PHYSICAL REVIEW A VOLUME 31, NUMBER 4 APRIL 1985

Scalar polarizabilities and avoided crossings of high Rydberg states in Rb

M. S. O'Sullivan and B. P. Stoicheff
Department of Physics, University of Toronto,
Toronto, Ontario, M5S 1A7 Canada

(Received 10 August 1984)

Two-photon Doppler-free spectroscopy has been used to measure Stark shifts of $n\,^2S$ Rydberg states of ^{85}Rb (for $n=15$ to 80). Scalar polarizabilities α_0 were found to obey the relation α_0 [in MHz/(V/cm)^2] $=2.202(28)\times10^{-9}n^{*6}+5.53(13)\times10^{-11}n^{*7}$, in good agreement with theoretical values. Avoided crossings were also determined, and first avoided crossings (with the $n-3$ manifold of levels) fitted the relation E (in V/cm) $=4.638(6)\times10^{8}/n^{*5}+1.528(18)\times10^{10}/n^{*7}$.

Earlier studies of the Stark effect in Rb have included calculations of Stark maps in the spectral region of $n=15$,[1] measurements of field ionization,[2] and determinations of polarizabilities of low-lying S, P, and D states.[3–6] These experiments were based on atomic beams and the most recent used either pulsed or cw lasers with field ionization or photon-counting detection. However, they did not make use of the high resolution (~1 MHz) afforded by two-photon Doppler-free absorption spectroscopy. This is the main consideration of the present Brief Report, which gives measurements of Stark shifts of highly excited states ($n>15$) in precisely known electric fields. While our experiments were in progress, Penent, Delande, Biraben, and Gay[7] published a brief account of their results on Rb atoms in crossed electric and magnetic fields using this spectroscopic technique.

In this paper we wish to report the results of precise Stark-shift measurements for $n\,^2S$ states of Rb, with principal quantum number n from 15 to 80. Relatively low electric fields were used throughout, since Stark shifts were clearly quadratic only over the range zero to the first avoided crossing (AC) for each level. From these measurements, values of scalar polarizabilities for highly excited Rydberg states of Rb were determined for the first time. The experimental arrangement is similar in many respects to experiments already reported from this laboratory.[8,9] Two cells with Rb vapor at 3–5 mTorr pressure were used: one a reference cell with no electric field and the other fitted with Stark electrodes. Both cells contained thermionic diode detectors.[10] Two-photon Doppler-free transitions are induced with cw dye-laser radiation, a part of which is also directed to a 1.2-m confocal étalon. As the dye-laser frequency is tuned, signals from the two cells are recorded on a 3-pen chart recorder, along with transmission fringes from the étalon. This arrangement allows the measurement of relative shifts and splittings at various electric fields, to within 2 MHz.

The Stark electrodes consist of two circular (stainless-steel) plates, 6 cm in diameter, ground flat to within 5 μm, and separated by a distance of 1.024 45(8) cm at 22 °C, by three fused quartz spacers. A fine grid of two lines per millimeter was spark cut in the top plate, while maintaining surface quality. The field direction was chosen so that positive ions drift through this grid (with 25% transmission) and into the thermionic diode detector. The Stark plates are connected to a well-regulated voltage supply (0.2-mV ripple), and voltages read by a calibrated voltmeter to 0.3% accuracy. Small corrections were made to the electric field arising from electrons from the diode which entered the field region. These corrections were determined from observations of the dependence of electric field of an avoided crossing (AC) of levels as a function of laser beam-waist position between the Stark plates. For the position selected in the experiments, this correction to the field was $<0.1\%$ at power supply voltages above 4 V, and increased to $\sim0.3\%$ at 1 V and to $\sim2\%$ at 0.15 V. Also, it is to be noted that from the observed broadening of the shifted $15S\leftarrow5S$ line at the highest field used in these experiments (983 V/cm) a field inhomogeneity of 0.03% was estimated.

For each transition, $n\,^2S\leftarrow5\,^2S$, groups of three or more laser scans were recorded at each of six different field strengths between zero and the first avoided crossing (AC). A typical scan is shown in Fig. 1 for the transition $15\,^2S\leftarrow5\,^2S$ at 492(2) V/cm. The observed line is symmetrical and sharp [4 MHz full width at half maximum (FWHM)], and shifted by 957.0 MHz to lower frequency. The average fluctuation in measured Stark shifts within a group of scans was 1.6 MHz, most of this being the uncertainty in locating centers of spectral lines and of étalon fringes. Over the electric-field range studied for each n, the measured Stark shifts were quadratic in the field to within 1.3 MHz. Thus values of scalar polarizability α_0 were determined by a least-squares fit of shifts to a quadratic function of electric field. The resulting values are listed in Table I,

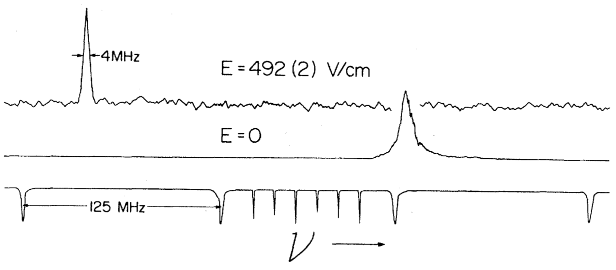

FIG. 1. Spectra of the $15\,^2S\leftarrow5\,^2S$ transition at $E=0$ and 492(2) V/cm recorded together with étalon fringes to provide a frequency scale. Note that the speed of the dye-laser scan has been increased for most of the Stark shift. The total shift is 7.662 orders $\times$ 124.9 = 957.0 MHz.

TABLE I. Scalar polarizabilities of ^2S states of Rb, and ranges of electric fields used in their evaluation.

Level $n\,^2S$	Polarizability α_0 [MHz/(V/cm)^2]	Electric-field range (V/cm)
15S	$7.929(45)\times 10^{-3}$	98–492
20S	$7.229(29)\times 10^{-2}$	28–188
25S	$3.745(45)\times 10^{-1}$	19–75
30S	1.390(15)	4–28
35S	4.202(36)	4–14
40S	10.57(20)	1–5
45S	24.9(1.0)	1.4–3.2
50S	50.61(32)	0.7–2
55S	96.4(1.8)	0.6–1
60S	171.1(7.4)	015–0.7
63S	249.1(8.0)	0.35–0.7
65S	318(12)	0.2–0.7
67S	380(13)	0.15–0.45
70S	534(20)	0.15–0.3
75S	802(80)	0.003–0.2
80S	1340(130)	0.1–0.16

along with the electric-field ranges used in their evaluations. The quoted uncertainties are twice the statistical error: accuracies are ~1% or better up to $n=35$, and then increase to ~10% at $n=80$ because of the limited range of electric field available (before the first AC) for determination of α_0. The values of α_0 obtained here conform to the following function of n^* (the effective quantum number):

$$\alpha_0 = 2.202(28)\times 10^{-9} n^{*6} + 5.53(13)\times 10^{-11} n^{*7} , \quad (1)$$

measured in MHz/(V/cm)^2. A test of this relation by extrapolating to values of α_0 for levels 9S and 10S, yielded the values $1.031(20)\times 10^{-4}$ and $2.709(40)\times 10^{-4}$ MHz/(V/cm)^2, respectively. These are in very good agreement with values measured earlier:[3] $1.02(9)\times 10^{-4}$ and $2.80(25)\times 10^{-4}$ MHz/(V/cm)^2.

Measurements of electric fields for the first avoided crossings of some $n\,^2S$ levels with the $n-3$ level manifold were also made. These fields were determined from observed intensity variations in a two-photon transition with changing electric fields. In the immediate vicinity of an AC, coupling between the near-degenerate states results in a lowering of intensity, which is acutely sensitive to the magnitude of the electric field. The relative size of such intensity variations increases with decreasing n, from ~30% at $n=52$ to at least 60% for $n<30$. An example is shown in Fig. 2 for $n=20$, with four laser scans taken at intervals ~1 V/cm, and demonstrating a sharp reduction of signal intensity at 379.0 V/cm which is taken to be the first AC. Values of AC fields obtained in this way are given in Table II for a number of levels from $n=18$ to 52. These values all fit the relation

$$E_{\mathrm{AC}} = \frac{4.638(6)\times 10^{8}}{n^{*5}} + \frac{1.528(18)\times 10^{10}}{n^{*7}} , \quad (2)$$

with E_{AC} given in V/cm. The first term is consistent with the value $4.5\times 10^8/n^{*5}$ expected using hydrogenic theory, and corresponds to the field which causes a near coincidence in energy of an $n\,^2S$ state and a Stark-shifted $(n-3)F$ state in Rb.

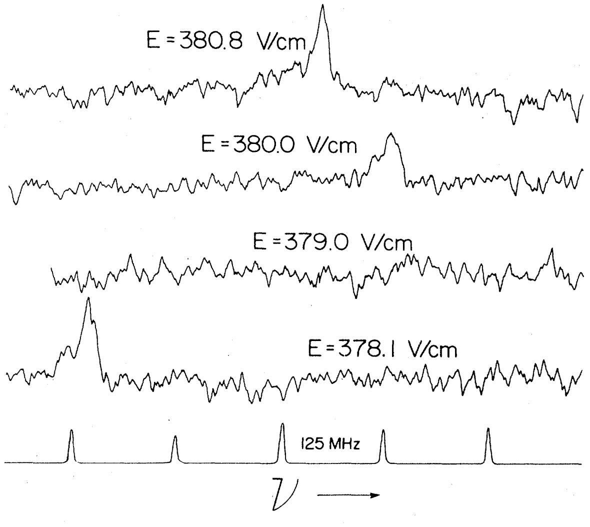

FIG. 2. Spectra of the $20\,^2S \leftarrow 5\,^2S$ transition at four values of the electric field near the first avoided crossing at 379.0 V/cm. Note that the signal intensity has decreased to that of the noise, between $E=378.1$ and 380.0 V/cm.

Our measurements include values for the 37S and 18S states, for which a measurement and a calculation, respectively, have already been reported. For 37S, Penent *et al.*[7] report on measurements from which an AC field of 11.69 V/cm may be deduced. However, this does not agree with our value for the first AC, 10.69 V/cm, but it is in reasonable agreement with the field of 11.57(4) V/cm which we obtained for the second AC of 37S with the $n=34$ manifold. Zimmerman, Littman, Kash, and Kleppner[1] have calculated the entire Stark structure in the vicinity of the 18S level of Rb. From their graph we measure 755(5) V/cm for the field of the first AC, which is corrected to 738(5) V/cm

TABLE II. Measured electric fields for first avoided crossings of $n\,^2S$ levels with $n-3$ level manifold.

Level $n\,^2S$	First avoided crossing field (V/cm)
18	733.5[a]
20	379.0
25	99.36
30	34.72
35	14.59
37	10.69
40	6.959
43	4.704
45	3.674
46	3.260
47	2.901
48	2.592
49	2.320
50	2.078
51	1.870
52	1.691

[a] All values of electric fields for first avoided crossings are considered to be accurate to 0.4%.

when the more recent value of the quantum defect, 3.1320,[8,11] is used in place of the 3.135(1) used in their calculations. This field of 738(5) V/cm is in good agreement with our measurement of 733.5(3.0) V/cm.

It is possible to obtain a good approximation to Eq. (1) for scalar polarizabilities from known theory and available radial matrix elements. When fine-structure interactions are neglected, the scalar polarizabilities for nS states are given by the expression

$$\alpha_0(nS)=\tfrac{2}{3}e^2\sum\frac{|\langle nS|r|iP\rangle|^2}{E_{iP}-E_{nS}} \quad . \tag{3}$$

Here, the summation is carried over all P states; E_{iP} and E_{nS} are the energies of iP and nS states, and e, r have their usual meanings.

The n dependence of Eq. (3) can be approximated using available radial matrix elements.[12] In the low-field limit, more than 99% of the scalar polarizability of each nS state is due to perturbations with its two nearest iP states, those with $i=n$ and $n-1$. The corresponding matrix elements may be written in a form consistent with the Bates-Damgaard approximation:

$$\langle nS|r|iP\rangle=\tfrac{3}{2}A(i)a_0(n^*+x_i)[(n^*+x_i)^2-1]^{1/2} \quad . \tag{4}$$

Here, n^* is the effective quantum number of the nS state, x_i the difference in effective quantum numbers of iP and nS states, and $A(i)$ can be calculated from known values of radial matrix elements. For excited states with $n>20$ in Rb, the $A(i)$ are essentially constants, namely,

$$A(n)=0.7491,\quad A(n-1)=0.7241 \quad .$$

When these values and Eq. (4) are substituted in Eq. (3) we obtain, in MHz/(V/cm)^2,

$$\alpha_0=2.228(20)\times10^{-9}n^{*6}+4.90(60)\times10^{-11}n^{*7} \quad .$$

(Terms of lower order than n^{*6} are insignificant, and the errors arise from an assumed uncertainty of 0.5% in calculated radial matrix elements.) The coefficients in n^{*6} and n^{*7} are in good agreement with the values $2.202(28)\times10^{-9}$ and $5.53(13)\times10^{-11}$, respectively, derived from the present measurements of Stark shifts at low fields.

The application of two-photon, Doppler-free spectroscopy to the measurement of Stark shifts in Rb, has permitted the evaluation of scalar polarizabilities for levels 15S to 80S. The values are found to increase by a factor of 2×10^5 over this range of levels, and are in good agreement with theoretical calculations.

We are indebted to E. Latal, J. Legge, and D. Warwick for their skillful construction and assembly of the Stark cell.

---

[1] M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A **20**, 6 (1979).
[2] S. Liberman and J. Pinard, Phys. Rev. A **20**, 507 (1979).
[3] K. Frederiksson and S. Svanberg, Z. Phys. A **281**, 189 (1977).
[4] W. Hogervorst and S. Svanberg, Phys. Scr. **12**, 67 (1975).
[5] S. Svanberg, Phys. Scr. **5**, 132 (1972).
[6] R. Marrus, D. McColin, and J. Yellin, Phys. Rev. **147**, 55 (1966).
[7] F. Penent, D. Delande, F. Biraben, and J. C. Gay, Opt. Commun. **49**, 184 (1984).
[8] B. P. Stoicheff and E. Weinberger, Can. J. Phys. **57**, 2143 (1979).
[9] B. P. Stoicheff and E. Weinberger, Phys. Rev. Lett. **44**, 733 (1980).
[10] K. C. Harvey and B. P. Stoicheff, Phys. Rev. Lett. **38**, 537 (1977); K. C. Harvey, Rev. Sci. Instrum. **52**, 204 (1981); D. C. Thompson and B. P. Stoicheff, *ibid.* **53**, 822 (1982).
[11] C.-J. Lorenzen and K. Niemax, Phys. Scr. **27**, 300 (1983).
[12] F. Gounand, J. Phys. (Paris) **40**, 457 (1979).