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POLARIZABILITIES OF CESIUM S, P, D, AND F STATES

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Abstract—The scalar and tensor polarizabilities of over sixty S, P, D, and F states of cesium are evaluated using a Coulomb approximation. The expected scaling of polarizabilities as n^{*7}, where n^* is the effective principal quantum number, was found to hold well for the higher excited states. The computed results agree closely with experimental measurements which are reviewed.

1. INTRODUCTION

Atomic polarizabilities are of interest as a test of atomic theory.[1] Due to recent progress in both experiment and theory, significantly more accurate data have been reported.[2–6] Alkali atoms are favourites of theorists because of their relatively simple atomic structure, which is based on a model of a single valence electron interacting with the nucleus that is shielded by inner core electrons. Cesium, in particular, is important for experiments such as parity violation,[7] electric dipole searches,[8] and application of laser cooling to atomic clocks.[9]

The polarizability of an atom governs its response to an applied electric field. The Hamiltonian describing the interaction of an electron in a state having quantum numbers J (angular momentum) and m (Zeeman component of angular momentum) with an external electric field E is[10]

$$H = -\frac{1}{2}\left[\alpha_0 + \alpha_2 \frac{3m^2 - J(J+1)}{J(2J-1)}\right]E^2. \tag{1}$$

Here, α_0 is the scalar polarizability which is given by

$$\alpha_0 = -\frac{2K_0}{[3(2J+1)]^{1/2}}, \tag{2}$$

and α_2, the tensor polarizability, is

$$\alpha_2 = 4\sqrt{\frac{5}{6}}K_2\left[\frac{J(2J-1)}{(2J+3)(2J+1)(J+1)}\right]^{1/2}, \tag{3}$$

where

$$K_L = \frac{3(2J+1)r_0}{8\pi^2}\sum_{J'} \lambda^2_{JJ'} f_{J'J} W(J, J, 1, 1,; L, J')(-1)^{J'-J}. \tag{4}$$

Here $r_0 = e^2/mc^2$ is the classical electron radius, $\lambda_{JJ'}$ is the wavelength for a transition from a state having angular momentum J' to a state with angular momentum J, $f_{J'J}$ is the transition oscillator strength, and W is a Racah coefficient. As is evident from Eqs. (2) and (3), the scalar and tensor polarizabilities are weighted sums of oscillator strengths or, equivalently, matrix elements of the valence electron position.

Theoretical work has focused so far on the ground and first excited states of cesium, primarily. The present paper reports the polarizabilities evaluated by using the Coulomb approximation of over sixty S, P, D, and F levels of cesium. The results are found to compare well with experimental

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observations. The paper is organized as follows. Section 2 discusses the theoretical work. The various experimental methods are reviewed in Sec. 3 and are compared with the computed results in Sec. 4.

2. COMPUTATION OF POLARIZABILITIES

Polarizabilities have been calculated using a variety of methods.[11] Marrus et al[12] used oscillator strengths found using the Hartree Fock method[13] to evaluate the polarizabilities of the $6S_{1/2}$ and $6P_{1/2,3/2}$ states. A different approach has been developed by Norcross[14] who uses a semiempirical potential. The latter is comprised of a Thomas-Fermi potential plus a term describing the polarization of the inner electron core. The various potential parameters are adjusted to obtain optimum agreement of computed and measured excited state energies.[15] Most recently, transition matrix elements $\langle 6, 7S_{1/2}|r|6, 7, 8P_{1/2,[FS43/2}\rangle$ where r is the position of the valence electron, have been computed using many body perturbation theory.[16] Extensive calculations have been made independently by two groups who obtain results agreeing to better than 1%.[2,3] Blundell et al used

Table 1. Computed polarizabilities.

State	n^*	$\alpha_0(a_0^3)$	$\alpha_2(a_0^3)$
$6S_{1/2}$	1.869	$3.94E2$	
$7S_{1/2}$	2.920	$6.14E3$	
$8S_{1/2}$	3.934	$3.79E4$	
$9S_{1/2}$	4.940	$1.53E5$	
$10S_{1/2}$	5.944	$4.75E5$	
$11S_{1/2}$	6.945	$1.24E6$	
$12S_{1/2}$	7.946	$2.84E6$	
$13S_{1/2}$	8.947	$5.90E6$	
$14S_{1/2}$	9.947	$1.14E7$	
$15S_{1/2}$	10.948	$2.06E7$	
$16S_{1/2}$	11.948	$3.54E7$	
$17S_{1/2}$	12.948	$5.77E7$	
$6P_{1/2}$	2.329	$1.29E3$	
$7P_{1/2}$	3.374	$2.94E4$	
$8P_{1/2}$	4.389	$2.21E5$	
$9P_{1/2}$	5.396	$1.02E6$	
$10P_{1/2}$	6.399	$3.49E6$	
$11P_{1/2}$	7.402	$9.86E6$	
$12P_{1/2}$	8.404	$2.44E7$	
$13P_{1/2}$	9.405	$5.40E7$	
$14P_{1/2}$	10.405	$1.10E8$	
$6P_{3/2}$	2.362	$1.60E3$	$-2.23E2$
$7P_{3/2}$	3.406	$3.69E4$	$-4.28E3$
$8P_{3/2}$	4.421	$2.82E5$	$-3.02E4$
$9P_{3/2}$	5.428	$1.31E6$	$-1.34E5$
$10P_{3/2}$	6.432	$4.51E6$	$-4.49E5$
$11P_{3/2}$	7.434	$1.28E7$	$-1.25E6$
$12P_{3/2}$	8.436	$3.16E7$	$-3.05E6$
$13P_{3/2}$	9.327	$7.03E7$	$-6.70E6$
$14P_{3/2}$	10.438	$1.43E8$	$-1.36E7$
$5D_{3/2}$	2.548	$-4.18E2$	$3.80E2$
$6D_{3/2}$	3.528	$-5.32E3$	$8.62E3$

Continued opposite

Table 1—*continued.*

State	n^*	$\alpha_0(a_0^3)$	$\alpha_2(a_0^3)$
$7D_{3/2}$	4.525	−6.52*E*4	7.04*E*4
$8D_{3/2}$	5.525	−3.66*E*5	3.36*E*5
$9D_{3/2}$	6.525	−1.40*E*6	1.19*E*6
$10D_{3/2}$	7.525	−4.22*E*6	3.41*E*6
$11D_{3/2}$	8.525	−1.09*E*7	8.52*E*6
$12D_{3/2}$	9.524	−2.51*E*7	1.91*E*7
$13D_{3/2}$	10.525	−5.22*E*7	3.89*E*7
$5D_{5/2}$	2.555	−5.18*E*2	7.04*E*2
$6D_{5/2}$	3.536	−7.95*E*3	1.70*E*4
$7D_{5/2}$	4.534	−8.71*E*4	1.40*E*5
$8D_{5/2}$	5.534	−4.72*E*5	6.75*E*5
$9D_{5/2}$	6.534	−1.77*E*6	2.38*E*6
$10D_{5/2}$	7.534	−5.30*E*6	6.85*E*6
$11D_{5/2}$	8.534	−1.36*E*7	1.71*E*7
$12D_{5/2}$	9.534	−3.11*E*7	3.83*E*7
$13D_{5/2}$	10.534	−6.47*E*7	7.85*E*7
$4F_{5/2}$	3.978	4.51*E*3	1.34*E*3
$5F_{5/2}$	4.974	1.12*E*6	−3.84*E*5
$6F_{5/2}$	5.972	7.77*E*6	−2.72*E*6
$7F_{5/2}$	6.971	−3.67*E*5	3.09*E*5
$8F_{5/2}$	7.970	−9.34*E*5	7.87*E*5
$9F_{5/2}$	8.969	−2.13*E*6	1.79*E*6
$10F_{5/2}$	9.969	−4.44*E*6	3.73*E*6
$11F_{5/2}$	10.969	−8.58*E*6	7.21*E*6
$4F_{7/2}$	3.978	4.14*E*3	1.99*E*3
$5F_{7/2}$	4.974	1.11*E*6	−4.44*E*5
$6F_{7/2}$	5.972	7.72*E*6	−3.14*E*6
$7F_{7/2}$	6.970	−4.04*E*5	4.03*E*5
$8F_{7/2}$	7.970	−1.03*E*6	1.03*E*6
$9F_{7/2}$	8.969	−2.37*E*6	2.37*E*6
$10F_{7/2}$	9.968	−4.94*E*6	4.94*E*6
$11F_{7/2}$	10.969	−9.57*E*6	9.57*E*6

their matrix values to determine ionization energies and hyperfine constants which were found to agree with observed values to better than 1%.[3] The same matrix elements have been used by us to compute the polarizability of the $6S_{1/2}$ state which is listed in Table 3.

In this work, polarizabilities were computed using oscillator strengths found using the method of Bates and Damgaard.[17] It determines transition matrix elements using experimentally measured excited state energies and assumes a Coulomb potential to describe the interaction of the valence electron and the atom. Hence, it best describes excited states whose wavefunctions have minimal penetration of the inner electron core. This method has been used to determine polarizabilities of the lighter alkali atoms for which the computed values were found to agree with the experimental data to within a few percent.[18]

The results, shown in Table 1, were computed using excited state energies determined by Moore.[19] The polarizabilities are predicted to scale as n^{*7} where n^* is the effective principal quantum number, for highly excited states where effects of the inner core electrons are negligible.[20] This was tested by plotting α/n^{*7} versus n^* for the nL_J states where L is the orbital angular momentum. Figure 1 shows the plot obtained for the $(6\text{–}17)S_{1/2}$ states. The scaled polarizabilities α/n^{*7} asymptotically

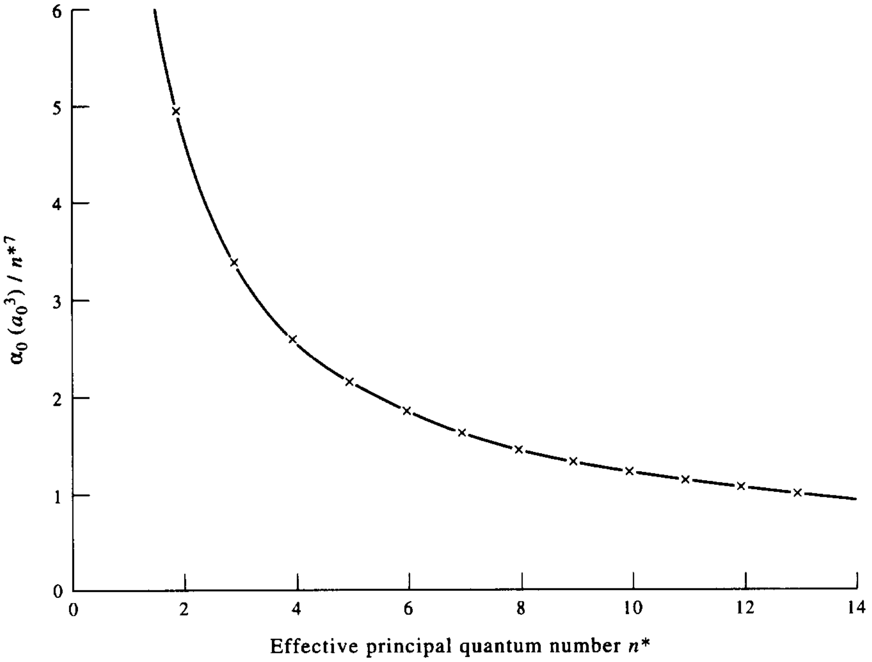

Fig. 1. The dependence of polarizability α_0 upon effective principal quantum number n^* for $(6\text{–}17)S_{1/2}$ states. The crosses are data listed in Table 1 which are fit to the curve as described in the text. The measured values of α_0 are not shown since they overlap the theoretical values.

approach a limiting value as n^* increases, which was found by using a least squares fit of the data to the function $\alpha/n^{*7} = A + Bn^{*-C}$. The polarizability values obtained using the fitted constants A, B, and C listed in Table 2, agreed with the theoretical results to within 2%. This is quite remarkable considering that the polarizability increases very rapidly with n^*. For the $(6\text{–}17)S_{1/2}$ states, α_0 increases by over five orders of magnitude. The scaling relations given by Table 2 are therefore useful for extrapolating the polarizabilities of higher states.

Table 2. Fitted parameters of curves $\alpha/n^{*7} = A + Bn^{*-C}$ determined using the states listed.

Scalar Polarizability $\alpha_0(a_0^3)$			
States	$A(a_0^3)$	$B(a_0^3)$	C
$(6-17)S_{1/2}$	0.05	8.51	0.88
$(6-14)P_{1/2}$	8.73	-24.43	1.81
$(6-14)P_{3/2}$	11.53	-27.88	1.50
$(6-13)D_{3/2}$	-5.17	15.38	0.99
$(6-13)D_{5/2}$	-6.22	17.99	1.00
$(7-11)F_{5/2}$	-0.46	0.0	
$(7-11)F_{7/2}$	-0.51	0.0	
Tensor Polarizability $\alpha_2(a_0^3)$			
States	$A(a_0^3)$	$B(a_0^3)$	C
$(6-14)P_{3/2}$	-1.03	3.40	2.26
$(6-13)D_{3/2}$	3.34	-8.59	1.14
$(6-13)D_{5/2}$	7.01	-16.25	1.02
$(7-11)F_{5/2}$	0.38	0.0	
$(7-11)F_{7/2}$	0.51	0.0	

3. EXPERIMENTAL METHODS

The first method developed to determine atomic polarizabilities is to measure the deflection of an atomic beam caused by an inhomogeneous electric field. It is particularly well suited for studying the ground state since the beam atoms must predominantly populate a single level. Table 3 lists the most recent result of $\alpha_0(6S_{1/2})$ made by Hall et al.[21] Bedersen et al have used both inhomogeneous magnetic H and electric E fields to measure the cesium ground state polarizability.[22] The atomic beam is not deflected if the magnetic and electric forces cancel, i.e., $\alpha E \frac{dE}{dz} = \mu \frac{dH}{dz}$. The polarizability is found provided the field gradients $\frac{dE}{dz}$, $\frac{dH}{dz}$ and the magnetic moment μ are known.

The first experimental determination of an excited state polarizability was done by Marrus et al.[16] A beam of cesium atoms passed through a uniform electric field generated by two parallel plates. On either side of the plates, was a region of inhomogeneous magnetic field. The atoms were illuminated by a cesium lamp when they were between the electric field plates. The lamp spectrum contained two peaks corresponding to transitions from the $6P_{1/2}$ state to the two ground state hyperfine levels. When zero voltage was applied to the plates, the lamp excited atoms to the $6P_{1/2}$ state. The effect of this excitation and subsequent radiative decay back to the ground state, is to transfer some atoms that initially occupy the $m = 1/2$ ground state Zeeman sublevel to the

Table 3. Experimental and theoretical values of polarizabilities.

State	$\alpha_0(a_o^3)$	$\alpha_2(a_o^3)$	Method	Reference
$6S_{1/2}$	$(4.27 \pm .31)E2$		Atomic beam deflection	21
	$(4.02 \pm .08)E2$		Atomic beam E-H balance	22
	$(4.54)E2$		Hartree Fock Theory	12
	$(4.006)E2$		Semiempirical Potential Model	14
	$(3.845)E2$		Many Body Perturbation Theory	2,3
$7S_{1/2}$	$(6.67 \pm .39)E3$		Laser using Fabry-Perot	24
	$(6.111 \pm .021)E3$		Laser using Fabry-Perot	25
	$(6.061)E3$		Semiempirical Potential Model	14
$10S_{1/2}$	$(4.96 \pm .24)E5$		Laser using Fabry-Perot	23
	$(4.791 \pm .011)E5$		Laser using Acousto-Optic Modulator	6
$11S_{1/2}$	$(1.30 \pm .065)E6$		Laser using Fabry-Perot	23
	$(1.2463 \pm .0010)E6$		Laser using Acousto-Optic Modulator	6
$12S_{1/2}$	$(2.90 \pm .18)E6$		Laser using Fabry-Perot	23
	$(2.8713 \pm .0023)E6$		Laser using Acousto-Optic Modulator	6
$13S_{1/2}$	$(6.65 \pm .69)E6$		Laser using Fabry-Perot	23
	$(6.0010 \pm .0049)E6$		Laser using Acousto-Optic Modulator	6
$6P_{1/2}$	$(1.26 \pm .20)E3$		Atomic Beam Deflection	12
	$(1.262)E3$		Hartree Fock Theory	12
	$(1.327)E3$		Semiempirical Potential Model	14
$7P_{1/2}$	$(2.96 \pm .06)E4$		Lamp excitation of cell using hyperfine structure	27

Continued overleaf

Table 3—*continued.*

State	$\alpha_0(a_o^3)$	$\alpha_2(a_o^3)$	Method	Reference
$6P_{3/2}$	$(1.61 \pm .24)E3$	$(-2.62 \pm .04)E2$	Atomic beam deflection	12
	$(1.596)E3$	$(-2.46)E2$	Hartree Fock Theory	12
	$(1.6383)E3$	$(-2.617)E2$	Semiempirical Potential Model	14
$7P_{3/2}$	$(3.79 \pm .08)E4$	$(-4.43 \pm .12)E3$	Lamp excitation of cell using hyperfine structure	26
		$(-4.33 \pm .17)E3$	Level Crossing	10
		$(-4.00 \pm .08)E3$	Experimental	27
$8P_{3/2}$		$(-3.07 \pm .12)E4$	Laser using Fabry-Perot	23
$8D_{3/2}$		$(3.33 \pm .16)E5$	Laser using Fabry-Perot	23
$9D_{3/2}$	$(-1.45 \pm .12)E6$	$(1.26 \pm .06)E6$	Laser using Fabry-Perot	23
$10D_{3/2}$	$(-4.64 \pm .69)E6$	$(3.39 \pm .16)E6$	Laser using Fabry-Perot	23
$8D_{5/2}$		$(7.34 \pm .40)E5$	Laser using Fabry-Perot	23
$9D_{5/2}$	$(-2.05 \pm .10)E6$	$(2.66 \pm .14)E6$	Laser using Fabry-Perot	23
$10D_{5/2}$	$(-5.40 \pm .52)E6$	$(7.14 \pm .36)E6$	Laser using Fabry-Perot	23
$11D_{5/2}$	$(-1.53 \pm .14)E7$	$(1.62 \pm .16)E7$	Laser using Fabry-Perot	23
$13D_{5/2}$		$(7.66 \pm .40)E7$	Laser using Fabry-Perot	23

$m = -1/2$ sublevel. The deflection of these atoms by the second magnet is opposite that which they experienced due to the first magnet. In general, an applied electric field shifts the atomic levels out of resonance with the lamp. The atoms then remain in the same Zeeman sublevel and the second magnet adds to the beam deflection. However, when the Stark shift equals the ground state hyperfine splitting of 9.193 GHz, the lamp is again in resonance with the atoms. Some of the beam atoms can then transfer between the two ground state Zeeman sublevels and experience no net deflection. The polarizabilities of the $6P_{1/2}$ and $6P_{3/2}$ states were found by measuring this electric field.

A second method for studying excited states is level crossing spectroscopy. The atoms are first excited using either lamps or lasers, and fluorescence resulting from their subsequent radiative decay is detected. An electric field perturbs the excited state hyperfine levels, changing the fluorescent intensity. This effect is strongest when two energy levels cross. The polarizability is then found by measuring the required crossing field and using the known excited state hyperfine splitting. This method was used to determine α_2 of the $7P_{3/2}$ state.[10]

The development of narrow linewidth lasers enabled the direct measurement of Stark shifts. Lasers can excite a desired state which fluoresces when it radiatively decays. The change in the laser frequency needed to keep an atom in resonance when an electric field is applied, can be measured using a Fabry-Perot interferometer. Fredriksson et al have determined a large number of excited state polarizabilities attaining an accuracy of about 5%.[23] This uncertainty arises primarily from the accuracy with which changes in laser frequency can be determined, which was done using an etalon having a free spectral range of 75 MHz.

The measurement of changes in laser frequency is greatly improved using an acousto-optic modulator in conjunction with a signal synthesizer to precisely frequency shift a laser. This was

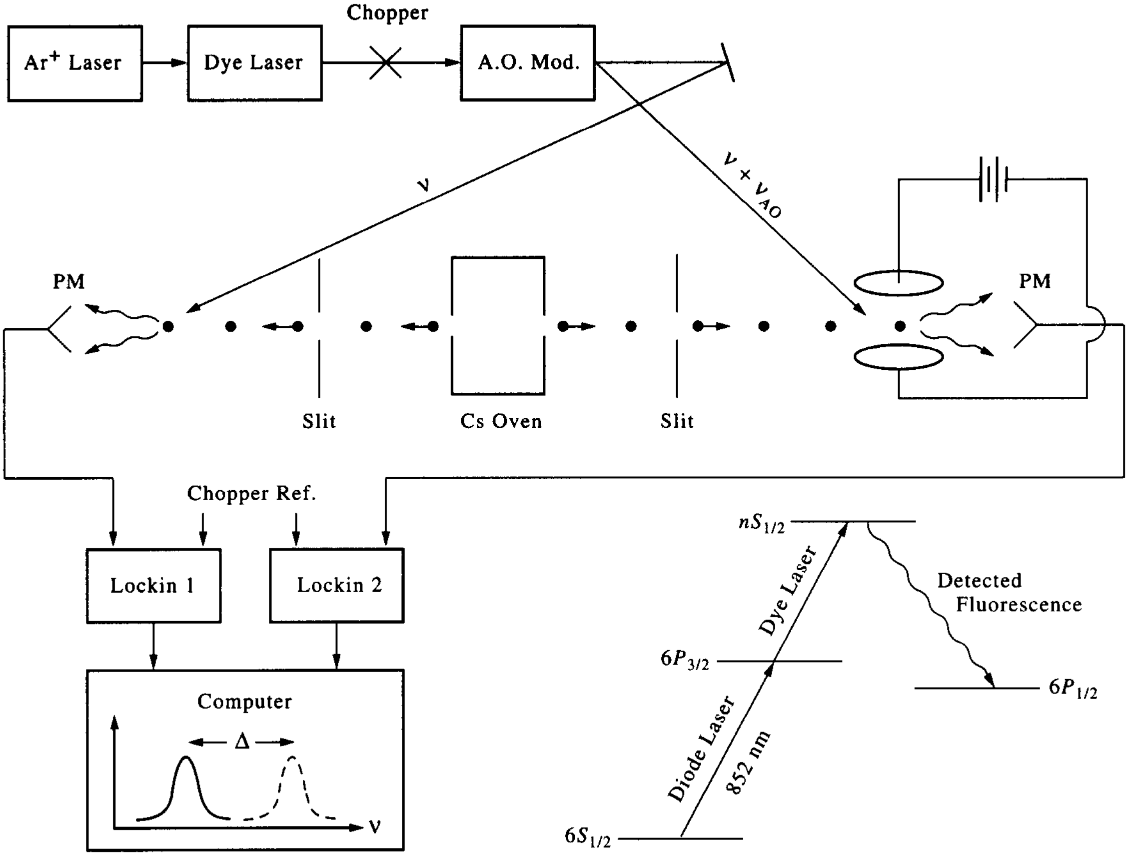

Fig. 2. The apparatus used to determine $\alpha_0(10\text{–}13S_{1/2})$. The Stark shift is measured using a laser with a part of its frequency shifted by an acousto-optic modulator.

done by Tanner et al in their study of the $6S_{1/2} \rightarrow 6P_{3/2}$ transition of cesium.[5] The Stark shift of the cesium $6S_{1/2} \rightarrow 6P_{1/2}$ transition was measured by Hunter et al using two diode lasers.[4] One laser excited an atomic beam that passed through an electric field while the other laser traversed a cesium cell. The fluorescent signals were then used to lock the laser frequencies. The Stark shift was found by measuring the beat frequency of the two diode lasers using a fast detector.

The advantage of atomic beams as compared to cells is that transitions are not Doppler broadened. In addition, cells cannot sustain as high electric fields as can exist in vacuum. Figure 2 shows the apparatus used to measure the polarizability of the $(10\text{–}13)S_{1/2}$ states.[6] Two atomic beams are first excited by a circularly polarized diode laser to the $F = 5$ hyperfine level of the $6P_{3/2}$ state. The $6P_{3/2} \rightarrow (10\text{–}13)S_{1/2}$ transition is then excited by a ring dye laser. Part of the dye laser is shifted by an acousto-optic modulator and intersects the atomic beam passing between two circular plates used to generate an electric field. The unshifted laser beam excites the other atomic beam. The dye laser is then scanned across the resonance which is found by detecting fluorescence using a photomultiplier in conjunction with a lock-in amplifier. The polarizability is found by measuring the field such that both atomic beams are simultaneously excited. The accuracy of better than 0.1% is limited by uncertainties in the measurements of high voltages and the distance separating the field plates.

4. DISCUSSION AND CONCLUSIONS

Table 3 lists polarizabilities obtained from experiment and theory. The values computed using the Coulomb approximation listed in Table 1 lie within 1% of all the experimental results except for the $6P_{1/2,3/2}$ states. The Stark shifts of transitions to these states from the ground state have been determined as follows,[4,5]

$$\alpha_0(6P_{1/2}) - \alpha_0(6S_{1/2}) = 923.35 \pm 7.12\alpha_0^3$$

$$\alpha_0(6P_{3/2}) + \alpha_2(6P_{3/2}) - \alpha_0(6S_{1/2}) = 977.8 \pm 1.9a_0^3$$

where a_0 is the Bohr radius. The corresponding values obtained from Table 1 are 900 and $981a_0^3$. This discrepancy is not surprising since the Coulomb potential does not adequately describe the effects of the core electrons nor take into account the spin orbit interaction which is largest for the $6P_{1/2,3/2}$ states. These two effects decrease rapidly in size as the principal quantum number increases. Hence, the polarizabilities obtained using the Coulomb potential are expected to agree well with the experimental results of the highly excited states. This was indeed found to be the case. Improved agreement with the measured Stark shifts of the $6P_{1/2,3/2}$ states has been achieved by Norcross using a semiempirical potential who obtained 926 and $976a_0^3$ respectively.[14]

In conclusion, we note that a Coulomb approximation can account for nearly all the polarizabilities that have been measured. Polarizabilities of higher states yet to be examined by experiment can be reliably estimated using simple scaling relations involving the effective principal quantum number n^*.

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