Dan,
This paper came
first
• Apparatus was operational
• 2 dye lasers for excitation
• N₂ laser pumped
• Optical detection using single photon timing

STARK MIXING SPECTROSCOPY IN CESIUM[‡]

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We present a new technique for selectively populating excited states which are inaccessible by dipole excitation from the ground state. The method uses a static electric field to introduce a component of a dipole-allowed state into the state of interest. We have applied the method to cesium to measure lifetimes and a Stark mixing coefficient. The results are $\tau(6\,^2D_{5/2}) = 64(2)$ ns, $\tau(7\,^2D_{5/2}) = 92.5(15)$ ns, and $\langle 6\,^2D_{5/2}|\mathcal{E}ez|7\,^2P_{3/2}\rangle/(E_{7P} - E_{6D}) = 0.7(3) \times 10^{-3}\,\mathcal{E}$, where $\mathcal{E}$ is in kV/cm.

Atomic levels which are accessible from the ground state by electric dipole transitions have traditionally received the most exhaustive study. Recently, a number of new techniques have been introduced for studying levels which were previously inaccessible from the ground state. Among these are: 1) Optical cascade [1] where, for example, alkalis in high-lying P states are populated via allowed dipole transitions from an S ground state. Lower lying S and D levels are then populated by cascade from the P states; 2) Stepwise excitation [2] which makes use of two consecutive allowed transitions by way of an intermediate level to populate a high-lying level; 3) Two-photon excitation [3] which uses a second-order transition process, for instance, an S → D, or S → S transition, via absorption of two photons. The sum of the photon energies equals the energy difference between the two levels.

We present here a new technique, "Stark mixing spectroscopy", in which an applied constant electric field "mixes" a small amount of a dipole-accessible state with a nearby dipole-forbidden state [4]. This allows radiative dipole excitation of the perturbed state from the ground state. This method is related to two-photon excitation; the role of a transverse electric field is replaced by a longitudinal static electric field, what we might describe as a "zero frequency photon". The method allows one to populate selectively a given "inaccessible" state. In this letter we report application of the technique to measure several previously unobserved lifetimes in atomic cesium, and to measure the Stark mixing coefficient which couples two of its close-lying excited states.

The levels of cesium, fig. 1, are effectively those of a single electron outside a spherically symmetric core. Let us look at the case of exciting the $6\,^2D_{5/2}$ level from the $6\,^2S_{1/2}$ ground state. A static electric field is used to mix a small amount of the $7\,^2P_{3/2}$ state into the 6D state.

The effect of the electric field can most easily be described by first-order perturbation theory. The electric field mixes all P and F states with the 6D state, but the only significant contribution is from the 7P state:

$$|6D\rangle' \approx |6D\rangle^0 + a(6D, 7P)|7P\rangle^0 . \qquad (1)$$

Here $|6D\rangle'$ is the perturbed wave function; $|6D\rangle^0$ and $|7P\rangle^0$ are unperturbed wave functions. The Stark mixing coefficient, $a(6D, 7P)$, is given by
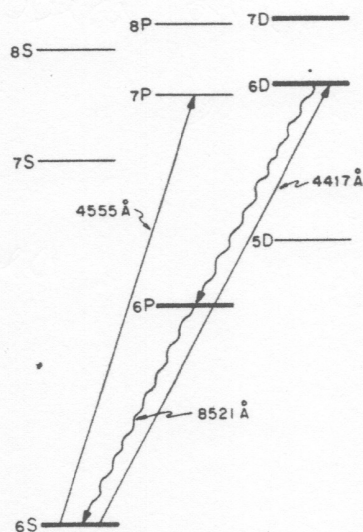
Fig. 1. Relevant energy levels of Cs.

Fig. 2. Schematic diagram of apparatus.

$$a(6D, 7P) = \frac{\mathcal{E}\langle 6D|ez|7P\rangle}{E_{7P} - E_{6D}}.$$

$\mathcal{E}$ is the magnitude of the static electric field, whose direction lies along the z-axis.

We can now excite the $|6D\rangle'$ level with light at the 6S–6D transition, 4417 Å. For a given incident photon flux and atomic beam density the rate of absorption is given by

$$R_{6S-6D'} \alpha |\langle 6S|ez|6D\rangle|^2 = |a(6D, 7P)|^2 |\langle 6S|ez|7P\rangle|^2 .$$

$$(2)$$

With moderate fields, this absorption rate can be large enough to provide a useful population in the 6D state. In addition, the Stark mixing coefficient is of interest itself. To measure it, let us compare $R_{6S-6D'}$ with the rate of allowed excitation of the 7P level under the same photon flux and atom density:

$$R_{6S-7P} \alpha |\langle 6S|ez|7P\rangle|^2 .$$

$$(3)$$

Combining eq. (2) and eq. (3) we see that:

$$|a(6D, 7P)|^2 = \frac{R_{6S-6D'}}{R_{6S-7P}}.$$
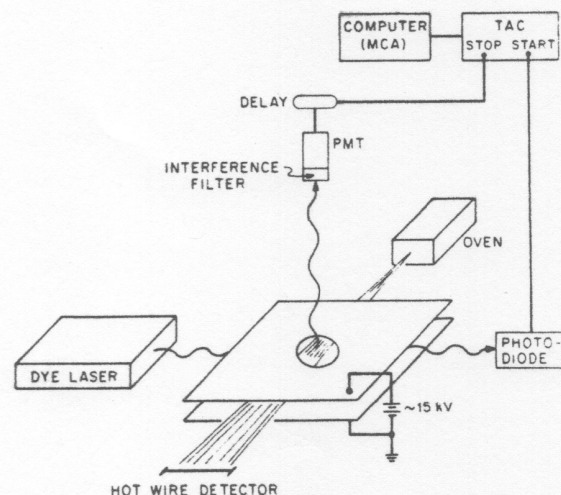
$$(4)$$

Thus, a measurement of the ratios of excitation

rates of the "forbidden" transition to the allowed transition yields a value for $a(6D, 7P)$, the Stark mixing coefficient.

To perform the experiment measuring $a(6D, 7P)$, we employed the arrangement illustrated in fig. 2. The source of excitation was a pulsed, tunable dye laser of a design similar to Hänsch's [5], sidepumped by an AVCO C950 N_2 laser with a repetition rate of up to 100 Hz. The duration of the dye laser pulses was ~5 nsec. The laser light entered an atomic beam apparatus normal to the flow of the cesium atoms. The cesium emanated from a conventional effusive oven source, and the atomic beam flux was monitored with a hot wire detector.

The laser and atomic beam intersected between parallel electric field plates having a 1 cm separation. One plate was at ground potential, the other was at a variable potential of up to 20 kV. A typical electric field strength in the interaction region was 15 kV/cm. Fluorescence was observed through an aperture in the field plate, normal to the plane formed by the laser and atomic beam (see fig. 2), using an RCA C31034 PMT. The aperture was covered with wire mesh to minimize distortion of the electric field.

In order to compare the excitation rates of the 7P and 6D levels, we monitored the fluorescence originating from each of these levels following a laser pulse of appropriate wavelength. The fluorescent intensity

provided a measure of the relative populations of the two levels.

The signal observed was the 8521 Å 6P → 6S cascade transition (see fig. 1). An interference filter centered at this wavelength was placed in front of the PMT. This transition was chosen because it is a common channel for both the 7P and 6D decay. Thus, no correction is needed for detection efficiency at different wavelengths. Also, the C31034 PMT is not sensitive to the 6D → 6P decay wavelength, 9200 Å.

The output of the PMT was sent to a gated scalar which recorded the total number of fluorescent counts observed. The number of counts was compared for 4555 Å (6S ↮ 7P) excitation and for 4417 Å (6S → 6D) excitation with a fixed electric field strength. An important aspect of this method of determining $a(6D, 7P)$ is that almost all the measurements are ratios: the ratio of fluorescent counts, the ratio of laser power at 4555 Å and 4417 Å, and the ratio of atomic beam fluxes. This eliminates many potential difficulties caused by experimental geometry and absolute detection efficiencies.

Our determination of the Stark mixing coefficient gives:

$$a(6D, 7P) = 0.7(3) \times 10^{-3} \, \mathcal{E}$$

where $\mathcal{E}$ is in kV/cm. The error is the limit of error from five independent determinations. The chief sources of error were uncertainty in the atomic beam flux, and difficulty in monitoring the laser power. The statistical error was negligible. It should be possible to reduce substantially the systematic errors. The present result, however, provides the first experimental determination of this matrix element. The value calculated from the Bates and Damgaard method is $a(6D, 7P) = 1.4 \times 10^{-3} \, \mathcal{E}$.

Electric quadrupole transitions directly between the 6S and 6D levels were also observed. The excitation rate of the quadrupole transition was measured and from this the emission rate was deduced. The value, $A(6D - 6S) = 63(30) \, s^{-1}$, is in general agreement with previous determinations [7, 8]. We are currently measuring the rate more accurately.

The Stark mixing method has also been used to determine lifetimes of a number of inaccessible states. Again, let us consider the $6\,^2D_{5/2}$ state in cesium. As before, we selectively populate this state with a pulse of light at 4417 Å in an electric field of 15 kV/cm, and monitor the 8521 Å cascade fluorescence.
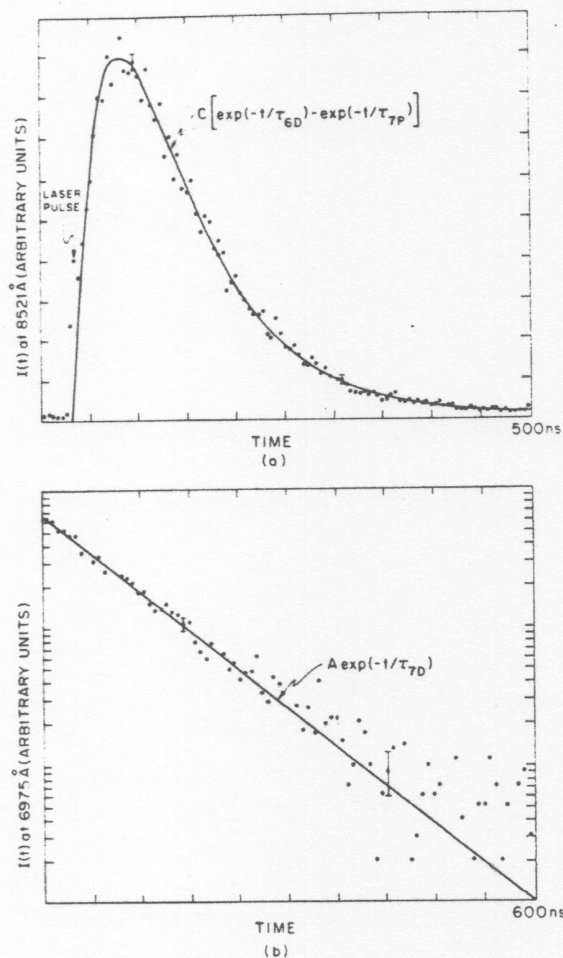
Fig. 3. (a) Data and least-squares fit for cascade decay from $6\,^2D_{5/2}$ level. Linear plot. (b) Data and least-squares fit for decay of $7\,^2D_{5/2}$ level. Log plot with background subtracted.

We are now interested in the time dependence of the fluorescence for lifetime determinations. Since we are monitoring the second leg of the cascade, the time dependence of the fluorescence is not a simple exponential. However, it can easily be shown that if the laser excitation pulse is short, the population of the $6\,^2P_{3/2}$ state in time and the fluorescent intensity at 8521 Å are given by

$$N_{6P}(t) = \frac{N_0 \, \tau_{6P}}{\tau_{6P} - \tau_{6D}} \left\{ \exp[-t/\tau_{6P}] - \exp[-t/\tau_{6D}] \right\} . \tag{5}$$

To measure the intensity as a function of time,

$I(t)$, we used a single photon-counting technique [9, 10] in which the probability or arrival of the first-observed fluorescent photon vs. time was determined. The distribution of arrival times is directly proportional to $I(t)$ in the limit of a low count rate. In order to measure the distribution of arrival times, a time-to-amplitude converter (TAC) was used to determine the delay between the initial laser pulse and the first fluorescent pulse from the PMT after laser excitation. The output of the TAC was sent to a computer configured to act like a multichannel analyzer (MCA) (see fig. 2).

The data for the $6\,^2D_{5/2}$ decay, shown in fig. 3a, closely follows the functional form of eq. (5). The values for the lifetimes were found by a least-square fit. The results are $(6\,^2P_{3/2}) = 31(1)$ ns, $\tau(6\,^2D_{5/2}) = 64(2)$ ns.

We have also determined the lifetime of the $7\,^2D_{5/2}$ state. The transition monitored was $7\,^2D_{5/2} \rightarrow 6\,^2P_{3/2}$ at 6975 Å. The pulsed dye laser was tuned to $6\,^2S_{1/2} \rightarrow 7\,^2D_{5/2}$, 3836 Å, and $I(t)$ was measured as before. The data are given in fig. 3b. Since in this case we monitor direct decay from the 7D level, we obtain a simple exponential characterized by the lifetime of the $7\,^2D_{5/2}$ state. The result is $\tau(7\,^2D_{5/2}) = 92.5(15)$ ns.

It should be noted that the lifetimes measured are those of the perturbed levels. In the present case, however, there is essentially no difference between the lifetimes of the perturbed and unperturbed states.

We have shown the applicability of the Stark mixing technique to the measurement of lifetimes of dipole-inaccessible states and to determining mixing coefficients in cesium. The technique can be extended to populate selectively states of other atomic systems and it opens the possibility of investigating states which are otherwise inaccessible.

References

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Ionization of highly excited hydrogenic atoms

The problem is to find the ionization rate of a highly excited hydrogenic atom in an applied electric field. The problem is important because ionization may limit the production of high-n hydrogenic atoms (i.e, highly excited alkalis), and because many be of interest to measure the rate and check the theory.

Reference: Bethe & Salpeter, p 235.

Schrödinger equation in an applied electric field is exactly soluble in parabolic coordinates.

Let's start by reviewing these

Parabolic coordinates:

Conic section: $r = \dfrac{r_0}{1 - \epsilon \cos\theta}$

$r = r_0 - \epsilon r \cos\theta = r_0 - \epsilon x$

$x^2 + y^2 = r_0^2 - 2r_0 \epsilon x + \epsilon^2 x^2$

$y^2 = r_0^2 - 2r_0 \epsilon x + (\epsilon^2 - 1) x^2$

For ellipse, take $\epsilon = \pm 1$ $x = \pm\left(\dfrac{r_0}{2} + \dfrac{z^2}{2r_0}\right)$
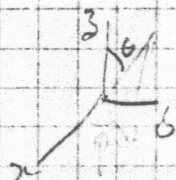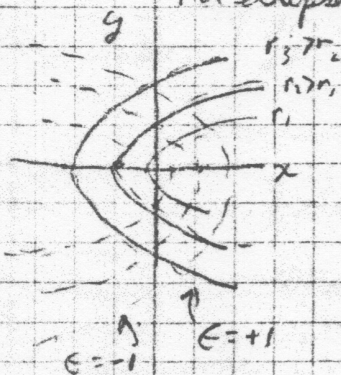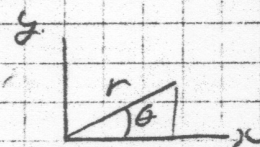
The parabolas are described by r_0, ϵ

$r - \epsilon r \cos\theta = r_0$

Let parabolas be centered around y axis,

$r - \epsilon z = r_0$

$\epsilon = \pm 1, \quad r_0 = r \mp z$

$\epsilon = -1 \qquad \epsilon = +1$

③ Started weekly discussions → MW 10:45
on high n studies
2 Oct, 7 Oct, 9 Oct, 16 Oct,
21 Oct, 1 Nov, 11 Nov, 10 Dec   30 Sept 74

High N stuff

Ref. Bethe & Salpeter

Hydrogen problem

Review of Hydrogenic Eif. (ref. B&S chap 1

$$\nabla^2 u + \frac{2m}{\hbar^2}(E - V) u = 0$$

$$V = -\frac{Ze^2}{r}$$

$$\nabla^2 u + \frac{2m}{\hbar^2}\left(E + \frac{Ze^2}{r}\right) u = 0 \xrightarrow{A.U.} \nabla_a^2 u + a_0^2\left(\frac{2E}{\hbar^2/m} + \frac{2Z m e^2/\hbar^2}{r}\right) u = 0$$

$$\frac{\hbar^2}{me^2} \equiv a_0 \quad \text{Bohr radius}$$

$$\frac{me^4}{\hbar^2} = \frac{e^2}{a_0} = \text{unit energy}$$
"Hartree"

$$\nabla^2 u + 2\left(E + \frac{Z}{r}\right) u = 0$$

digression → for other pot
↓

eg.

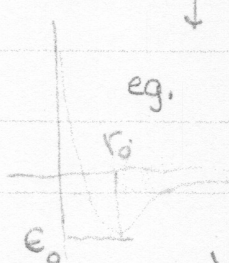$$\nabla^2 u + \frac{2\mu\epsilon_0}{\hbar^2}\left[\frac{E}{\epsilon_0} + f\left(\frac{r}{r_0}\right)\right] u = 0$$

$$\nabla_x^2 = r_0^2 \nabla^2$$

$$\nabla_x^2 u + \underbrace{\frac{2\mu\epsilon_0 r_0^2}{\hbar^2}}_{\beta}\left[E_r + f\left(\frac{r}{r_0}\right)\right] u = 0$$

$$V(r) = \epsilon_0 f(x)$$
$$x = \frac{r}{r_0}$$

mass M
reduced mass μ

$\beta = 2$ for hydrogen

④ Letter to Dan
- sweeping electric field
- using pulsed ionization
- Computer moved to bldg 26
- Rick Freeman working with us

Cambridge, Mass
5 March 1975

Dear Dan,

    As you can tell from the manuscript many mysteries still remain concerning the expt. This selection rule business that is discussed in the paper ($\vec{J}$ vs $\vec{F}$ rules depending on how long atom lives in $3^2 P_{3/2}$ intermediate state [3 nsec hyperfine period]) is certainly not well understood on this side of the Atlantic. Looking back at Franken's treatment of pulsed excitation I am somewhat convinced that it is at best incomplete as that following the treatment one cannot explain the observed phenomenon. Can you suggest a ref. that might contain a detailed calc. treatment for pulsed excitation? We also tried this out using the $3^2 P_{1/2}$ intermediate state and if the laser were coincident in time we got the same dramatic result. This is not to be expected in that the hyperfine period of the $P_{1/2}$ state is 3 times shorter than the $P_{3/2}$, one might expect some effect but not as dramatic.

    We spent some time making stark shift measurements and rapidly came to the concl. that an estimate about the order of mag. of

Stark shift vs field is needed. Rick redid
some of the calc. and found some errors.
It appears that if one applies a D.C. field
(laser tuned say to an S state) that
the nearby D state is stark shifted
into resonance with the laser before the
S state moves a laser linewidth ($\sim \frac{3}{4} cm^{-1}$).
This work was done where n was about 30.
Some interesting results were obtained for
lower n (~ 25). For these the laser was
tuned a little red of the S state ...

200 V/cm
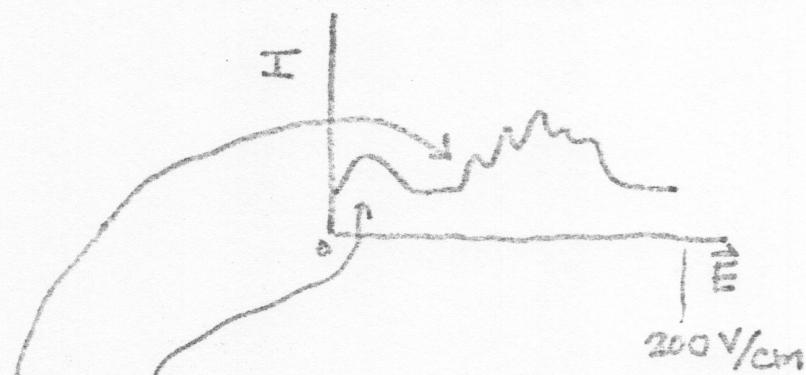
tentative identification is that the 1st large
bump is due to the s state stark shifted
red thru the laser line and the broad
multibumped blob is due to the D state
stark shifted into resonance. The bumps
appear (?) to be different m sublevels
in that their strength depends
on the polarization of the laser (i.e. can
get rid of 1st D bump by circular. polar.
lasers.) Of course this is all speculative
but $\tilde{g}$ we are soon to be looking at this problem.

One improvement that can be made right away
I believe is narrowbanding the blue laser.

I wanted to tell you more about some
other newsfronts but my hand is getting
tired so I'll mention them very briefly.
The delay problem (funct. of ion, field) in
the pulsed case is better documented
and I believe all effects here are .
instrumental. In the D.C. case we still
need to look more closely. Some crude
calc. have been made for R.F. transitions
between adjacent levels (for Hydrogen)
and the dipole moments are huge so the
R.F. powers need is miniscule (maybe
nanowatts). QUESTION: If applied
E&M field has bandwidth less than
natural linewidth characterizing
transition what should one use for
$\rho(\sigma)$ [spectral power density]?
(ref. please). Still no good way to
~~pulsed~~ integrate although progress
has been made on electronic front.
Vacuum apparatus working well and
it loves LN_2 (causes penning guage
to go off scale). Much more, will save
for another letter.

Mike

P.S. computer is in Bldg 26 & it is running!
cost for move ~$100. (service call.) Lab

⑤ Pulsed ionization of Na
- Started before Rick joined group
- Rick contributed as experiment started to be operational

Stark Ionization of High-Lying States of Sodium*†

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(Received 14 April 1975)

By using stepwise excitation in an atomic beam, we have excited slow-moving atoms to pure high-lying quantum states at densities low enough to avoid loss by collision. The atoms were detected with high efficiency by Stark ionization. Results are presented of a study of the threshold field for ionization for s states of sodium with principal quantum number n from 26 to 37.

We report the production of highly excited sodium atoms in pure quantum states, and observations on their ionization threshold in an electric field. There has recently been a growing interest in the properties of high-lying states of atoms near the ionization limit. These states, which are essentially hydrogenic, are characterized by very long lifetimes, large polarizabilities and

Stark shifts, low binding energies ($[2n^2]^{-1}$ a.u.) and large radii (proportional to n^2 a.u.). They are so fragile and large that until recently the only data on atoms in pure high-lying quantum states have come from radioastronomical observations of the recombination lines in interstellar hydrogen.[1] Measurements on lifetimes in sodium s and d states with n up to 13 using laser excitation in a gas cell have been reported by Gallagher, Edelstein, and Hill.[2] Bayfield and Koch[3] created a beam of hydrogen with n in the range of 63 to 69 by charge exchange of a fast proton beam with xenon. Cook, West, Dunning, and Stebbings[4] have excited high-lying levels of rare gases. In our work, we have used an atomic beam to reduce collisional ionization which can limit the levels observable in a cell. By using tunable lasers, we have been able to excite selectively individual quantum states, in contrast to collisional techniques which generally excite a wide distribution of states.

Sodium atoms in an atomic beam were excited stepwise by two pulsed dye lasers[5] pumped by a common nitrogen laser. The first dye laser saturated the D_1 line (5890 Å), creating a large population in the $3^2P_{3/2}$ state, while the second (~ 4100 Å) caused transitions from the p state to high-lying s or d states. The lasers both had a spectral width of 1 cm^{-1}, a pulse length of 5 nsec, and a peak power of 1 kW.

We first detected atoms in high-lying levels by observing resonance fluorescence at the ~ 4100 Å line from the high level. A photomultiplier tube detected the light emitted at right angles to both the atomic beam and the incident lasers. We were able to observe s and d states from $n=15$ to about 30, but not higher levels because of rapidly decreasing signal strength. The intensity loss was due to two factors, each varying as n^{-3}: the decreasing transition moment, and the loss of signal which occurs when the radiative lifetime becomes longer than the time for an atom to pass out of the observation region, about 10 μsec.

For $n > 23$ the excited atoms were detected by direct ionization in an applied field. In addition to avoiding the problem of signal loss due to long radiative lifetime, the method provides close to 100% detection efficiency and very low background. The laser beams intersected the atomic beam between electric field plates. A pulsed ionizing electric field was applied after laser excitation and the resulting ions were observed with a channel electron multiplier. As expected, for reasons explained below, the approximate ion-ization field for a state with principal quantum number n was $[16n^4]^{-1}$ a.u. (386 V/cm for $n=30$). For a typical atomic-beam density of 10^8 cm^{-3} and an interaction region of 0.02 cm^3, we observed the equivalent of approximately 10^4 ions/ pulse for $n=30$.

To confirm the identity of excited levels as s or d, we used the fact that optical selection rules for stepwise two-photon processes are strongly affected by nuclear coupling in the intermediate state. The electron-nuclear hyperfine interaction mixes states of different values of m_J and m_I (where $\hbar J$ and $\hbar I$ are the electronic and nuclear angular momentum, respectively) to create a state of total angular momentum $\hbar F$. This tends to scramble together states of all possible values of m_J and m_I satisfying $m_J + m_I = m_F$. The result is that cw absorption of two photons circularly polarized in the same sense gives rise to both $s \to p \to s$ and $s \to p \to d$ transitions. In contrast, if such photons are absorbed successively in a time interval which is short compared to the hyperfine period of the intermediate state, the electron does not have sufficient time to precess about the nucleus to a new spatial orientation before it absorbs the second photon. In this situation the electric dipole selection rules for the two-step process are the same as those for an atom with no nuclear spin. For such an atom $s \to p \to d$ transitions are still allowed, but $s \to p \to s$ transitions are forbidden.

In our experiment we found, consequently, that when the lasers were circularly polarized in the same sense, the population of the s levels was a sensitive function of the delay between the pulses. When the two pulses excited the sodium in rapid succession (≤ 3 nsec delay, a time short compared to the hyperfine period in the $3P_{3/2}$ state) the transition rate to the s state was radically suppressed, whereas the rate to a d state was essentially unchanged. When the second pulse was delayed approximately 8 nsec, the s-state transition rate became far less affected by circular polarization of the lasers.

The energy levels of sodium have the form $W_n^0 = [2(n-\Delta)^2]^{-1} = [2(n^*)^2]^{-1}$, where the quantum defect Δ depends on the orbital angular momentum. The high-lying levels form a slowly varying pattern shown in Fig. 1. The term spacing is $\Delta W_n^0 = (n^*)^{-3}$. For $n=30$, $\Delta W_{30}^0 = 9.3$ cm^{-1}. The states ns and $(n-1)d$ form a pair separated by about 0.35 of the term spacing. Individual levels were resolved for $n=23$ to $n=38$. The laser frequency was determined by a Spex 14018 monochromator
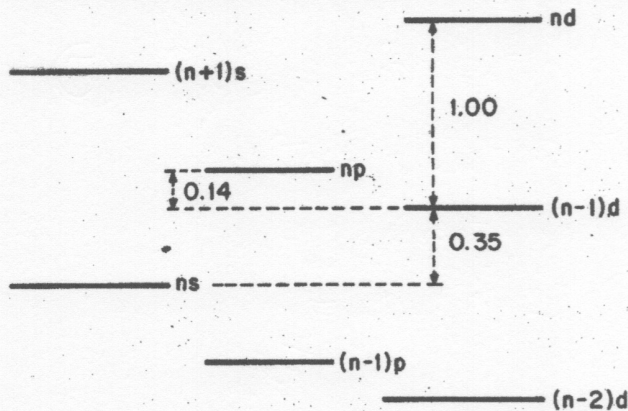
FIG. 1. Energy-level diagram showing relative positions of ns, np, and nd levels. Energy differences are in units of $\Delta W_n{}^0 = |W_n{}^0 - W_{n-1}{}^0|$.

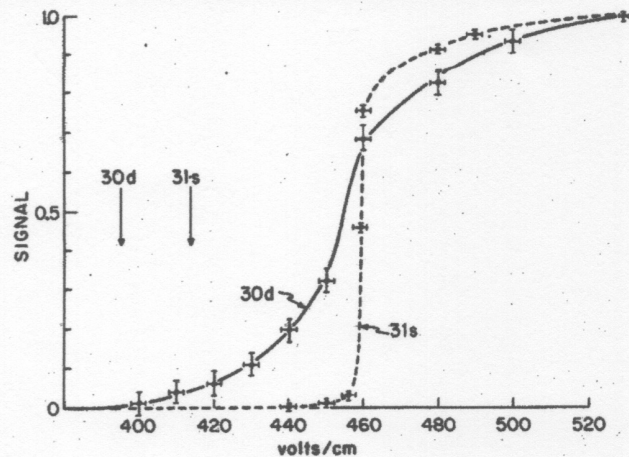FIG. 2. Ionization signal as a function of the pulsed ionizing electric field. The onset of ionization is broader for the 30d level than for the 31s. This is due to the Stark splitting in the ionizing field of the 30d. The 31s remains a single-shifted level. The vertical arrows indicate the critical ionization field of the unshifted levels from Eq. (1).

to an accuracy of about 1 cm^{-1} and the quantum defects for all n were determined to be $\Delta(s) = 1.35(4)$, $\Delta(d) = 0.00(4)$, in good agreement with the values at lower levels.[6] For $n > 38$ the laser resolution was insufficient to resolve the splitting between adjacent ns and $(n-1)d$ levels, though individual pairs could be resolved to $n = 50$. With a narrower laser line, individual states should be resolved for much higher values of n.

By studying the ionization probability as a function of electric field for different levels we have been able to observe the Stark shift at the onset of ionization. This problem has evoked continued interest over the years because it represents the extreme case of distortion of a free atom by an electric field.[7] Consistent results on the ground state of hydrogen have been obtained by several authors.[8] Huschfelder and Curtiss[9] have considered the $n = 5$ case, in detail, but to our knowledge a complete theoretical treatment for high-n states is lacking.

The alkali system differs from hydrogen in the important respect that the degeneracy of the low-angular-momentum states is broken, regardless of n. This presents a useful experimental advantage, for it allows selection of a single member of an otherwise n-fold-degenerate manifold of angular momentum states. This is illustrated in the data on the ionization threshold shown in Fig. 2. The 31s-state curve exhibits the abrupt onset of ionization that characterizes a single energy level; the width of the curve for the 30d state is attributed to its multiplicity.

The Coulombic potential in an applied field, $V = -1/r - Ez$ (a.u.) has a maximum $V_{max} = -2\sqrt{E}$.

The ionization threshold for high-n states is well defined because the range of fields for which tunneling would be observed is extremely narrow. Ionization occurs when $V_{max} = W_n$, where W_n is the term energy. If we neglect the Stark effect, then $W_n = -[2(n^*)^2]^{-1}$, and the threshold field would be

$$E_n{}^0 = [16(n^*)^4]^{-1}. \tag{1}$$

We have measured the critical ionization fields, E_n, for s levels with $n = 26$–37. The results of Eq. (1) and the data are compared in Fig. 3. E_n is consistently larger than $E_n{}^0$. This difference arises because of the Stark shift.

A simplified analysis of the critical-field data can be made by assuming the wave function remains essentially Coulombic at the ionization threshold. We can write the term energy at threshold as

$$W_n = W_n{}^0 + \delta W_n, \tag{2}$$

where $W_n{}^0 = -[2(n^*)^2]^{-1}$, and δW_n is taken as the Stark shift at ionization. Values for δW_n may be extracted from the data in Fig. 3 using the relation

$$\delta W_n = -2\sqrt{E_n} - W_n{}^0. \tag{3}$$

An estimate of the Stark shift can be obtained by assuming that the ns state mixes chiefly with the np state. With this two-level model we calculate values for the Stark shift at ionization which are in general agreement with the values
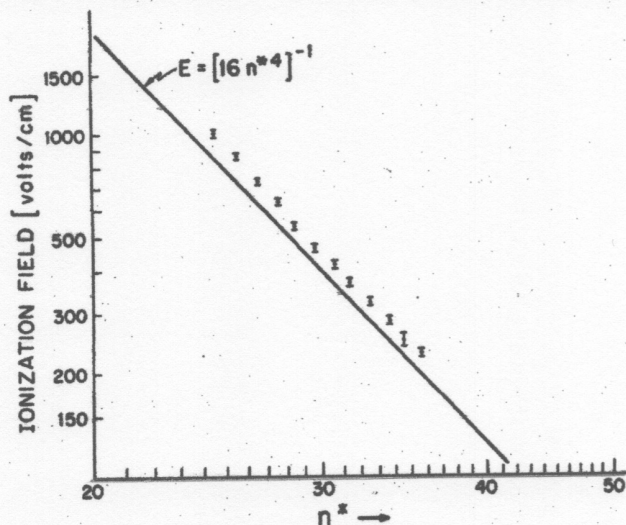
FIG. 3. Log-log plot of critical ionization field versus n^* for $n = 26$–37. The straight line represents the predictions of Eq. (1) for no Stark shift. The actual critical field is consistently larger than the prediction. This is a result of the Stark shift at ionization.

extracted from the data, but are typically 30% low. For example, at $n = 30$, $\delta W_n = 5.6 \times 10^{-5}$ a.u. from Eq. (3), and 4.0×10^{-5} a.u. from the two-level analysis. More precise measurements and more realistic calculations which consider the contribution to the Stark shift of all important states are in progress.

The technique described here should be useful for studying a variety of other problems involving highly excited atoms. In addition to questions of polarizabilities and tunneling phenomena, there is interest in the behavior of atoms when the magnetic energy dominates the Coulomb energy[10] and in photoionization and other radiation phenomena. The technique of high-level excitation and ionization provides a selective way to detect excited atoms with very high efficiency and virtually no background. It may also have useful applications in a photon detector.

We would like to thank Myron Zimmerman for assistance in carrying out this work.

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Clarendon Laboratory
July 11

Dear Mike,

Congratulations to you and to Myron for obtaining such interesting results on the Stark shift. Narrowing the laser appears to have payed major dividends. The computer plot is certainly suggestive of the physics of the situation, but it looks like some more work will be needed to give definite assignments to all the peaks. The wide peaks are suggestive of the overlap between positive and negative Stark components- the plot shows one at 400V. which looks very much like the data- but the intermediate weak peaks suggest that perhaps some distant terms not included in the computer plot are playing a role.

The new collimators should be on the way very shortly. I hope that these will help you recover some of the power lost in narrowing the laser. The delay was due to difficulty in removing the old coatings, baked MgF. It turned out that nothing would touch it, and the lenses had to go back to the factory for repolishing. However, they are now properly coated- a fairly broadband coating centered at 4100A, and there should be no further delay. The new coatings are unbaked MgF. The coatings are reasonably rugged- they can be wiped with a cloth quite safely- but not as hard as the usual coatings.

A reference on Stark ionization which you will find very informative is Bailey, Hiskes and Riviere, Nuclear Fusion 5,41 (1965). They extend the Rice and Good method to n=25, and present the results in useful plots. The Rice & Good method should apply to Na for orbits with m greater than two or three, for these orbits are essentially non-penetrating. For lower values of m we shall have to do some work- perhaps we can use the Rice-Good method with alkali wave functions in the spirit of the Bates Damgard method.

I agree with you that measuring the Stark ionization rates would be extremely interesting, and I am sure that there are many possibilities. An important requisite is the ability to work with a single Stark component, which I think you have already shown how to do by your Stark excitation experiment. According to Bailey et al the ionization rate for n=15 low-lying Stark level varies from $.7 \times 10^6$/s to 6×10^8/s as the field varies between 9.8 and 11.0kv/cm. The Lanczos method gives rates about 10X smaller, so it should be easy to distinguish the two methods. (Incidentally, I have some translations of Lanczos'paper- given to me by Riviere- which I will bring back) We will probably need to make some effort to get rid of background ionization in the system.

I have been trying to find a simple argument to obtain the 'critical' field for ionization, analagous to the expression $1/16n^4$ for an unperturbed Coulombic system in an s state. It is easy to get an expression taking into account the first order Stark effect, but my second order solution doesn't make sense. I've enclosed a note- perhaps you can get somewhere with the problem.

Say hello to everyone for me.

Regards,

Dan