weak dependence on detuning. Fits to the detuning-dependent polarization data then yielded linear relationships among the parameters; estimates of P and Q further allowed extraction of a quite precise value for the constant matrix element ratio R. Although this is found to be insufficient for the precision of the data presented here, we report here our values, making the same assumptions as in earlier work. That approach yields R=2.1068(36) + 443.2(1.2)P - 362(362)Q, where the uncertainty (in parenthesis) in the numerical values represents one standard deviation. Note that although the coefficient of Q has an uncertainty on the order of its value, its value is correlated with that of R and P, and so it cannot be neglected. The final uncertainty in R derived from calculated values of P and Q shows that the correction due to Q is on the order of the uncertainty in R. This equation represents the equivalent sum rule to those presented in earlier reports on our measurements in Rb.

However, P and Q depend weakly on detuning, and so to obtain the highest precision, it is desirable to calculate P and Q as a function of detuning directly from the most accurate experimental and theoretical matrix elements. We use a combination of the most reliable experimental and theoretical values for dominant contributions with n ≤ 9. For n = 6, measured resonance line (n = 6) matrix elements of Rafac, et al. [12] are employed, while for the second resonance doublet (n = 7), we use the recently reported precision measurements of Vasilyev et al. [13]. Other values having n ≤ 9 are obtained from the relativistic all-order calculations of Safronova et al. [14]; these are listed in Table 1. For multiplets with n > 9, which make only a small overall contribution to R, we use Dirac-Hartree-Fock matrix elements [28]; these values are also summarized in Table 1, along with the energies associated with each intermediate np level. For reference, it is found that P and Q, calculated with these values, are fit very well by second order polynomials in detuning ∆ = ω_{1} - ω_{10}. They are given by P = -2.437 x 10^{-4} + 3.257 x 10^{-9}∆ - 3.8847 x 10^{-12}∆^{2} and Q = 3.260 x 10^{-6} - 7.683 x 10^{-9}∆ + 1.645 x 10^{-13}∆^{2}. The detuning ∆ is in vacuum cm^{-1} units. We estimate the total uncertainty in P and Q to be on the order of 1%.

To obtain the matrix element ratio defined in Eq. (6), a nonlinear least squares fit of the polarization data of Fig. 7 is made to the theoretical expressions, using the calculated values of P and Q for each detuning. The result of the fit is R = 2.0024(24), where the dominant error in R comes from the statistical uncertainty in the measured polarization. The quality of the fit is illustrated in Fig. 8. In the figure, measurements are compared to the deviation of the polarization from the values obtained when R = 2, P = Q = 0; these are represented by the horizontal line passing through zero deviation. The experimental measurements, represented by the data points, are seen to be in excellent agreement with the solid curve, this being calculated from the fitting parameters.

The ratio of excited-state dipole matrix elements R_{6s-6p} = <8s|d|6p3/2>/<8s|d|6p1/2> may be obtained from Eq. (6) by combining the measured value of R with the high precision 6p resonance line matrix element ratio of 1.4074(3) measured by Rafac et al. [11]. This gives the excited state matrix element ratio of R_{6s-6p} = 1.423(2), which has the uncertainty of about 0.15%, with roughly equal contributions from statistical uncertainty in the measurements reported here and from
the resonance line matrix element ratio of [11]. Finally, we point out that the analysis may be reversed, if it is assumed that the bare transition matrix element ratio is calculated precisely. Then, by recognizing that the off-resonance terms represented by $P$ and $Q$ contribute an average of about -0.1091(37) to the value of $R$, the measurements determine the much smaller combination of dipole matrix elements in Eqs. (7) - (8) to about 3%. As these terms are dominated by transitions through the 6s - 7p and 6s - 8p multiplets, the measurements serve as a consistency check on existing measurements [13] at that level.

III. RELATIVISTIC ALL-ORDER CALCULATION OF TRANSITION MATRIX ELEMENTS

In order to compare the experimental result $R_{8s-6p} = 1.423(2)$ with the high-precision theoretical value we carry out relativistic all-order calculation of electric-dipole matrix elements in Cs. In particular, we calculate $8s - np$, $n = 6, 7, 8, 9$ electric-dipole reduced matrix elements using a relativistic all-order method including single and double (SD) excitations [28]. The resulting values of the $8s - 7p$, $8s - 8p$, and $8s - 9p$ electric-dipole matrix elements were used to determine $P$ and $Q$.

In the SD all-order method, the wave function of the valence electron $v$ is represented as an expansion

$$|\Psi_v\rangle = \left[1 + \sum_{ma} \rho_{ma} a_{m}^\dagger a_{a} + \frac{1}{2} \sum_{mnab} \rho_{mnab} a_{m}^\dagger a_{n}^\dagger a_{b} a_{a} + \sum_{m \neq v} \rho_{mv} a_{m}^\dagger a_{v} + \sum_{mna} \rho_{mnva} a_{m}^\dagger a_{n}^\dagger a_{a} a_{v}\right]|\Phi_v\rangle$$

(10)

In this equation $|\Phi_v\rangle$ is the lowest-order atomic state wave function, which is taken to be the frozen-core Dirac-Hartree-Fock (DHF) wave function of a state $v$ and $a_v^\dagger$ and $a_v$ are creation and annihilation operators, respectively. The indices $a, b$ designate core electrons and indices $m, n$ designate any states above the core. The equations for excitations coefficients $\rho_{ma}, \rho_{mv}, \rho_{mnab},$ and $\rho_{mnva}$ are obtained by substituting the wave function $|\Psi_v\rangle$ into the many-body Schrödinger equation

$$H|\Psi_v\rangle = E|\Psi_v\rangle,$$

(11)

where $H$ is the relativistic no-pair Hamiltonian [29]. The equations are solved iteratively until the corresponding correlation energy for the state $v$ is sufficiently converged. The resulting excitation coefficients are then used to calculate matrix elements. The one-body matrix element of the operator $Z$ given by

$$Z_{wv} = \frac{\langle \Psi_w | Z | \Psi_v \rangle}{\langle \Psi_v | \Psi_v \rangle},$$

(12)

is expressed in terms of excitation coefficients as

$$Z_{wv} = \frac{z_{wv} + Z^{(a)} + \cdots + Z^{(t)}}{(1 + N_v)(1 + N_w)},$$

(13)

where $z_{wv}$ is the lowest-order DHF matrix element, the terms $Z^k$, $k = a...t$ are linear or quadratic functions of the excitation coefficients, and normalization terms $N_v$ and $N_w$ are quadratic functions of the excitation coefficients. As a result, certain sets of many-body perturbation theory (MBPT) terms are summed to all orders. This method is shown to yield high-accuracy results for the primary transition electric-dipole matrix elements in alkali-metal atoms [27, 28, 30]. The results for the reduced electric-dipole matrix elements for $8s - 6p, 8s - 7p, 8s - 8p,$ and $8s - 9p$ transitions are listed in Table I.

Next, we investigate the effect of the correlation to the $8s - 6p$ matrix elements to evaluate the uncertainty of their ratio. We find that the total correlation correction to the $8s - 6p$ matrix elements is small, 3%. However, it actually results from severe cancellations of large contributions illustrated in Table I where we give the breakdown of the all-order SD calculation for both $8s - 6p$ transitions. The lowest-order (DHF) value is listed in the first row of Table II. Three larger terms, $Z^{(a)}, Z^{(c)},$ and $Z^{(d)}$

$$Z^{(a)} = \sum_{ma} z_{am} \tilde{\rho}_{wva} + \sum_{ma} z_{ma} \tilde{\rho}_{wma}$$

$$Z^{(c)} = \sum_{m} z_{wm} \rho_{mv} + \sum_{m} z_{mv} \rho_{mw}^*$$

$$Z^{(d)} = \sum_{mn} z_{mn} \tilde{\rho}_{mva} \rho_{nv},$$

(14)

where

$$\tilde{\rho}_{wva} = \rho_{wva} - \rho_{wma}$$

(15)
obtained from Dirac-Hartree-Fock calculations.

TABLE I: Energy and matrix elements (ME) used in calculating the contributions of far-off-resonance transitions to the measured linear polarization spectrum. ME for \( n \geq 10 \) are obtained from Dirac-Hartree-Fock calculations.

| Relay Level Energy (cm\(^{-1}\)) | 6s-npj (a.u.) | 8s-npj (a.u.) |
|----------------------------------|---------------|---------------|
| 6p1/2                            | 11178.2       | 4.489\(^a\)   | -1.027\(^d\) |
| 6p3/2                            | 11732.4       | 6.324\(^a\)   | -1.462\(^d\) |
| 7p1/2                            | 21732.4       | 0.276\(^b\)   | -9.251\(^d\) |
| 7p3/2                            | 21765.7       | 0.586\(^b\)   | -14.00\(^d\) |
| 8p1/2                            | 25791.8       | 0.218\(^c\)   | 24.46\(^d\)  |
| 8p3/2                            | 25763.7       | 0.043\(^c\)   | 1.743\(^d\)  |
| 9p1/2                            | 27802.0       | 0.127\(^c\)   | 2.969\(^d\)  |
| 10p1/2                           | 28727.1       | 0.047          | 0.634        |
| 10p3/2                           | 28753.9       | 0.114          | 1.158        |
| 11p1/2                           | 29403.7       | 0.034          | 0.348        |
| 11p3/2                           | 29421.1       | 0.085          | 0.667        |
| 12p1/2                           | 29852.9       | 0.026          | 0.228        |
| 12p3/2                           | 29864.7       | 0.067          | 0.451        |
| 13p1/2                           | 30166.0       | 0.021          | 0.165        |
| 13p3/2                           | 30174.5       | 0.055          | 0.334        |
| 14p1/2                           | 30393.2       | 0.017          | 0.127        |
| 14p3/2                           | 30399.5       | 0.046          | 0.262        |
| 15p1/2                           | 30563.3       | 0.015          | 0.102        |
| 15p3/2                           | 30568.0       | 0.039          | 0.213        |

\(^a\)Rafac, et al. \(^{[\text{12}]}\)
\(^b\)Vasilyev, et al. \(^{[\text{15}]}\)
\(^c\)Safronova, et al. \(^{[\text{30}]}\)
\(^d\)This work (all order)

We note that while terms \( Z(\text{a}) \) and \( Z(\text{c}) \) contain third-order terms as well as higher-order terms (see \(^{[\text{27}]}\) for details) the \( Z(\text{d}) \) term, being quadratic in valence single-excitation coefficients \( \rho_{\text{mv}} \) contains only terms starting from fifth order. Out of the remaining terms, the largest contribution comes from the normalization correction. For these transitions, the dominant contribution to \( N_{\text{v(w)}} \) in the denominator of Eq. \(^{[\text{13}]}\) comes from the term

\[
\sum_m \rho_{\text{mv}}^\text{a} \rho_{\text{mv}}^\text{c}.
\] (17)

This term contributes 97\% to the \( N_{\text{6s}} \) value and 64\% to the \( N_{\text{6p}} \) value. Again, this term is quadratic in single excitation coefficients \( \rho_{\text{mv}} \) and can contain only MBPT terms starting from fifth order. Thus, \( 8s - 8p \) matrix elements present an interesting case with large, but canceling, contributions from high orders in many-body perturbation theory. This cancellation occurs for both \( 8s - 6p_{1/2} \) and \( 8s - 6p_{3/2} \) matrix elements.

To evaluate the uncertainty in the theoretical ratio \( R_{\text{8s-6p}} \) we also calculate \( 8s - 6p_{3/2} \) and \( 8s - 6p_{1/2} \) electric-dipole matrix elements in different approximations and estimate some omitted contributions. The results are summarized in Table II. The lowest-order DHF values are listed in column labeled DHF. The results of the third-order many-body perturbation-theory calculation, which includes higher-order random-phase-approximation terms as described in \(^{[\text{31}]}\), are listed in column “III”. The third-order values, which include an estimate of the omitted fourth- and higher-order Brueckner-orbital (see classification and formulas in \(^{[\text{31}]}\)) corrections obtained by the scaling procedure described in \(^{[\text{31}]}\), are listed in column “III\_sc”. Single-double all-order data from Table I are listed in column “SD”. The results obtained by including partial contribution of the triple excitations are listed in column labeled “SDpT”. These data are obtained by adding a triple-excitation valence term to Eq. \(^{[\text{10}]}\) and making corresponding corrections to correlation energy and single-valence excitation coefficient equations \( \rho_{\text{mv}} \) as described in Refs. \(^{[\text{23}, \text{24}, \text{30}]}\). The \( \rho_{\text{mv}} \) excitation coefficients give rise to the largest terms, \( Z(\text{c}) \) and \( Z(\text{d}) \), and the dominant part of \( N_{\text{v}} \).

All-order SD and SDpT calculations include a complete third-order contribution (see \(^{[\text{27}]}\) for a detailed comparison) but omit some classes of higher-order terms starting from fourth order. We have estimated some omitted correlation corrections resulting from triple and higher excitations using the scaling described, for example, in Ref. \(^{[\text{24}]}\). Briefly, single-particle excitation coefficients \( \rho_{\text{mv}} \) are multiplied by the ratio of the experimental and theoretical correlation energies. The modified excitation coefficients are then used to re-calculate matrix elements. Such scaling estimates only certain classes of the omitted contributions (mainly Brueckner-orbital contributions, see Ref. \(^{[\text{24}]}\) for classification of the perturbation theory terms). The corresponding SD and SDpT scaled values are listed in columns “SD\_sc” and “SDpT\_sc”.

The third-order matrix elements do not differ signifi-
TABLE III: Electric-dipole 8s–6p, reduced matrix elements in Cs calculated using different approximations: Dirac-Hartree-Fock (DHF), third-order many-body perturbation theory (III), single-double all-order method (SD), single-double all-order method including partial triple contributions (SDpT) and the corresponding scaled values. $R_{8s-6p}$ is the ratio of the 8s–6p$_{3/2}$ and 8s–6p$_{1/2}$ matrix elements. Absolute values of the matrix elements in a.u. are given.

| Level | Ab initio | Scaled |
|-------|-----------|--------|
|       | DHF       | III SD | SDpT | III sc | SDsc | SDpTsc |
| j=1/2 | 1.0584    | 1.0231 | 1.0260 | 1.0321 | 1.0315 | 1.0223 | 1.0327 |
| j=3/2 | 1.5145    | 1.4519 | 1.4618 | 1.4709 | 1.4712 | 1.4556 | 1.4705 |
| $R_{8s-6p}$ | 1.4309    | 1.4191 | 1.4247 | 1.4252 | 1.4262 | 1.4238 | 1.4240 |

significantly from the all-order results indicating very accurate cancellation between large higher-order terms. The Brueckner-orbital correction is dominant in a third-order calculation for both transitions and is relatively more important for the 8s–6p$_{3/2}$ transition, thus giving a ratio differing from more accurate SD all-order value by 0.6%. We note that the ratio of the scaled third-order values, which includes an estimate of higher-order Brueckner-orbital terms, agrees with the all-order result. As one can see from Table III the total correlation correction contribution to the ratio $R_{8s-6p}$ is very small, 0.4%, owing to the cancellation of dominant terms. The difference between all-order data in different approximations is only 0.1%. As the largest contributions to these matrix elements contain valence single-excitation coefficients $\rho_{mv}$ the inclusion of the partial triple contributions and the scaling described above should give a good estimate of the omitted higher-order terms as both these methods are aimed at correcting $\rho_{mv}$. We recommend the SD $ab$ initio value $R_{8s-6p} = 1.425(2)$ as the final theoretical value for the ratio. The uncertainty is obtained by combining the uncertainty of the dominant terms, determined to be 0.1% based on the spread of SD, SDpT and scaled values, and the total uncertainty in all other, much smaller, contributions taken to be 0.1% not to exceed the uncertainty of the dominant terms.

IV. DISCUSSION OF RESULTS AND CONCLUSIONS

Comparison of the theoretical matrix element ratio of $R = 1.425(2)$, and the corresponding experimental result of $R = 1.423(2)$ shows excellent agreement. This is remarkable, given the various contributing theoretical and experimental factors that significantly affect the final result in each case. It is interesting to reiterate that many of the two-photon transition matrix elements in heavy alkali atoms show significant relativistic modification. For example, the 5s $^2S_{1/2} \rightarrow 5p ~^2P_j \rightarrow 5d ~^2D_{3/2}$ transition in Rb shows such variations at a level of nearly 7%. At the same time, other ratios, such as that measured in the present case, show significantly smaller relativistic modification, and yet in each case such modifications are generally well-described by the highest-level atomic structure calculations. Nevertheless, there is a significant sensitivity in the experimental measurements to other dipole transitions, and these transitions depend significantly on relativistic contributions. Naturally, it would be of interest to measure the absolute oscillator strengths of the individual multiplet transitions, in order to make direct comparison of the matrix element values reported here. Such measurements remain a significant challenge to experimental technique. Finally, the excited state transitions in atomic Cs associated with the 7p doublet are of particular interest, as those second resonance line matrix elements show nearly a factor of 2 departure from the expected line strength ratio of 2.

In conclusion, we have used precision two-photon polarization spectroscopy to make measurements of the transition matrix element ratio associated with the 6s $^2S_{1/2} \rightarrow np ~^2P_j \rightarrow 8s ~^2S_{1/2}$ transition in atomic Cs. The measurements are combined with other experimental data and calculations in order to extract the ratio of excited-state matrix elements. The experimental value is found to be in excellent agreement with the ratio of reduced matrix elements calculated using a relativistic all-order method.

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