

# Determination of electric-dipole matrix elements in K and Rb from Stark shift measurements

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Stark shifts of potassium and rubidium  $D1$  lines have been measured with high precision by Miller *et al.* [Phys. Rev. A **49**, 5128 (1994)]. In this work, we combine these measurements with our all-order calculations to determine the values of the electric-dipole matrix elements for the  $4p_j-3d_j$  transitions in K and the  $5p_j-4d_j$  transitions in Rb to high precision. The  $4p_{1/2}-3d_{3/2}$  and  $5p_{1/2}-4d_{3/2}$  transitions contribute on the order of 90% to the respective polarizabilities of the  $np_{1/2}$  states in K and Rb, and the remaining 10% can be accurately calculated using the relativistic all-order method. Therefore, the combination of the experimental data and theoretical calculations allows us to determine the  $np-(n-1)d$  matrix elements and their uncertainties. We compare these values with our all-order calculations of the  $np-(n-1)d$  matrix elements in K and Rb for a benchmark test of the accuracy of the all-order method for transitions involving  $nd$  states. Such matrix elements are of special interest for many applications, such as determination of “magic” wavelengths in alkali-metal atoms for state-insensitive cooling and trapping, and determination of blackbody radiation shifts in optical frequency standards with ions.

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The values of the various electric-dipole matrix elements in alkali-metal atoms are needed for a variety of applications ranging from reducing decoherence in quantum logic gates [1] to the study of fundamental symmetries [2,3]. The all-order method that includes all single and double (SD) excitations of the Dirac-Fock (DF) wave function to all orders of perturbation theory was shown to give values for the primary  $np$ - $ns$  transitions in alkali-metal atoms in excellent agreement with high-precision experiments [4]. There are many interesting applications involving  $np$ - $n'd$  transitions in alkali-metal atoms and other monovalent systems, but there are very few benchmark experiments, such as lifetime measurements, to compare with theoretical calculations. Moreover, the only high-precision lifetime measurements of the lowest  $nd$  states of any alkali-metal atom [5,6], carried out for Cs, are in disagreement [7] with the Stark shift values for cesium  $D1$  and  $D2$  lines [8,9]. The breakdown of the correlation correction terms is very different for the  $np$ - $ns$  and  $np$ - $n'd$  transitions, creating a need for additional benchmark tests. In this work, we determine the values of the  $4p_j-3d_j$  transitions in K and  $5p_j-4d_j$  transitions in Rb to high precision using experimental values of the Stark shifts of  $D1$  lines [10] in these systems.

The motivation for this work is twofold. First, we provide the recommended values for these transitions to be used for various applications, such as determination of the “magic” wavelengths in alkali-metal atoms for state-insensitive cooling and trapping and calculation of the  $nd$  state polarizabilities. Second, we also conduct all-order calculations of these transitions in order to carry out a benchmark comparison of the accuracy of the all-order method. The conclusions reached in this work allow us to provide recommended values for a variety of the transition properties of monovalent systems and more accurately evaluate their uncertainties. Such transition properties are needed for the evaluation of

the blackbody radiation and quadrupole shifts in ions, light shifts and quadrupole polarizabilities in  $Ba^+$  which were recently measured in Refs. [11,12], branching ratios for various decay channels, and other applications. Such benchmarks are also useful for the understanding of the accuracy of the all-order calculations conducted for the analysis of the experimental studies of parity violation with heavy atoms and search for an electron electric-dipole moment.

The  $D1$  line Stark shifts in alkali-metal atoms were measured with high precision by Miller *et al.* [10] and Hunter *et al.* [8,13] using a pair of cavity-stabilized diode lasers locked to resonance signals. The K and Rb measurements, 39.400(5) and 61.153 kHz/(kV/cm)<sup>-2</sup>, respectively, represent a three order of magnitude improvement in accuracy upon previous experimental results. In this paper, we determine the  $4p_{1/2}-3d_{3/2}$  and  $5p_{1/2}-4d_{3/2}$  electric-dipole ( $E1$ ) matrix elements in K and Rb, respectively, using these experimental Stark shifts. We also compare these values with our all-order calculations of the  $np-(n-1)d$  matrix elements in K and Rb for a benchmark test of the accuracy of the all-order method for transitions involving  $nd$  states. The values of the  $4p_{3/2}-3d_j$  and  $5p_{3/2}-4d_j$  electric-dipole matrix elements in K and Rb, respectively, are obtained by combining our recommended values for the  $4p_{1/2}-3d_{3/2}$  and  $5p_{1/2}-4d_{3/2}$  transitions with the corresponding accurate theoretical ratios.

We start by expressing the experimental Stark shifts as the difference of the ground  $ns$  and the first excited  $np_{1/2}$  state polarizabilities of the respective atoms [10]. It is convenient for this purpose to use the system of atomic units (a.u.), in which  $e$ ,  $m_e$ ,  $4\pi\epsilon_0$ , and the reduced Planck constant  $\hbar$  have the numerical values 1. Polarizability in a.u. has the dimensions of volume, and its numerical values presented here are thus measured in units of  $a_0^3$ , where  $a_0 \approx 0.052\,918$  nm is the Bohr radius. The atomic units for  $\alpha$  can be converted to

SI units via  $\alpha/h[\text{Hz}/(\text{V}/\text{m})^2]=2.488\,32\times 10^{-8}\alpha$  (a.u.), where the conversion coefficient is  $4\pi\epsilon_0 a_0^3/h$  and the Planck constant  $h$  is factored out.

The Stark shifts in the  $D1$  lines of K and Rb yield the following values of the differences of the scalar dipole polarizabilities of the  $np_{1/2}$  and  $ns$  states [10]:

$$\Delta_{\text{K}} = \alpha_0(4p_{1/2}) - \alpha_0(4s) = 317.11(4)a_0^3, \quad (1)$$

$$\Delta_{\text{Rb}} = \alpha_0(5p_{1/2}) - \alpha_0(5s) = 492.20(7)a_0^3. \quad (2)$$

The static  $E1$  atomic polarizabilities of the alkali-metal atoms are dominated by the valence contribution  $\alpha_v$  for all valence states. The small ionic core contribution evaluated in Ref. [14] using the random-phase approximation approach does not affect the present calculation as it is the same for  $\alpha_0(np_{1/2})$  and  $\alpha_0(ns)$  and does not contribute to the Stark shifts. The counterterm  $\alpha_{vc}$  that needs to be introduced to correct ionic core polarizability for an occupancy of the valence shell is very small for the  $ns$  state and is entirely negligible for the  $np$  state. The valence polarizabilities are calculated using the sum-over-states approach:

$$\alpha_v = \frac{2}{3(2j_v + 1)} \sum_k \frac{\langle k||D||v \rangle^2}{E_k - E_v}, \quad (3)$$

where  $\langle k||D||v \rangle$  is the reduced electric-dipole matrix element for the transition between states  $k$  and  $v$ , and  $E_i$  is the energy corresponding to the level  $i$ . The sum over the intermediate states  $k$  converges very rapidly. In fact, the first two  $ns$ - $np_{1/2}$  and  $ns$ - $np_{3/2}$  transitions entirely dominate the ground state polarizabilities. Since these two matrix elements can be obtained from the corresponding experimental lifetimes [15], our values for the ground state polarizabilities contain very little theoretical input. Such a calculation has been described before [4,16–18], and we do not repeat the details here. Our resulting values are  $\alpha_0(4s)=289.6(6)a_0^3$  and  $\alpha_0(5s)=318.35(62)a_0^3$ . These results are in agreement with the values of Refs. [4,16]. The uncertainty comes nearly entirely from the uncertainties of the experimental values of the  $np$  lifetimes.

The polarizabilities of the  $np_{1/2}$  states are dominated by a single transition,  $np_{1/2}-(n-1)d_{3/2}$ , allowing us to use Eqs. (1) and (2) to derive the matrix elements that are the subject of the present work. As a result, Eqs. (1) and (2) can be rewritten as

$$\begin{aligned} \Delta_{\text{K}} &= \alpha_0(4p_{1/2}) - \alpha_0(4s) \\ &= \frac{1}{3} \frac{\langle 3d_{3/2}||D||4p_{1/2} \rangle^2}{E_{3d_{3/2}} - E_{4p_{1/2}}} + \alpha_0^{\text{other}}(4p_{1/2}) - 289.6(6) \\ &= 317.11(4), \end{aligned} \quad (4)$$

$$\begin{aligned} \Delta_{\text{Rb}} &= \alpha_0(5p_{1/2}) - \alpha_0(5s) \\ &= \frac{1}{3} \frac{\langle 4d_{3/2}||D||5p_{1/2} \rangle^2}{E_{4d_{3/2}} - E_{5p_{1/2}}} + \alpha_0^{\text{other}}(5p_{1/2}) - 318.35(62) \\ &= 492.20(7), \end{aligned} \quad (5)$$

where we substituted the ground state polarizability values

and separated the contribution from the  $np_{1/2}-(n-1)d_{3/2}$  transition. However, the remaining contributions to the  $np_{1/2}$  polarizabilities grouped together as  $\alpha_0^{\text{other}}(np_{1/2})$  still give 10% to the polarizabilities of the  $np_{1/2}$  states and need to be evaluated accurately for our approach to yield high-precision values. We describe this calculation below. For completeness, we describe the full theoretical evaluation of the  $np_{1/2}$  polarizabilities first and then remove the dominant contribution to determine  $\alpha_0^{\text{other}}(np_{1/2})$ .

We separate the valence polarizabilities into two parts,  $\alpha^{\text{main}}$ , containing the contributions from the states near the valence state, and the remainder,  $\alpha^{\text{tail}}$ . We calculate the matrix elements contributing to the main term using the SD all-order method. We conduct additional semi-empirical scaling of our all-order values ( $\text{SD}_{\text{sc}}$ ), where we expect scaled values to be more accurate based on the analysis of the dominant correlation correction contributions. We refer the reader to Refs. [2,4,19,20] for the description of the all-order method and the scaling procedure. The experimental energies from [21,22] are used in all main term contributions. The remaining terms from highly excited one-electron states are included in the  $\alpha^{\text{tail}}$  part. The  $\alpha^{\text{tail}}$  is calculated in the DF approximation using complete basis set functions that are linear combinations of  $B$  splines [23]. We use 70 splines of order 11 for each angular momentum state. A spherical cavity radius of 220 a.u. is chosen to accommodate all valence orbitals included in the calculation of  $\alpha^{\text{main}}$ . We chose to include as many states as possible into  $\alpha^{\text{main}}$  in order to decrease the uncertainty of the remainder term.

The contributions to the scalar polarizabilities of the  $4p_{1/2}$  state in K and  $5p_{1/2}$  state in Rb and their uncertainties are listed in Tables I and II, respectively. The corresponding experimental energy differences [21,22,24,25] and the absolute values of the lowest-order (DF) and final all-order electric-dipole reduced matrix elements are also listed. The lowest-order values are listed in order to illustrate the size of the correlation correction for all transitions. We use the experimental numbers from Ref. [15] along with their uncertainties for the primary  $ns$ - $np_{1/2}$  transitions (for example, the  $5s$ - $5p_{1/2}$  transition in Rb). High-precision relativistic SD or scaled SD all-order values are used for all the remaining main term transitions. The uncertainties given for the matrix elements are equal to the differences between the SD *ab initio* and scaled values.

The tail contribution is rather small for the  $ns$  sum, but is significant for the  $nd_{3/2}$  sum. In order to evaluate the uncertainties in the tail contributions, we calculated a few last main terms by using the DF approximation and compared the resulting values with the all-order values quoted in Tables I and II. In the case of the  $ns$  states, the DF values differ from our all-order values by only 7–10%, with the difference decreasing with increasing principal quantum number  $n$ . As a result, we assign a 10% uncertainty to the  $ns$  tail values. In the case of the  $nd_{3/2}$  states, the correlation corrections are extremely large and nearly cancel the lowest-order contribution for K. The differences between the all-order and DF values are on the order of 90% for K and 60% for Rb. As we mentioned above, the large  $nd_{3/2}$  tail contributions are the reason for the inclusion of so many states into the main term.

TABLE I. The contributions to the scalar polarizability of the  $4p_{1/2}$  state in K and their uncertainties. The corresponding energy differences [21,22,24,25], the absolute values of the lowest-order (DF) electric-dipole reduced matrix elements, and final all-order matrix elements are also listed. The energy differences are given in  $\text{cm}^{-1}$ . Electric-dipole matrix elements are given in atomic units ( $ea_0$ ), and polarizabilities are given in  $a_0^3$ , where  $a_0$  is the Bohr radius.

Contribution	$k$	$\langle 4p_{1/2}    D    k \rangle^{\text{DF}}$	$\langle 4p_{1/2}    D    k \rangle^{\text{SD}}$	$E_k - E_{4p_{1/2}}$	$\alpha_0(4p_{1/2})$
$\alpha^{\text{main}}(ns)$	4s	4.555	4.102	-12 985	-94.8(2)
	5s	3.974	3.885	8041	137.3(1.4)
	6s	0.925	0.903	14 466	4.127(3)
	7s	0.485	0.477	17 289	0.962
	8s	0.319	0.315	18 780	0.386
	9s	0.233	0.231	19 663	0.198
	10s	0.181	0.180	20 229	0.117
$\alpha^{\text{tail}}(ns)$					1.6(0.2)
$\alpha^{\text{main}}(nd_{3/2})$	3d <sub>3/2</sub>	8.596	7.949	8552	540.5(9.7)
	4d <sub>3/2</sub>	0.769	0.097	14 413	0.05(5)
	5d <sub>3/2</sub>	0.105	0.336	17 201	0.48(47)
	6d <sub>3/2</sub>	0.030	0.340	18 711	0.45(30)
	7d <sub>3/2</sub>	0.063	0.296	19 613	0.33(18)
	8d <sub>3/2</sub>	0.069	0.253	20 193	0.23(11)
	9d <sub>3/2</sub>	0.067	0.216	20 587	0.17(8)
$\alpha^{\text{tail}}(nd_{3/2})$					4.5(4.5)
$\alpha^{\text{core}}$					5.5(3)
Total					602(11)

TABLE II. The contributions to the scalar polarizability of the  $5p_{1/2}$  state in Rb and their uncertainties. The corresponding energy differences [21,22,24,25], the absolute values of the lowest-order (DF) electric-dipole reduced matrix elements, and final all-order matrix elements are also listed. The energy differences are given in  $\text{cm}^{-1}$ . Electric-dipole matrix elements are given in atomic units ( $ea_0$ ), and polarizabilities are given in  $a_0^3$ , where  $a_0$  is the Bohr radius.

Contribution	$k$	$\langle 5p_{1/2}    D    k \rangle^{\text{DF}}$	$\langle 5p_{1/2}    D    k \rangle^{\text{SD}}$	$E_k - E_{5p_{1/2}}$	$\alpha_0(5p_{1/2})$
$\alpha^{\text{main}}(ns)$	5s	4.819	4.231	-12 579	-104.11(15)
	6s	4.256	4.146	7554	166.5(2.2)
	7s	0.981	0.953	13 733	4.835(16)
	8s	0.514	0.502	16 468	1.120(7)
	9s	0.338	0.331	17 920	0.448(3)
	10s	0.247	0.243	18 783	0.230(2)
	11s	0.192	0.189	19 338	0.135(1)
$\alpha^{\text{tail}}(ns)$					1.9(0.2)
$\alpha^{\text{main}}(nd_{3/2})$	4d <sub>3/2</sub>	9.046	8.017	6777	694(30)
	5d <sub>3/2</sub>	0.244	1.352	13 122	10.2(9)
	6d <sub>3/2</sub>	0.512	1.067	16 108	5.2(1.1)
	7d <sub>3/2</sub>	0.447	0.787	17 701	2.6(4)
	8d <sub>3/2</sub>	0.369	0.605	18 643	1.4(2)
	9d <sub>3/2</sub>	0.307	0.483	19 243	0.89(10)
$\alpha^{\text{tail}}(nd_{3/2})$					10.5(10.5)
$\alpha^{\text{core}}$					9.08(45)
Total					805(31)

We take the uncertainty in the  $nd_{3/2}$  tail contributions to be 100%.

While such an estimate represents a high bound on the value of the Rb tail based on the comparison of the DF and all-order values, the case of K requires some additional consideration owing to larger discrepancies of the DF and all-order values even for  $n=9$ . We have conducted additional all-order calculations of the K  $4p_{1/2}-nd_{3/2}$   $E1$  matrix elements with  $n>9$ . We found that our calculational scheme becomes impractical for  $n>15$ . Such a problem is expected because the lowest-order energies of the higher basis set states are large and positive, resulting in cancellations in the denominators of the many body perturbation theory (MBPT) terms and consequently leading to the divergence of the all-order iteration procedure. In our approach, the tail does not sufficiently converge at  $n=15$  to significantly reduce the tail error. As a result, we chose a different approach to ensure that we do not significantly underestimate the tail in the K calculation. We compared our final results for K and Rb with experimental values for the  $np_{1/2}$  state polarizabilities by combining the  $D1$  line Stark shifts from [10] with recommended ground state polarizability values from [16]. Our K and Rb values differ from the experimental results 606.7(6) and 810.6(6) a.u. by 0.69% and 0.77%, respectively. If the tail in the K calculation were significantly underestimated, we should have seen a significant mismatch of the K and Rb comparison since the tail problem is specific to the K calculation.

Tables I and II show that the uncertainties of the  $nd_{3/2}$  tail values give the overwhelmingly dominant contributions to the uncertainties of  $\alpha_0^{\text{other}}(np_{1/2})$ .

Subtracting the contribution of the  $np_{1/2}-(n-1)d_{3/2}$  states from our final theoretical values for  $np_{1/2}$  polarizabilities and removing the corresponding uncertainties from the total error budget, we obtain

$$\alpha_0^{\text{other}}(4p_{1/2}) = 61.6(4.8)a_0^3,$$

$$\alpha_0^{\text{other}}(5p_{1/2}) = 111(11)a_0^3.$$

Substituting the  $\alpha_0^{\text{other}}$  values and experimental energies from [24] into Eq. (4), we obtain the following absolute values of the  $E1$  matrix elements:

$$\text{K: } \langle 4p_{1/2} || D || 3d_{3/2} \rangle = 7.984(35)ea_0, \quad (6)$$

$$\text{Rb: } \langle 5p_{1/2} || D || 4d_{3/2} \rangle = 8.051(63)ea_0. \quad (7)$$

The uncertainties of these values are essentially determined by the uncertainties of the respective  $nd_{3/2}$  tail contributions for both K and Rb. The contributions of the uncertainties in the Stark shift measurements and ground state polarizabilities to the uncertainty of the recommended matrix element values given by Eqs. (6) and (7) are negligible.

We compare these final recommended results with our *ab initio* and scaled values in Table III. Since the contributions of the triple excitation are important for the accurate evaluation of these matrix elements, we also conducted another all-order calculation partially including the triple excitations

TABLE III. Comparison of the recommended values for the  $np_{1/2}-(n-1)d_{3/2}$  electric-dipole reduced matrix element in Rb and K derived from Stark shifts in this work (listed in row labeled “Present work”), with our theoretical calculations carried out using single-double all-order method (SD), single-double all-order method including partial triple excitations (SDpT), and their scaled values. Absolute values are given. Units:  $ea_0$ .

	K( $4p_{1/2}-3d_{3/2}$ )	Rb( $5p_{1/2}-4d_{3/2}$ )
Present work	7.984(35)	8.051(63)
SD	7.868	7.846
SD <sub>sc</sub>	7.949	8.017
SDpT	7.956	7.994
SDpT <sub>sc</sub>	7.953	8.015
Final theory	7.949(80)	8.02(17)

to the extent described in Ref. [4]. We refer to these results as SDpT values in Table III and text below. The corresponding scaled values are listed in the row labeled “SDpT<sub>sc</sub>”. We take the SDpT<sub>sc</sub> values as our final values (see, for example, Refs. [20,26] for the discussion of this issue). We note that SD<sub>sc</sub> and *ab initio* SDpT values are essentially the same in the case of K and very close in the case of Rb. The uncertainty of the final values is taken to be the maximum difference between the final values and SD, SDpT, and SDpT<sub>sc</sub> ones. Our all-order values are in excellent agreement with the values derived from the  $D1$  line Stark shift. We also conclude that our procedure for the uncertainty evaluation of the theoretical matrix element values for the  $np-(n-1)d$  transitions overestimates the uncertainty, especially for Rb.

We also evaluated the recommended values of the  $4p_{3/2}-3d_j$   $E1$  matrix elements in K and  $5p_{3/2}-4d_j$   $E1$  matrix elements in Rb using our recommended values (6) and (7) and appropriate theoretical ratios. The ratios  $\langle 4p_{1/2} || D || 3d_{3/2} \rangle / \langle 4p_{3/2} || D || 3d_{3/2} \rangle$  and  $\langle 4p_{3/2} || D || 3d_{3/2} \rangle / \langle 4p_{3/2} || D || 3d_{5/2} \rangle$  in K are essentially independent of the correlation correction, i.e., the DF and all-order values are the same to better than 0.1%. Therefore, the theoretical values of the ratio are accurate enough for such a recalculation. The case of Rb is exactly the same as that of K. The complete set of our recommended values for all six  $np-(n-1)d$  transitions considered in this work is given in Table IV.

In summary, relativistic all-order calculations of the  $ns_{1/2}$  and  $np_{1/2}$  state polarizabilities are presented. The calculations are combined with the experimental Stark shift values in

TABLE IV. Recommended absolute values of the  $np-(n-1)d$  electric-dipole reduced matrix element in K and Rb. Units:  $ea_0$ .

	K	Rb
$4p_{1/2}-3d_{3/2}$	7.984(35)	$5p_{1/2}-4d_{3/2}$ 8.051(63)
$4p_{3/2}-3d_{3/2}$	3.580(16)	$5p_{3/2}-4d_{3/2}$ 3.633(28)
$4p_{3/2}-3d_{5/2}$	10.741(47)	$5p_{3/2}-4d_{5/2}$ 10.899(86)

order to determine the  $np_{1/2}-(n-1)d_{3/2}$  matrix elements in K and Rb atoms with high precision. The values of the matrix elements calculated using the experimental Stark shifts are found to be in excellent agreement with the values of the reduced matrix elements evaluated using the relativistic all-order method. This work provides a benchmark test for the

all-order matrix elements involving  $nd$  states of monovalent systems.

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