

$5p^2P_j \rightarrow 5d^2D_{3/2}$ transition matrix elements in atomic ^{87}Rb

S. B. Bayram,* M. Havey,† M. Rosu, and A. Sieradzan

Department of Physics, Central Michigan University, Mt. Pleasant, Michigan 48859

A. Derevianko‡ and W. R. Johnson

Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556

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A combined precision experimental and theoretical study of $5p^2P_j \rightarrow 5d^2D_{3/2}$ electric-dipole transition matrix elements in atomic ^{87}Rb has shown that they are dominated by electron correlation. The relative size and sign of the measured matrix element ratio is found to be 1.068(8), in very good agreement with the value of 1.135 obtained from relativistic third-order many-body perturbation theory.

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Determination of multipole matrix elements, quantities that define a scale of interaction between electromagnetic radiation and atomic matter [1], remains a fundamental problem of considerable theoretical and experimental interest. Such quantities are important to a wide range of basic and applied problems, including determination of astrophysical abundances of atoms in stellar atmospheres [2] and analysis of atomic parity-violation experiments [3]. As opposed to observables related to energy intervals in atoms or molecules, which may be determined to a precision of $\sim 10^{-11}$ [4,5], the best measurements of atomic dipole matrix elements have only recently achieved a precision of $\sim 5 \times 10^{-4}$ [6]. And despite remarkable advances in the development of relativistic many-body techniques in atomic physics, calculations of electric-dipole matrix elements for heavier atoms such as cesium have been limited to an accuracy of about 0.5% [7,9,10]. Nevertheless, at this level insight into important physical effects determining the matrix elements may be given. For example, a recent report of calculation of magnetic dipole matrix elements associated with $nS^2S_{1/2} \rightarrow n'S^2S_{1/2}$ transitions in the alkali-metal atoms has revealed that negative-energy states can produce significant anomalies in the magnetic transition strength [11].

A number of novel techniques have recently been applied to precise absolute and relative measurements of the strength of resonance transitions in the heavier alkali-metal atoms. These have included refined lifetime measurements in atomic-beam experiments [12], photoassociative spectroscopy of cold atoms confined to atomic traps [13–15], and direct measurements of the natural width of the $3s^2S_{1/2} \rightarrow 3p^2P_{3/2}$ transition in Na [16]. Meticulous light-absorption measurements have determined the relative strength of the Cs resonance transitions [17] to $\sim 5 \times 10^{-4}$. It has also been shown [18] that the frequency dependence of polarized Rayleigh and Raman scattering of light from atoms depends sensitively on the relative strengths of radiative transitions be-

tween manifolds of atomic levels. Further, recent experiments [19] on the $5s^2S_{1/2} \rightarrow 5p^2P_j \rightarrow 8s^2S_{1/2}$ transition in atomic Rb demonstrated that precision measurements of the polarization dependence of the two-photon transition rate in atoms can yield sum rules for combinations of atomic electric-dipole matrix elements.

We have made a combined experimental and theoretical study of the $5s^2S_{1/2} \rightarrow 5p^2P_j \rightarrow 5d^2D_{3/2}$ transition in atomic ^{87}Rb . In this case, precision measurements of the nonresonant two-photon, two-color linear depolarization spectrum show remarkable polarization-dependent interference structure which is partly due to the near degeneracy of the two photons driving the process. From the measurements it was possible to extract the ratio of the reduced excited-state transition matrix elements for two contributing pathways: $5p^2P_{3/2} \rightarrow 5d^2D_{3/2}$ and $5p^2P_{1/2} \rightarrow 5d^2D_{3/2}$. The ratio of reduced matrix elements for the multiplet transition shows a significant departure from the nonrelativistic limit of unity. To understand the results, we have performed third-order many-body perturbation theory [7] calculations of the matrix elements, including spin-orbit and relativistic effects in an *ab initio* fashion. The calculations reveal that, although the main departure appears in the Dirac-Hartree-Fock values, exceptionally large correlation contributions dominate the excited-state transition matrix elements.

The experimental scheme is illustrated in the inset to Fig. 1, which shows low-lying energy levels for atomic Rb. Two independently tunable lasers having frequencies ω_1 and ω_2 are adjusted to satisfy the two-photon resonance condition $\omega_1 + \omega_2 = \omega_0$, where ω_0 is the resonance frequency of the $5s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition. Averaged over hyperfine structure, $\omega_0 = 25\,700.56 \text{ cm}^{-1}$. In the weak laser fields of the present experiment, there are two contributing virtual levels; these are indicated by the horizontal broken lines in Fig. 1. In the present case, these merge when one laser frequency is offset from the $5s^2S_{1/2} \rightarrow 5p^2P_{3/2}$ transition by about 33.7 cm^{-1} . In this paper detuning is defined as $\Delta_1 = \omega_1 - \omega_{3/2}$, where $\omega_{3/2} = 12\,816.58 \text{ cm}^{-1}$ is the resonance frequency of the $5s^2S_{1/2} \rightarrow 5p^2P_{3/2}$ transition. For the $5s^2S_{1/2} \rightarrow 5p^2P_{1/2}$ transition, $\omega_{1/2} = 12\,578.96 \text{ cm}^{-1}$. Detection of atoms promoted to the $5d^2D_{3/2}$ levels is achieved by monitoring the $6p^2P_j \rightarrow 5s^2S_{1/2}$ fluorescence around 420 nm.

*Present address: University of Michigan, Ann Arbor, MI.

†Permanent address: Old Dominion University, Norfolk, VA.

‡Present address: Harvard Smithsonian Center for Astrophysics, Cambridge, MA.

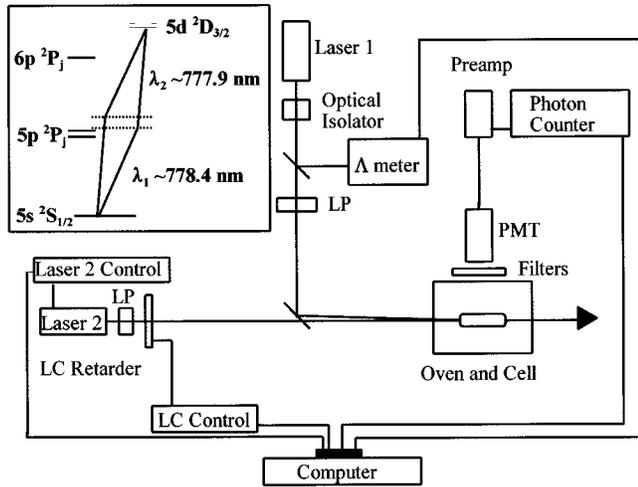


FIG. 1. Schematic diagram of the excitation scheme and the experimental apparatus.

A diagram of the experimental apparatus is also shown in Fig. 1. In the figure, laser 1 is a temperature-tuned diode laser delivering around 40 mW in a bandwidth on the order of 1 MHz, but having a combined jitter and drift of about 50 MHz in one minute. Feedback to the laser was prevented by a Faraday-type optical isolator that provided more than 35 dB of isolation over the spectral range of importance to the present experiment. In order to monitor ω_1 , about 5% of the output of the laser was directed to a wavemeter having a precision of 30 MHz; calibration of the instrument against the atomic resonance lines established the accuracy to about 100 MHz. The linear polarization of the laser-1 output was purified by a Glan-Thompson prism having an extinction ratio of about 10^{-5} . Laser 2 was an external-cavity tuned diode laser operated in a single longitudinal mode. The laser provided 6–9 mW of power in a bandwidth typically stable to less than 10 MHz over the several minutes required to make a scan over the two-photon resonance. The laser was piezoelectrically tuned over the two-photon resonances. The linear polarization of the laser was purified and made parallel to that of laser 1 by means of a polarizing beam splitter having an extinction ratio of greater than 10^{-3} . The linear polarization direction of laser 2 was controlled to be either collinear with, or perpendicular to that of laser 1 by means of a liquid crystal retarder. The overall analyzing power of the resulting polarimeter was $A > 0.999$.

The two beams were made to pass nearly collinearly (< 0.01 rad) through a heated oven-cell arrangement. The oven provided a temperature stability of ± 0.1 K over the range of 320–420 K used in the experiment. This corresponded to a Rb density range $\sim 10^{11}$ cm^{-3} to 10^{13} cm^{-3} . The cylindrical Pyrex cell had flat and uncoated windows and contained isotopically purified ^{87}Rb . The signal fluorescence at 420 and 422 nm was collected at right angles to the laser beams by a short focal length lens ($f \sim 3.8$ cm) and spectrally separated from scattered laser light and other fluorescence channels by a colored-glass filter. The fluorescence was detected by a blue-sensitive photomultiplier tube operating in a photon-counting mode. The signals were amplified

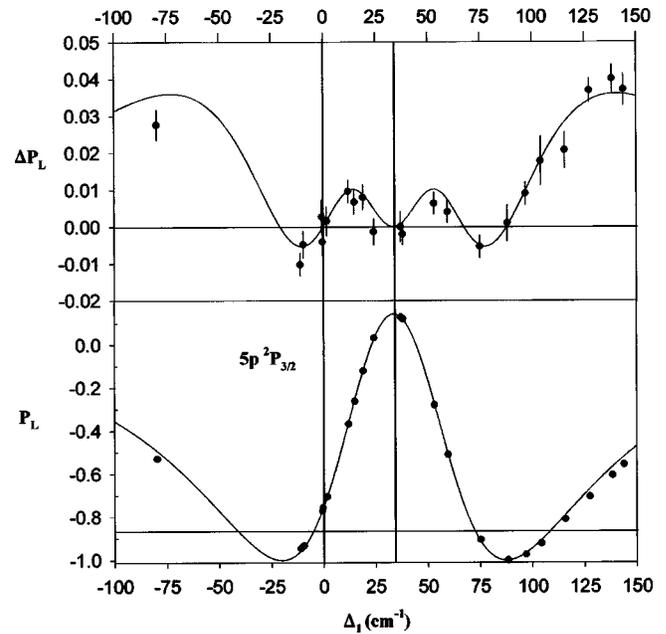


FIG. 2. Measured P_L and fitted ΔP_L polarization spectrum for the $5p\ ^2P_j - 5d\ ^2D_{3/2}$ ($j=1/2, 3/2$) transition in Rb.

with a fast preamplifier, and accumulated by a 100-MHz photon counter–discriminator system. The typical counting rate was about 10^4 s^{-1} , making dead-time corrections negligible. Laser scanning, polarization switching, and acquisition and storage of signals were managed by a LabVIEW based computer-controlled data acquisition system [20].

The integrated and background-corrected signal when the two lasers have collinear polarization directions is S_{\parallel} , and when they are perpendicular is S_{\perp} . These may be combined to form a linear polarization degree P_L ,

$$P_L = \frac{S_{\parallel} - S_{\perp}}{S_{\parallel} + S_{\perp}}.$$

The polarization degree, determined as a function of the detuning Δ_1 , is the main quantity measured in the experiment and compared with theoretical expressions. Polarization measurements were made over a Δ_1 range of about 200 cm^{-1} , and for excitation out of both $F=1$ and $F=2$ ground-level hyperfine components. The P_L values formed from S_{\parallel} and from S_{\perp} were free of measurable systematic variations due to the intensity of either laser 1 or laser 2. Measured P_L values for excitation out of either the $F=1$ or the $F=2$ ground-state hyperfine levels were also independent of the Rb density. Final polarization measurements have an average statistical uncertainty of about $\pm 0.4\%$.

The measured linear polarization spectrum associated with the $5s\ ^2S_{1/2} \rightarrow 5p\ ^2P_j \rightarrow 5d\ ^2D_{3/2}$ transition is shown in the lower half of Fig. 2. First note that the spectrum is, within the statistics of the measurements, independent of the initial ground-state hyperfine level (not shown in Fig. 2). This is due to the fact that the $5d\ ^2D_{3/2}$ level decays predominantly to the $6p\ ^2P_{1/2}$ level, which can support no alignment. Thus the radiation distribution cannot depend on alignment transfer, and only the considerably weaker $6p\ ^2P_{3/2}$

$\rightarrow 5s^2S_{1/2}$ would show any F dependence. Estimates of the size of the effect are consistent with the null experimental result. The polarization spectrum shows distinctive interference structure that is symmetric about a detuning of 33.7 cm^{-1} from the $5s^2S_{1/2} \rightarrow 5p^2P_{3/2}$ transition. The structure has two origins. First, at this detuning the two-photon resonance condition is satisfied when laser 1 and laser 2 have the same frequency. For detunings on either side of this, there are comparable contributions to the excitation amplitude from either temporal order of photon absorption. Second, excitation of the $5d^2D_{3/2}$ level has contributing amplitudes (in principle) from all the np levels; in the present case the $5p^2P_{3/2}$ and $5p^2P_{1/2}$ levels dominate. Even for the largest detuning of about 150 cm^{-1} , the other np levels contribute at most a fraction of about 10^{-3} of these levels.

Neglecting hyperfine structure, expressions for the frequency-dependent intensities for the $5s^2S_{1/2} \rightarrow 5p^2P_j \rightarrow 5d^2D_{3/2}$ transition are given by

$$I_{\parallel} = \left[\frac{5}{\omega_1 - \omega_{1/2}} + \frac{5}{\omega_2 - \omega_{1/2}} + \frac{R}{\omega_1 - \omega_{3/2}} + \frac{R}{\omega_2 - \omega_{3/2}} + p \right]^2,$$

$$I_{\perp} = \frac{3}{4} \left[\frac{-5}{\omega_1 - \omega_{1/2}} + \frac{2R}{\omega_1 - \omega_{3/2}} - \frac{3R}{\omega_2 - \omega_{3/2}} + q_1 \right]^2$$

$$+ \frac{25}{4} \left[\frac{1}{\omega_1 - \omega_{1/2}} + \frac{2}{\omega_2 - \omega_{1/2}} + \frac{4R/5}{\omega_1 - \omega_{3/2}} - \frac{R/5}{\omega_2 - \omega_{3/2}} + q_2 \right]^2.$$

In these expressions p , q_1 , and q_2 represent contributions of np multiplet levels other than the $5p^2P_j$ levels. These quantities are weakly frequency dependent, and may be estimated from existing atomic data [21] to have the approximate values of $p \cong 10^{-4}$, $q_1 \cong -6 \times 10^{-5}$, and $q_2 \cong 3.6 \times 10^{-5}$; they have a negligible effect on the polarization determined from the above equations over the frequency range of the data. The quantity $R = R_1(5s_{1/2} - 5p_j)R_2(5p_j - 5d_{3/2})$ is a product of ratios of reduced dipole matrix elements [22,8] for the two-photon transition. It represents the ratio of transition matrix elements for the $j \rightarrow j'$ multiplet transition normalized by evaluating associated angular-momentum recoupling coefficients. In the absence of relativistic influence on the reduced matrix elements, $R = 1$.

We have used these expressions to calculate the linear polarization spectrum with $R = 1$, and with the p , q_1 , and q_2 values indicated; the result is shown as the solid line in the lower half of Fig. 2. From the figure it is evident that the experimental data depart significantly from this prediction, particularly at larger detunings. This may be seen more clearly in the upper part of Fig. 2, which shows the deviation of the polarization from the curve as a function of detuning. We have made a least-squares fit of the data, using R as an adjustable parameter. The solid curve shown in the upper portion of Fig. 2 minimizes the deviations and is obtained for $R = 1.067(7)$, representing a strong departure from the nonrelativistic limit of $R = 1$. Note that the sign of R is also determined from the measurements to be positive. The

TABLE I. Breakdown of many-body contributions to selected electric-dipole matrix elements (in a_0) in Rb.

	$5d_{5/2} - 5p_{3/2}$	$5d_{3/2} - 5p_{3/2}$	$5d_{3/2} - 5p_{1/2}$
DHF	0.3182	0.3033	0.2115
RPA	0.0848	0.0855	0.0874
BO	0.6325	0.6447	0.6129
SR+Norm	-0.0117	-0.0119	-0.0118
Total	1.0238	1.0216	0.9000

value may be further reduced by using recent precision lifetime measurements of the Rb resonance transitions [12], which give $R_1(5s_{1/2} - 5p_j) = 0.999(1)$ for the matrix element ratio. Thus we obtain the excited-state transition ratio of matrix elements as $R_2(5p_j - 5d_{3/2}) = 1.068(8)$. Although the sign of $R_1(5s_{1/2} - 5p_j)$ is not determined from the lifetime measurements, it is unlikely that the reduced matrix elements change sign within the $5p$ resonance line doublet. Thus the ratio $R_2(5p_j - 5d_{3/2})$ should also be positive.

We have employed relativistic third-order many-body perturbation theory (MBPT), described in detail in Ref. [7], to calculate the $5p_j - 5d_j$ transition matrix elements. The calculations start from ‘‘frozen core’’ Dirac-Hartree-Fock (DHF) orbitals and include spin-orbit and other relativistic effects in an *ab initio* manner. To make relativistic effects apparent, we present in Table I a breakdown of the various many-body contributions to the $5p_j - 5d_j$ reduced matrix elements, divided by corresponding values of reduced C_1 matrix elements, where C_1 is the normalized spherical harmonic [8]. In the present case, retardation effects are dominated by the many-body contributions.

From Table I, we note that correlation corrections constitute more than 70% of the total value, in contrast to the principal $5s_{1/2} - 5p_j$ transition, where correlations contribute at only a 5% level [9]. The dominant effect is due to Bruckner orbital (BO) corrections, corresponding to a reaction of the valence electron to the induced core polarization. The BO contributions were calculated using the second-order self-energy operator and include both third- and fourth-order corrections. The random-phase approximation (RPA) corrections, describing shielding of the applied field and usually, equally important, are also significantly lower. Structural radiation (SR) and normalization (Norm) diagrams are contributing at an even lower level. We note that matrix elements connecting the $5p^2P_{3/2}$ level with the $5d^2D_{3/2}$ level and with the $5d^2D_{5/2}$ level are close in value, demonstrating that d states are minimally affected by the relativistic core region due to the centripetal barrier. The penetrating p states are effected by spin-orbit interaction much more strongly; the ratio of $5p_{3/2} - 5d_{3/2}$ and $5p_{1/2} - 5d_{3/2}$ reduced matrix elements in the lowest (DHF) order is 1.435. The core-polarization (BO) correction, being a long-range effect, differs for these transitions at only a 4% level and together with other many-body contributions brings the matrix element ratio to $R_2(5p_j - 5d_{3/2}) = 1.135$, in very good agreement with the experimental value. We point out that third-order MBPT estimates for matrix elements of the principal $5p_{1/2} - 5s_{1/2}$ transition are known to agree with experiment at a 1% level.

Due to the much larger correlation contribution to $5p_j - 5d_j$ amplitudes, we expect the present third-order result to be less accurate. Even better precision could be achieved by calculations in the framework of relativistic all-order methods [9]. Finally, note that due to the large correlations, the results calculated in the length and velocity form agree at a 10% level. However, the corresponding ratio R_2 differs by only $\sim 2\%$.

The points to be emphasized are the following. First, precise linear polarization spectra have been determined for the $5s^2S_{1/2} \rightarrow 5p^2P_j \rightarrow 5d^2D_{3/2}$ multiplet transitions in ^{87}Rb . The spectra show marked interference structure associated with both orders of photon absorption, and with contributions from the two $5p$ multiplet components to the transition amplitude. The high quality of the measurements, which have a fractional uncertainty of $\sim 8 \times 10^{-3}$, reveals a significant departure in the transition matrix element ratio from the nonrelativistic case. The associated intensity ratios would be modified by about 15% in this case. Second, third-order rela-

tivistic many-body calculations of the matrix elements allow extraction of the important physical contributions. The resulting calculations are in very good agreement with the measurements, permitting interpretation of the effect as due to modification of Dirac-Hartree-Fock values by the differential effect of core polarization in the $5p$ multiplet components; the DHF values and the resulting electron correlation contribution to the reduced transition matrix elements account for nearly all the departure from the nonrelativistic value. Finally, as the d state does not strongly penetrate the core, and the main effect is on the $5p$ wave functions, transitions involving one penetrating state and a relatively nonpenetrating state may serve as better indicators of relativistic effects than transitions between two penetrating orbitals.

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- [1] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Applications* (Wiley, New York, 1992).
- [2] D. Mihalas, *Stellar Atmospheres* (Freeman, San Francisco, 1970).
- [3] C. S. Wood, S. C. Bennett, D. Cho, B. P. Masterson, J. L. Roberts, C. E. Tanner, and C. E. Wieman, *Science* **275**, 1759 (1997).
- [4] P. Shao, W. Lichten, H. Layes, and J. Bergquist, *Phys. Rev. Lett.* **58**, 1293 (1987).
- [5] T. W. Hansch, in *Atomic Physics 14*, edited by D. Wineland, C. E. Wieman, S. J. Smith, AIP Conf. Proc. No. **323** (AIP, New York, 1995).
- [6] W. I. McAlexander, E. R. I. Abraham, and R. G. Hulet, *Phys. Rev. A* **54**, R5 (1996).
- [7] W. R. Johnson, Z. W. Liu, and J. Sapirstein, *At. Data Nucl. Data Tables* **64**, 280 (1996).
- [8] D. A. Varshalovich, A. N. Moskalev and V. K. Khersonskii, *Quantum Theory of Angular Momentum* (World Scientific, Singapore, 1988).
- [9] See, for example, M. S. Safronova, W. R. Johnson, and A. Derevianko, *Phys. Rev. A* **60**, 996 (1999), and references therein.
- [10] V. A. Dzuba, V. V. Flambaum, and O. P. Sushkov, *Phys. Rev. A* **51**, 3454 (1995).
- [11] I. M. Savukov, A. Derevianko, H. G. Berry, and W. R. Johnson, *Phys. Rev. Lett.* **83**, 2914 (1999).
- [12] U. Volz, M. Jajerus, H. Liebel, A. Schmitt, and H. Schmoranzler, *Phys. Rev. Lett.* **76**, 2862 (1996); U. Volz and H. Schmoranzler, *Phys. Scr.* **T65**, 48 (1996); C. E. Tanner, A. E. Livingston, R. J. Rafac, F. G. Serpa, K. W. Kukla, H. G. Berry, L. Young, and C. A. Kurtz, *Phys. Rev. Lett.* **69**, 2765 (1992).
- [13] C. R. Ekstrom, J. Schmiedmayer, M. S. Chapman, T. D. Hammond, and D. E. Pritchard, *Phys. Rev. A* **51**, 3883 (1995).
- [14] K. M. Jones, P. S. Julienne, P. D. Lett, W. D. Phillips, E. Tiesinga, and C. J. Williams, *Europhys. Lett.* **35**, 85 (1996).
- [15] H. Wang, J. Li, X. T. Wang, C. J. Williams, P. L. Gould, and W. C. Stwalley, *Phys. Rev. A* **55**, R1569 (1997).
- [16] C. W. Oates, K. R. Vogel, and J. L. Hall, *Phys. Rev. Lett.* **76**, 2866 (1996).
- [17] R. J. Rafac and C. E. Tanner, *Phys. Rev. A* **58**, 1087 (1998).
- [18] M. D. Havey, *Phys. Lett. A* **240**, 219 (1998).
- [19] A. I. Beger, M. D. Havey, and R. P. Meyer, *Phys. Rev. A* **55**, 3780 (1997).
- [20] Development by ScinetVI (1999).
- [21] A. A. Radzig and B. M. Smirnov, *Reference Data on Atoms, Molecules and Ions* (Springer, Berlin, 1985).
- [22] M. E. Rose, *Elementary Theory of Angular Momentum* (Wiley, New York, 1957).