Determination of dipole polarizabilities for Mg^+ and Ca^+ ions from precision lifetime measurements and transition-moment cancellations

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(Received 26 January 1995)

It is shown that precision lifetime measurements now available for the ns-np resonance transitions in alkali-metal-like atoms and ions and the existence of strong cancellation effects in ns-$n'p$ transitions for $n'>n$ can be combined and used to specify the electric dipole polarizabilities α_d of these systems to a precision comparable to that of the lifetime measurements. The method is described and applied to Mg^+ and Ca^+ ions. Existing precision lifetime measurements are coupled with Coulomb approximation with Hartree-Slater core calculations, which estimate the contribution of the $n'>n$ transitions in near cancellation, and the analysis yields to values $\alpha_d(Mg^+)=(34.62\pm0.26)a_0^3$ and $\alpha_d(Ca^+)=(70.89\pm0.15)a_0^3$.

PACS number(s): 32.10.Dk, 32.70.Cs, 32.80.Fb

I. INTRODUCTION

The dipole polarizability of an atom or ion describes the response in lowest order in the field strength of the electron cloud to an external electric field. This atomic-structure property plays an important role in many processes [1] that motivate its theoretical and experimental determination. It is used to specify core effects in the spectroscopy of high Rydberg states [2], to account for electron correlation in model potential [3–9] and transition moment [10,4–6] calculations, in atomic scattering processes [11] and in the formulation of van der Waals forces [12], refractive indices [13], ion mobility in gases, diamagnetic susceptibilities, etc. Most experimental determinations are made directly by observation of one or more of the quantities described above. The purpose of this paper is to reinforce an earlier suggestion [14] that, as a result of advances in the field of atomic mean-lifetime measurements, it has become possible to make very accurate indirect determinations in certain systems using lifetime measurements and a knowledge of fortuitous cancellations that occur in these systems [15–18]. We have applied this approach to obtain precise determinations for Mg^+ and Ca^+ ions.

A substantial database now exists for high-precision lifetime measurements of the lowest resonance transitions in alkali-metal-like systems. Primarily through the use of selective excitation using lasers, measurements accurate to within 1% are now available for ns-np transitions in Li [19,20], Na [20,21], Mg^+ [22], Ca^+ [23,24], Cu [25], Sr^+ [26], Ag [27], Cd^+ [28], Cs [29], and Ba^+ [30]. These measurements have been found to compare very well [31] with computations made using the semiempirical method of the Coulomb approximation with Hartree-Slater core (CAHS) [7–9]. Since the Li, Na, Mg^+, Ag, Cd^+, and Cs decay processes are unbranched, the lifetime values can be converted directly to absorption oscillator strengths f. Ca^+, Cu, Sr^+, and Ba^+ have weak alternative branches to ^2D terms and can also be converted to f values if accurate theoretical estimates of these small branching fractions are available. The f values for these intrashell ns-np transitions are characteristically large and close to unity, which implies (through f sum rules in the single-active-electron approximation) that extrashell ns-$n'p$ f values are small. This tendency is especially pronounced for the singly charged ions in several of the sequences because fortuitous cancellations [15–17] affect the ns-$n'p$ transition moments for all intershell ($n'>n$) cases. The consequences of these considerations will be discussed below.

II. CALCULATIONAL FORMULATION

The ground-state dipole polarizability can be expressed in the form

$$\alpha_d(n)=4\mathscr{R}^2\left[\sum_{n'}\frac{f_{nn'}}{(E_{n'}-E_n)^2}+\int_0^\infty\frac{df/dE}{(E-E_n)^2}\,dE\right], \quad (1)$$

where $f_{nn'}$ is the absorption oscillator strength for a transition from an $ns\ ^2S_{1/2}$ ground state to an $n'p\ ^2P_{J'}$ excited state, $E_{n'}-E_n\equiv E_{nn'}$ is the corresponding excitation energy, and $\mathscr{R}$ is the Rydberg constant. The summation notation includes an implicit sum over $J'=\frac{1}{2},\frac{3}{2}$ and so does the integral over continuum states. If the sum is dominated by the $n'=n$ term, a lower limit α_{d0} for the dipole polarizability can be deduced directly from the measured lifetime of the ns-np transition through its oscillator strength and transition energy

$$\alpha_{d0}\equiv f_{nn'}(2\mathscr{R}/E_{nn'})^2. \quad (2)$$

Thus if the $n'=n$ term dominates, the uncertainties in the determination can be accurately estimated by propagating the tolerances in the lifetime measurement and combining these with theoretical estimates of the magnitudes and uncertainties in the specification of the $n'>n$ transitions. Moreover, the contributions due to higher terms in the summation could also be examined in the context of the Thomas-Reiche-Kuhn sum rule

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$$N=\sum_{n'} f_{nn'}+\int_0^\infty \frac{df}{dE}\,dE, \tag{3}$$

where N is the number of active electrons (unity in the single-particle approximation). In the present case, departure from the $N=1$ value and the single particle approximation is expected, at least in the theoretical results, since our calculations explicitly include core polarization effects in the transition matrix elements. Because n represents the ground state, no transitions involve emission and all f values are positive. Since $E_{nn'}$ increases with increasing n', the value of α_d can be bracketed

$$\alpha_{d0}\leq\alpha_d\leq\alpha_{d0}+(N-f_{nn'})(2R/E_{n,n+1})^2. \tag{4}$$

Theoretical estimates of the contributions of higher n transitions can also be made using energy level data and the semiempirical Coulomb approximation. Calculations of this type were performed using the Coulomb approximation with a central potential to represent the core (CACP) [7] as implemented recently by Theodosiou and Themelis in the calculation of the Li $2s$ and $2p$ dipole polarizabilities [32]. Specifically, the oscillator strengths to all excited $n'p,n'<21$ were explicitly calculated and the contributions of transitions from $n'=21-\infty$ were obtained by an accurate Padé extrapolation. Finally, the transitions to continuum states up to about 500 eV above threshold were calculated and added to obtain the final values.

III. APPLICATIONS

The Mg^+ ion in the Na-like sequence and the Ca^+ ion in the K-like sequence are particularly interesting cases since both have practical applications, both have been determined to modest accuracy from spectroscopic measurements, and both exhibit very strong cancellation effects.

A. Mg^+ ion

The dipole polarizability for Mg^+ was experimentally determined through its use in the identification of lines from neutral Mg in the far infrared solar spectrum. The initial observation [33,34] of two strong unidentified solar emission lines in the 120-μm region was considered important to astrophysics because the lines were narrow, had a large Zeeman sensitivity, and (unlike most magnetically sensitive visible solar lines) were chromospheric rather than photospheric. Speculations concerning the origin of these lines ended when they were identified by Chang and Noyes [35] [on the basis of theoretical calculations [36–38] for the core polarizability $\alpha_d(Mg^+)$] as the $6h$-$7i$ and $6g$-$7h$ transitions in Mg I. Reference [35] then utilized these solar data to extract an experimental value for $\alpha_d(Mg^+)$. Theoretical and experimental values for this quantity are compared in Table I.

The oscillator strengths for the $3s\ ^2S_{1/2}-3p\ ^2P_{1/2}$ and $3s\ ^2S_{1/2}-3p\ ^2P_{3/2}$ transitions have been determined with high precision in the lifetime measurements of Ansbacher *et al.* [22]. The measurements of these unbranched decays yielded values $\tau(^2P_{1/2})=3.854\pm0.030$ ns and $\tau(^2P_{3/2})=3.810\pm0.040$ ns, which are in very good agreement with earlier CAHS calculations [8] and our present CACP values of 3.929 ns and 3.898 ns, respectively. We have extended these CACP calculations up to $n'=20$, using the measured energy levels and the Ritz parametrizations of the np levels obtained by Risberg [42]. The oscillator strengths corresponding to the measurement of Ref. [22], together with our calculations of the higher transition probabilities, are given in Table II. The values labeled "Present" under "Theory" in Table I were obtained using only theoretical values for the necessary oscillator strengths.

TABLE I. Experimental and theoretical values for α_d.

Ion	Present	Other experiments	Theory	
			Present	Other
Mg^+	34.62(26)	33.0(5)[a]	34.144	33.9[b], 38.7[c] 37.2[d], 34.0[e] 33.8[f], 38.9[g]
Ca^+	70.89(15)	75.3(4)[h], 72.5(19)[i]	70.872	74.7[j], 112.4[c] 96.2[d], 76.9[e] 77.2[f]

[a]Chang and Noyes, Ref. [35].
[b]Curtis, Ref. [36].
[c]Easa and Shukla, Ref. [37].
[d]Langhoff and Hurst, Ref. [38].
[e]Adelman and Szabo, Ref. [39].
[f]Deduced from f values of Black *et al.* Ref. [40].
[g]Kundu *et al.*, Ref. [41].
[h]Chang, Ref. [46], based on data of Ref. [43].
[i]Chang, Ref. [46], based on data of Ref. [44].
[j]Curtis, Ref. [48].

Notice that the calculated oscillator strengths for $4\leq n'\leq20$ make a negligible contribution to α_d and (in the single-active-electron approximation) over 85% of the oscillator strength is contained in the $3s$-$3p$ transitions. Assuming that the uncertainties in the two lifetime measurements are uncorrelated, they introduce an uncertainty of ±0.26 (0.7%) into the determination of α_d. While Eq. (4) brackets the value to only $\pm$ 0.31 (0.9%), the calculated estimate of the contribution for $n'>3$ is only ±0.13 (0.4%).

B. Ca^+ ion

Lines in the Ca I spectrum produced by Beigang and Wynne [43] (using multiphoton laser excitation) and by Vaidyanathan *et al.* [44,45] (using microwave spectroscopy) were classified by Chang [46] using the core polarization model together with theoretical calculations [37,38,40,47,48] for $\alpha_d(Ca^+)$. Reference [46] then used these data to deduce experimental values for $\alpha_d(Ca^+)$. Theoretical and experimental values for this quantity are also compared in Table I.

The lifetimes for the $4s\ ^2S_{1/2}-4p\ ^2P_{1/2}$ and $4s\ ^2S_{1/2}-4p\ ^2P_{3/2}$ transitions in Ca^+ have been measured by Gosselin *et al.* [23] and by Jin and Church [24]. The former yielded $\tau(^2P_{1/2})=7.07\pm0.07$ ns and $\tau(^2P_{3/2})=6.87\pm0.06$ ns and the latter yielded $\tau(^2P_{1/2})=7.098\pm0.020$ ns and $\tau(^2P_{3/2})=6.924\pm0.019$ ns. These values also agree well with earlier CAHS calculations [9] and our present values of 7.105 ns and 6.909 ns, respectively. Our calculations indicate branching fractions to $3d\ ^2D_J$ levels of 6.0% for $^2P_{1/2}$ and 6.1% for $^2P_{3/2}$. In view of the accuracy of our semiempirical lifetime values, we used these branching ratios as essentially

TABLE II. Determination for Mg^+.

n'	J'	$f_{nn'}$	$E_{nn'}$	$f_{nn'}(2\mathcal{R}/E_{nn'})^2$
3	1/2	0.304(2)[a]	35 669	11.52(9)
		0.2998		11.358
	3/2	0.610(6)[a]	35 761	22.99(24)
		0.6015		22.655
4	1/2	4.365×10^{-4}	80 620	3.240×10^{-3}
	3/2	7.704×10^{-4}	80 650	5.713×10^{-3}
5	1/2	7.080×10^{-4}	97 455	3.594×10^{-3}
	3/2	1.341×10^{-3}	97 469	6.806×10^{-3}
6	1/2	4.789×10^{-4}	105 622	2.069×10^{-3}
	3/2	9.165×10^{-4}	105 630	3.958×10^{-3}
7	1/2	3.122×10^{-4}	110 204	1.238×10^{-3}
	3/2	5.998×10^{-4}	110 208	2.379×10^{-3}
8	1/2	2.100×10^{-4}	113 030	7.918×10^{-4}
	3/2	4.043×10^{-4}	113 033	1.524×10^{-3}
9	1/2	1.467×10^{-4}	114 897	5.352×10^{-4}
	3/2	2.827×10^{-4}	114 899	1.032×10^{-3}
10	1/2	1.060×10^{-4}	116 194[b]	3.783×10^{-4}
	3/2	2.046×10^{-4}	116 195[b]	7.299×10^{-4}
11	1/2	7.894×10^{-5}	117 137[b]	2.772×10^{-4}
	3/2	1.524×10^{-4}	117 137[b]	5.351×10^{-4}
12	1/2	6.027×10^{-5}	117 827[b]	2.091×10^{-4}
	3/2	1.164×10^{-4}	117 827[b]	4.039×10^{-4}
13	1/2	4.702×10^{-5}	118 367[b]	1.617×10^{-4}
	3/2	9.084×10^{-5}	118 368[b]	3.123×10^{-4}
14	1/2	3.736×10^{-5}	118 787[b]	1.275×10^{-4}
	3/2	7.220×10^{-5}	118 787[b]	2.465×10^{-4}
15	1/2	3.017×10^{-5}	119 122[b]	1.024×10^{-4}
	3/2	5.831×10^{-5}	119 122[b]	1.979×10^{-4}
16	1/2	2.470×10^{-5}	119 393[b]	8.347×10^{-5}
	3/2	4.775×10^{-5}	119 393[b]	1.614×10^{-4}
17	1/2	2.048×10^{-5}	119 616[b]	6.894×10^{-5}
	3/2	3.959×10^{-5}	119 616[b]	1.333×10^{-4}
18	1/2	1.716×10^{-5}	119 801[b]	5.759×10^{-5}
	3/2	3.318×10^{-5}	119 802[b]	1.114×10^{-4}
19	1/2	1.452×10^{-5}	119 957[b]	4.861×10^{-5}
	3/2	2.808×10^{-5}	119 957[b]	9.399×10^{-5}
20	1/2	1.240×10^{-5}	120 090[b]	4.140×10^{-5}
	3/2	2.397×10^{-5}	120 090[b]	8.007×10^{-5}
21–∞	1/2	1.142×10^{-4}		3.815×10^{-4}
	3/2	2.209×10^{-4}		7.380×10^{-4}
continuum	1/2	5.119×10^{-2}		3.127×10^{-2}
	3/2	1.014×10^{-1}		6.137×10^{-2}
	Sum	1.085		34.62(26)
	CACP	1.062		34.144

[a]Ansbacher *et al.* Ref. [22].
[b]Extrapolated by Ritz parametrizations, Ref. [42].

TABLE III. Determination for Ca^+.

n'	J'	$f_{nn'}$	$E_{nn'}$	$f_{nn'}(2\mathcal{R}/E_{nn'})^2$
4	1/2	0.3128(9)[a]	25 191.51	23.74(7)
		0.3112		23.676
	3/2	0.6296(18)[a]	25 414.40	46.95(14)
		0.6292		46.911
5	1/2	9.101×10^{-4}	60 533.02	1.199×10^{-2}
	3/2	1.358×10^{-3}	60 611.28	1.785×10^{-2}
6	1/2	1.121×10^{-3}	74 485.95	9.744×10^{-3}
	3/2	1.944×10^{-3}	74 521.80	1.688×10^{-2}
7	1/2	7.440×10^{-4}	81 498[b]	5.399×10^{-3}
	3/2	1.324×10^{-3}	81 518[b]	9.602×10^{-3}
8	1/2	4.860×10^{-4}	85 529[b]	3.202×10^{-3}
	3/2	8.739×10^{-4}	85 541[b]	5.755×10^{-3}
9	1/2	3.288×10^{-4}	88 062[b]	2.043×10^{-3}
	3/2	5.946×10^{-4}	88 069[b]	3.694×10^{-3}
10	1/2	2.311×10^{-4}	89 756[b]	1.382×10^{-3}
	3/2	4.194×10^{-4}	89 761[b]	2.508×10^{-3}
11	1/2	1.681×10^{-4}	90 946[b]	9.790×10^{-4}
	3/2	3.056×10^{-4}	90 950[b]	1.780×10^{-3}
12	1/2	1.258×10^{-4}	91 814[b]	7.189×10^{-4}
	3/2	2.291×10^{-4}	91 817[b]	1.309×10^{-3}
13	1/2	9.647×10^{-5}	92 466[b]	5.436×10^{-4}
	3/2	1.759×10^{-4}	92 468[b]	9.913×10^{-4}
14	1/2	7.556×10^{-5}	92 969[b]	4.212×10^{-4}
	3/2	1.379×10^{-4}	92 971[b]	7.688×10^{-4}
15	1/2	6.025×10^{-5}	93 364[b]	3.330×10^{-4}
	3/2	1.101×10^{-4}	93 366[b]	6.083×10^{-4}
16	1/2	4.880×10^{-5}	93 681[b]	2.679×10^{-4}
	3/2	8.920×10^{-5}	93 682[b]	4.896×10^{-4}
17	1/2	4.007×10^{-5}	93 939[b]	2.188×10^{-4}
	3/2	7.327×10^{-5}	93 940[b]	4.000×10^{-4}
18	1/2	3.330×10^{-5}	94 151[b]	1.810×10^{-4}
	3/2	6.091×10^{-5}	94 152[b]	3.310×10^{-4}
19	1/2	2.797×10^{-5}	94 328[b]	1.514×10^{-4}
	3/2	5.118×10^{-5}	94 329[b]	2.771×10^{-4}
20	1/2	2.372×10^{-5}	94 478[b]	1.280×10^{-4}
	3/2	4.341×10^{-5}	94 478[b]	2.343×10^{-4}
21–∞	1/2	2.094×10^{-4}		1.114×10^{-3}
	3/2	3.838×10^{-4}		2.042×10^{-3}
continuum	1/2	5.267×10^{-2}		6.230×10^{-2}
	3/2	1.024×10^{-1}		1.185×10^{-1}
	Sum	1.11		70.89(15)
	CACP	1.108		70.872

[a]Jin and Church, Ref. [24].
[b]Extrapolated by Ritz parametrizations, Ref. [49].

exact to deduce the principal transition absorption oscillator strengths from the measured lifetime values [24]. We have extended our CACP calculations up to $n'=20$ using measured energy levels and the Ritz parametrizations obtained by Edlén and Risberg [49]. The oscillator strengths corresponding to this measurement, together with our calculated values, are given in Table III.

Notice that the calculated oscillator strengths for $5\leq n'\leq 20$ make a contribution of only 3 parts in 10^3 to α_d and that the experimentally determined oscillator strengths of the $4s$-$4p$ transitions alone sum to 0.9424(2). Assuming again uncorrelated uncertainties in the measurements of the two lifetimes, they introduce an uncertainty of

± 0.15 (0.2%) into the determination of α_d. Contributions from higher n' are less than 0.285 (0.4%) and Eq. (4) would (with the single-active-electron assumption) set brackets of very small width.

IV. CONCLUSION

The results presented here indicate that the availability of high precision measurements of lifetimes for the lowest resonance transitions in alkali-metal-like ions can be combined with a knowledge of the strong cancellation effects that often occur in these systems and used to specify dipole polarizabilities to comparable precision. The Mg^+ and Ca^+ ions provide only two examples of what is a widely applicable method.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy, Fundamental Interactions Branch, Office of Basic Energy Sciences, Division of Chemical Sciences, under Grant No. DE-FG05-88ER13958. L. J. C. acknowledges stimulating and historically enriching discussions with the late Professor Bengt Edlén.

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