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ATOMIC BEAM STUDY OF THE STARK EFFECT
IN THE CESIUM AND RUBIDIUM D LINES

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ABSTRACT

Details of a new atomic beam method for the study of the Stark effect in optical transition are presented. The method is then applied to a study of the transitions $6^2p_{1/2,3/2} \leftrightarrow 6^2s_{1/2}$ in cesium and $5^2p_{1/2,3/2} \leftrightarrow 5^2s_{1/2}$ in rubidium. The splitting by the electric field of the $p_{3/2}$ level into two levels is observed. It is shown that the characterization of the Stark effect in the ^2p levels by a simple scalar and tensor polarizability does not hold. Fine-structure effects giving rise to differences of the $^2p_{1/2}$ and $^2p_{3/2}$ radial functions are sufficiently strong so that the Stark effect of the ^2p level must be expressed in terms of three parameters. If the polarizability $\alpha(n^2p_Jm_J)$ is defined by the relation $\Delta W(n^2p_Jm_J) = -\frac{E^2}{2}\alpha(n^2p_Jm_J)$, where E is the electric field and ΔW the induced energy shift, then the following values of the polarizabilities are deduced. For cesium, $\alpha(6^2p_{1/2}) = 187(29) \times 10^{-24}$ cm^3; $\alpha(6^2p_{3/2} \pm \frac{3}{2}) = 196(30) \times 10^{-24}$ cm^3; and $\alpha(6^2p_{3/2} \pm \frac{1}{2}) = 273(42) \times 10^{-24}$ cm^3. For rubidium, $\alpha(5^2p_{1/2}) = 112(17) \times 10^{-24}$ cm^3; $\alpha(5^2p_{3/2} \pm \frac{3}{2}) = 102(15) \times 10^{-24}$ cm^3; and $\alpha(5^2p_{3/2} \pm \frac{1}{2}) = 148(23) \times 10^{-24}$ cm^3. The polarizabilities are compared with results deduced from Stone's recent oscillator strength calculations for cesium and with values deduced from the method of Bates and Damgaard.

INTRODUCTION

Recently, there has been a considerable revival of interest in the study of the Stark effect. New theoretical techniques have been developed for studying the infinite sums appearing in the expressions for the Stark shift.[1] From the experimental point of view, new techniques have been developed for observing small frequency shifts in hyperfine and Zeeman transitions.[2]

In this paper, details are given for an atomic beam technique for studying the Stark effect in optical transitions. The method is then applied to measurements of the Stark effect in the D line transitions in both cesium and rubidium. A detailed theory of the Stark effect in these states is developed with which the experimental results are compared. These results are of interest as a test of a recent calculation of cesium oscillator strengths. They also serve as an important preliminary to the measurement of the cesium and rubidium isotope shifts in the D lines.[3]

Surprisingly, there seems to have been no Stark-effect work on the cesium and rubidium D lines. Measurements have been made on the 6p-5s transitions in rubidium and the 7p-6s transitions in cesium.[4] However, in this work the splitting of $p_{3/2}$ into the predicted doublet was not observed, and is not useful as a test of the theory of the Stark effect.

THEORY

The perturbation of an energy level by an external electric field E is described by the Hamiltonian

$$\mathcal{H}' = - \underset{\sim}{p} \cdot \underset{\sim}{E} \quad , \tag{1}$$

where $\underset{\sim}{p}$ is the induced dipole moment and is given by $\underset{\sim}{p} = - e \sum_i \underset{\sim}{r}_i$, $\underset{\sim}{r}_i$ being the position vector of the ith electron. It is assumed that polarization of the nucleus is negligible. Specializing to an alkali for which we neglect perturbation

of electrons in closed shells, then $\underset{\sim}{p} = -e\underset{\sim}{r}$, $\underset{\sim}{r}$ being the position vector of the valence electron.

If the total Hamiltonian is denoted by $\mathcal{H}$, then $\mathcal{H} = \mathcal{H}_o + \mathcal{H}'$, and we ask what terms it is appropriate to consider as part of $\mathcal{H}_o$ for the states under investigation here. For the $^2s_{1/2}$ and $^2p_{1/2}$ states of rubidium and cesium, we include in $\mathcal{H}_o$ all terms through the hyperfine structure. More specifically, $\mathcal{H}_o$ includes the central field, the spin-orbit effect, and the hyperfine-structure operator. The inclusion of hyperfine structure is important for $s_{1/2}$, since the Stark shifts induced are of the same order as the hyperfine structure. For $p_{1/2}$, the Stark shift is considerably larger (by about an order of magnitude) than the hfs, but it is no inconvenience to include hfs in the zeroth-order Hamiltonian. For $^2p_{3/2}$, the hfs is an order of magnitude smaller than the Doppler width of the lamp, and almost two orders of magnitude smaller than the induced shifts. Accordingly, hfs is neglected for $p_{3/2}$.

A. Application to $^2s_{1/2}$ and $^2p_{1/2}$

It is well known that for states of well-defined parity the Hamiltonian Eq. (1) produces no first-order shift. Hence we can write the second-order shift due to Eq. (1) as

$$\Delta W(J = 1/2) = \sum_{\psi} \frac{|\langle\psi|e\underset{\sim}{r}\cdot\underset{\sim}{E}|n^2\ell_{1/2}IFm_F\rangle|^2}{\Delta E(\psi,\frac{1}{2})} \quad . \tag{2}$$

The electric field in this experiment is parallel to the fields and field gradients in the A and B magnets and may be taken along the z axis. It is convenient in evaluating Eq. (2) to employ spherical tensor methods. Therefore, we write

$$e\underset{\sim}{r}\cdot\underset{\sim}{E} = (4\pi/3)^{1/2}\, eErY_1^0(\theta,\varphi) \quad , \tag{3}$$

where Y_1^0 is the zeroth component of the spherical harmonic of rank one. The form of Eq. (3) limits the states ψ to those having the same m_F as the initial state. Thus, we rewrite Eq. (2) as

$$\Delta W(J = 1/2) = \frac{4\pi}{3} e^2E^2 \sum_{\substack{n',\ell',J' \\ F'}} \frac{|\langle n'^2\ell'_{J'}, IF'm_F|rY_1^0|n^2\ell_{1/2}IFm_F\rangle|^2}{\Delta E(\psi,\frac{1}{2})} \quad . \tag{4}$$

If we use standard tensor identities relating 3j symbols and the Biedenharn-Elliott sum rule,[5] it can be shown that Eq. (4) is independent of the quantum numbers F and m_F, provided only that the hyperfine energy of the states ψ is neglected in the denominator of Eq. (4). Under this circumstance, Eq. (4) can be written

$$\Delta W(J = 1/2) = \frac{2\pi}{3} e^2E^2 \sum_{n',\ell',J'} \frac{|\langle n'^2\ell'_{J'}\|rY_1\|n^2\ell_{1/2}\rangle|^2}{\Delta E(\psi_{n',\ell',J'};\frac{1}{2})} \quad . \tag{5}$$

For the case $n^2s_{1/2}$ this becomes

$$\Delta W(n^2s_{1/2}) = \frac{1}{9} e^2E^2 \sum_{n'} \left\{ \frac{|\langle n'^2p_{1/2}\|r\|n^2s_{1/2}\rangle|^2}{\Delta E(n'^2p_{1/2};n^2s_{1/2})} + \frac{2|\langle n'^2p_{3/2}\|r\|n^2s_{1/2}\rangle|^2}{\Delta E(n'^2p_{3/2};n^2s_{1/2})} \right\} , \tag{6}$$

and for $n^2p_{1/2}$ this becomes

$$\Delta W(n^2p_{1/2}) = \frac{1}{9} e^2E^2 \sum_{n'} \left\{ \frac{|\langle n'^2s_{1/2}\|r\|n^2p_{1/2}\rangle|^2}{\Delta E(n'^2s_{1/2};n^2p_{1/2})} + 2\,\frac{|\langle n'^2d_{3/2}\|r\|n^2p_{1/2}\rangle|^2}{\Delta E(n'^2d_{3/2};n^2p_{1/2})} \right\} . \tag{7}$$

The reduced matrix elements are related to integrals over radial wave functions in the usual way; i.e.,

$$\langle\psi_f\|r\|\psi_i\rangle = \int_0^\infty R_fR_i r dr \quad ,$$

where the radial part of the wave function is R/r. The square of this radial integral is proportional to the oscillator strength. Hence the study of the

Stark effect can be regarded as a method for the study of oscillator strengths or as a method for checking theoretical oscillator strengths.

B. Application to $^2p_{3/2}$

As discussed above, it is reasonable to neglect hyperfine structure for the $^2p_{3/2}$ states. Thus, the Stark perturbation takes the form

$$\Delta W(n^2p_{3/2}) = \frac{4\pi}{3} e^2E^2 \sum_{n',\ell',J'} \frac{|\langle n'^2\ell'_{J'}m_J|rY_1^0|n^2p_{3/2}m_J\rangle|^2}{\Delta E} \quad . \tag{8}$$

It follows that the splitting is proportional to $(m_J)^2$, so that states with the same absolute value of m_J remain degenerate under the action of the Stark field. Therefore, the $^2p_{3/2}$ energy level is split into two levels under the action of the Stark field, corresponding to $m_J = \pm 3/2$ and $m_J = \pm 1/2$. The evaluation of Eq. (8) leads to

$$\Delta W(n^2p_{3/2} \pm 3/2) = \frac{1}{25} e^2E^2 \sum_{n'} \left\{ \frac{|\langle n'd_{3/2}\|r\|np_{3/2}\rangle|^2}{\Delta E(n'd_{3/2};np_{3/2})} + 4 \frac{|\langle n'd_{5/2}\|r\|np_{3/2}\rangle|^2}{\Delta E(n'd_{5/2};np_{3/2})} \right\} , \tag{9}$$

$$\Delta W(n^2p_{3/2}\ m_J = \pm 1/2) = \frac{1}{225} e^2E^2 \sum_{n'} \left\{ \frac{|\langle n'd_{3/2}\|r\|np_{3/2}\rangle|^2}{\Delta E(n'd_{3/2};np_{3/2})} + 54 \frac{|\langle n'd_{5/2}\|r\|np_{3/2}\rangle|^2}{\Delta E(n'd_{5/2};np_{3/2})} + 50 \frac{|\langle n's_{1/2}\|r\|p_{3/2}\rangle|^2}{\Delta E(n's_{1/2};np_{3/2})} \right\} , \tag{10}$$

We now define polarizabilities (α) for each of the above energy levels according to the usual relation

$$\Delta W(n^2\ell_J m_J) = - \frac{E^2}{2} \alpha(n^2\ell_J m_J) \quad . \tag{11}$$

So far as it is possible to neglect differences in the radial wave functions for $n^2p_{1/2}$ and $n^2p_{3/2}$ and for $n'^2d_{3/2}$ and $n'^2d_{5/2}$, the following simple relation among the polarizabilities holds:

$$\alpha(^2p_{1/2}) = \frac{1}{2}\,[\alpha(^2p_{3/2}\, m_J = \pm 3/2) + \alpha(^2p_{3/2}\, m_J = \pm 1/2)] \quad . \qquad (12)$$

Such a relation can be deduced more directly from a decomposition of the Stark operator into scalar and tensor parts.[6] As we will see, however, such a relation does not hold for the cesium 6p state and the rubidium 5p states. Fine-structure effects are appreciable, and three parameters are needed to characterize the Stark effect in each of these levels.

EXPERIMENTAL METHOD

The method used here is that outlined by two of the authors in a recent letter.[3] The apparatus employed is a conventional atomic beam machine with flop-in magnet geometry. The C region consists of a pair of electric field plates, with a 0.036-in. gap, capable of sustaining large electric fields. The space between the plates is illuminated by filtered resonance radiation from a Varian X49-609 spectral lamp (see Fig. 1). For the cesium work, a lamp filled with ^{133}Cs was employed; for rubidium, a lamp of isotopically enriched ^{85}Rb was used. For both the D_1 and D_2 transitions in rubidium and in cesium the lamp output consists of a resolved doublet separated by the ground-state hyperfine structure (see Fig. 2). The excited-state hfs[7] is about 10% the ground state hfs for $p_{1/2}$ and even smaller for the $p_{3/2}$ state. It makes no essential difference in the discussion and is ignored.

Measurement of the Stark effect proceeds according to the following principles. It is well known that an atomic beam apparatus refocuses atoms that undergo the transition $m_J = +1/2 \longleftrightarrow m_J = -1/2$ in the C region. Consider now the action of a beam atom of the same isotopic species as the atom in the resonance lamp. At zero electric field the absorption lines of atoms in the beam coincide with the center of the emission lines in the lamp. Consequently, resonance absorption of photons takes place. In the subsequent

decay half of the atoms will undergo spin flip and will contribute to the flop-in signal at the detector. We describe the action of an electric field on the beam absorption lines for each of the two transition lines separately.

A. D_1 Transition ($^2p_{1/2} \leftrightarrow {}^2s_{1/2}$)

It is shown in the section on theory that to second order in the Stark perturbation all the hyperfine levels arising from a state with $J = 1/2$ are shifted by the same amount in the presence of an electric field. The relative shifts of the hyperfine levels and of the Zeeman sublevels can be deduced from recent measurements[2] to be smaller than the gross shift in the levels themselves by at least four orders of magnitude. Accordingly, an electric field serves to decrease in energy both the $p_{1/2}$ and $s_{1/2}$ levels and to decrease the net transition energy. When the transition energy is lowered by an amount equal to the emission linewidth of the lamp, the flop-in signal goes to zero. However, when the electric field is sufficiently large so as to shift the absorption lines by an amount equal to the ground-state hyperfine structure, a new overlap of the absorption lines with the emission lines of the lamp occurs (see Fig. 2) and another flop-in signal is observed. From the known ground-state hfs and the E^2 dependence characteristic of the Stark effect, the difference in the polarizabilities of the $p_{1/2}$ and $s_{1/2}$ states can be determined.

B. D_2 Transition ($^2p_{3/2} \leftrightarrow {}^2s_{1/2}$)

As pointed out in the section on theory, the hfs of the $p_{3/2}$ state is negligible. To this approximation the $p_{3/2}$ level is split into two levels corresponding to $m_J = \pm 3/2$ and $m_J = \pm 1/2$. As the difference in energy between each of these levels and the $^2s_{1/2}$ level is shifted by an amount equal to the ground-state hfs, new flop-in signals are observed (see Fig. 2). Hence, in addition to the zero field signal two new signals should be observed. From

a knowledge of the electric field at which these peaks occur and the ground-state hfs, the polarizabilities can be deduced.

DATA ANALYSIS AND RESULTS

A. Cesium

In Figs. 3A and 3B are shown the signals observed. The following qualitative features are of importance. First, there is only one flop-in peak observed with the D_1 optical line incident on the beam and two flop-in peaks with the D_2 optical line incident on the beam. This confirms the predictions made in the theory section. Second, the heights of the peaks are in agreement with theory; and third, the width of the peaks agrees with an independent measurement of the linewidth of the lamp. Perhaps the most important feature is the fact that the single $p_{1/2}$ lies significantly higher than the average of the two $p_{3/2}$ peaks. This is in violation of the prediction of Eq. (12) and must be taken as direct evidence for the importance of spin-orbit effects on the radial wave functions.

In order to understand the feature of the data we can use the well-known fact that the radial matrix elements involved are the same as those that determine the oscillator strengths for the transition. If spin-orbit effects modify the radial wave functions so as to invalidate relation (12), then this must show up in the oscillator strengths in the following way. Oscillator strengths from $p_{3/2}$ and $p_{1/2}$ to the same lower state must differ from the ratio of the statistical weights. Similarly, oscillator strengths from a common upper level to each of the p states must differ from the ratio of the statistical weights. Bearing on this point are recent calculations of the cesium oscillator strengths by Stone.[8] Stone's wave functions include spin-orbit effects and the resulting oscillator strengths for transitions to each of the 6p states from a common upper level which differ substantially from the appropriate weight factor. Using Stone's oscillator strengths and Eqs. (6), (7), (9), and (10), we have calculated the

polarizabilities for each of the observed levels. These are compared in Table I with the polarizabilities determined from our results. Our values for the polarizabilities of the 6p state are based on recent measurements of the ground-state polarizabilities by Bederson et al.[9] We also give in Table I results for the polarizabilities based on the method of Bates and Damgaard.[10] It is seen that the theoretical polarizabilities of both Stone and Bates and Damgaard are in excellent agreement with experiment.

B. Rubidium

In Figs. 4A and 4B are shown the rubidium signals with D_1 light and D_2 light incident, respectively. A lamp of separated ^{85}Rb was used and a beam of separated ^{85}Rb was employed so as to avoid complication from ^{87}Rb signals. Qualitatively, the results are similar to the cesium results. There are two features worth pointing out. First, the polarizabilities are smaller. Second, Eq. (12) is much better satisfied than in the case of cesium. This corresponds to the fact that the spin-orbit splitting in rubidium is much smaller than in cesium. In Table II the measured polarizabilities are compared with calculations based on the Bates-Damgaard method. Agreement here is also excellent.

The electric field was taken from the relation $E = V/d$. Our plates are sufficiently narrow relative to the length and height that this expression should hold to about 1%.

REFERENCES

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1. R. M. Sternheimer, Phys. Rev. 96, 951 (1954); C. Schwartz, Ann. Phys. (N.Y.) 6, 156 (1956).
2. R. D. Haun and J. R. Zacharias, Phys. Rev. 107, 107 (1957); E. Lipworth and P. G. H. Sandars, Phys. Rev. Letters 13, 14 (1964); B. Budick, S. Marcus, and R. Novick, Phys. Rev. 140, A1041 (1965); J. Blamont, Ann. Phys. (Paris) 2, 551 (1957).
3. R. Marrus and D. McColm, Phys. Rev. Letters 15, 813 (1965).
4. Y. T. Tao, Z. Physik 77, 307 (1932).
5. See, for example, A. R. Edmonds, Angular Momentum in Quantum Mechanics (Princeton University Press, Princeton, New Jersey, 1957).
6. P. G. H. Sandars and J. R. P. Angel, The Hyperfine Structure Stark Effect (to be submitted for publication).
7. H. Kleiman, J. Opt. Soc. Am. 52, 441 (1962).
8. P. M. Stone, Phys. Rev. 127, 1151 (1962).
9. A. Salop, E. Pollack, and B. Bederson, Phys. Rev. 124, 1431 (1961).
10. D. R. Bates and A. Damgaard, Phil. Trans. Roy. Soc. London A242, 101 (1949).

Table I. Cesium polarizabilities $\times 10^{24}$ cm^3.

	$\alpha(6s_{1/2})$	$\alpha(6p_{1/2})$	$\alpha(6p_{3/2} \pm 3/2)$	$\alpha(6p_{3/2} \pm 1/2)$
Stone	65	187	200	273
Bates and Damgaard	56	192	191	246
Measured[a]	52.5(6.5)	187(29)	196(30)	273(42)

a. The measured value for $\alpha(6s_{1/2})$ is taken from Ref. 9.

Table II. Rubidium polarizabilities $\times 10^{24}$ cm^3.

	$\alpha(5s_{1/2})$	$\alpha(5p_{1/2})$	$\alpha(5p_{3/2} \pm 3/2)$	$\alpha(5p_{3/2} \pm 1/2)$
Bates and Damgaard	46	116	108	151
Measured[a]	40(5)	112(17)	102(15)	148(23)

a. The measured value for $\alpha(5s_{1/2})$ is taken from Ref. 9.

FIGURE CAPTIONS

Fig. 1. Schematic diagram of atomic beam apparatus for studying Stark effect.

Fig. 2. Schematic diagram of energy levels. The lines A and B are both present in the lamp. At zero electric field the absorption lines 1 and 2 coincide with the emission line B. Signals are also observed at electric fields such that the lines 1 and 2 are made to resonate with the line A.

Fig. 3. A) Observed cesium signal with D_1 radiation only.

B) Observed cesium signal with D_2 radiation only.

Fig. 4. A) Observed rubidium signal with D_1 radiation only.

B) Observed rubidium signal with D_2 radiation only.

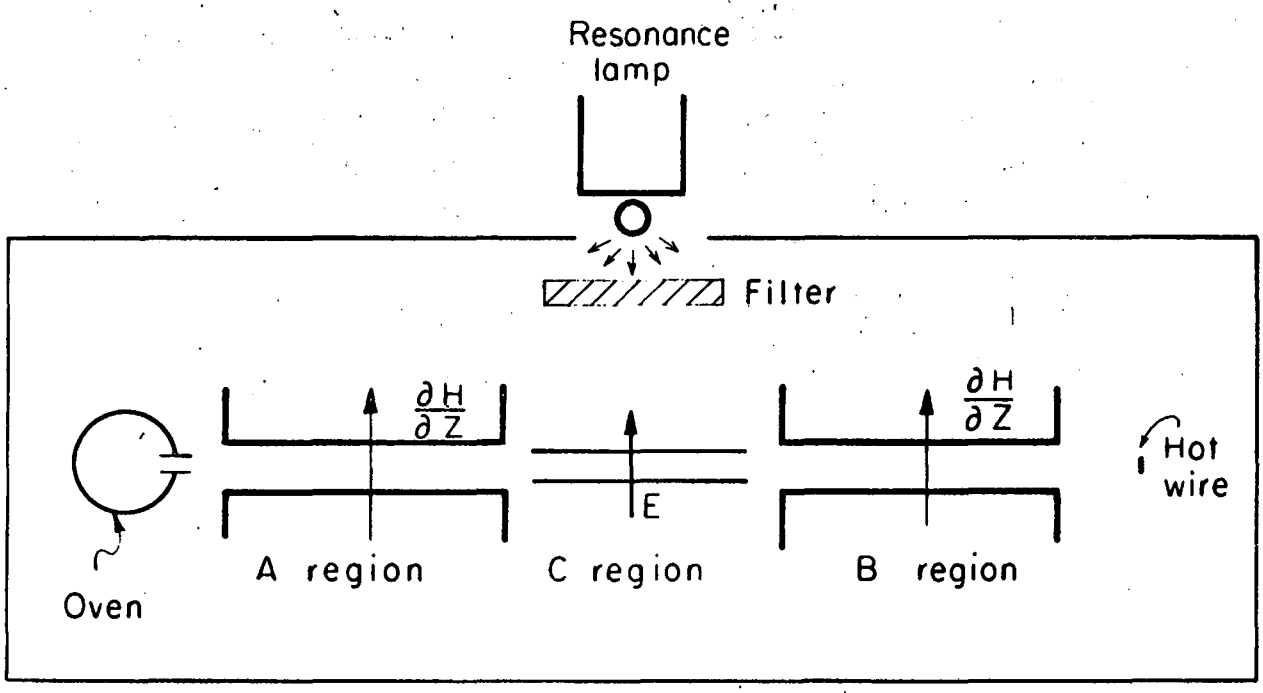

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Fig. 1

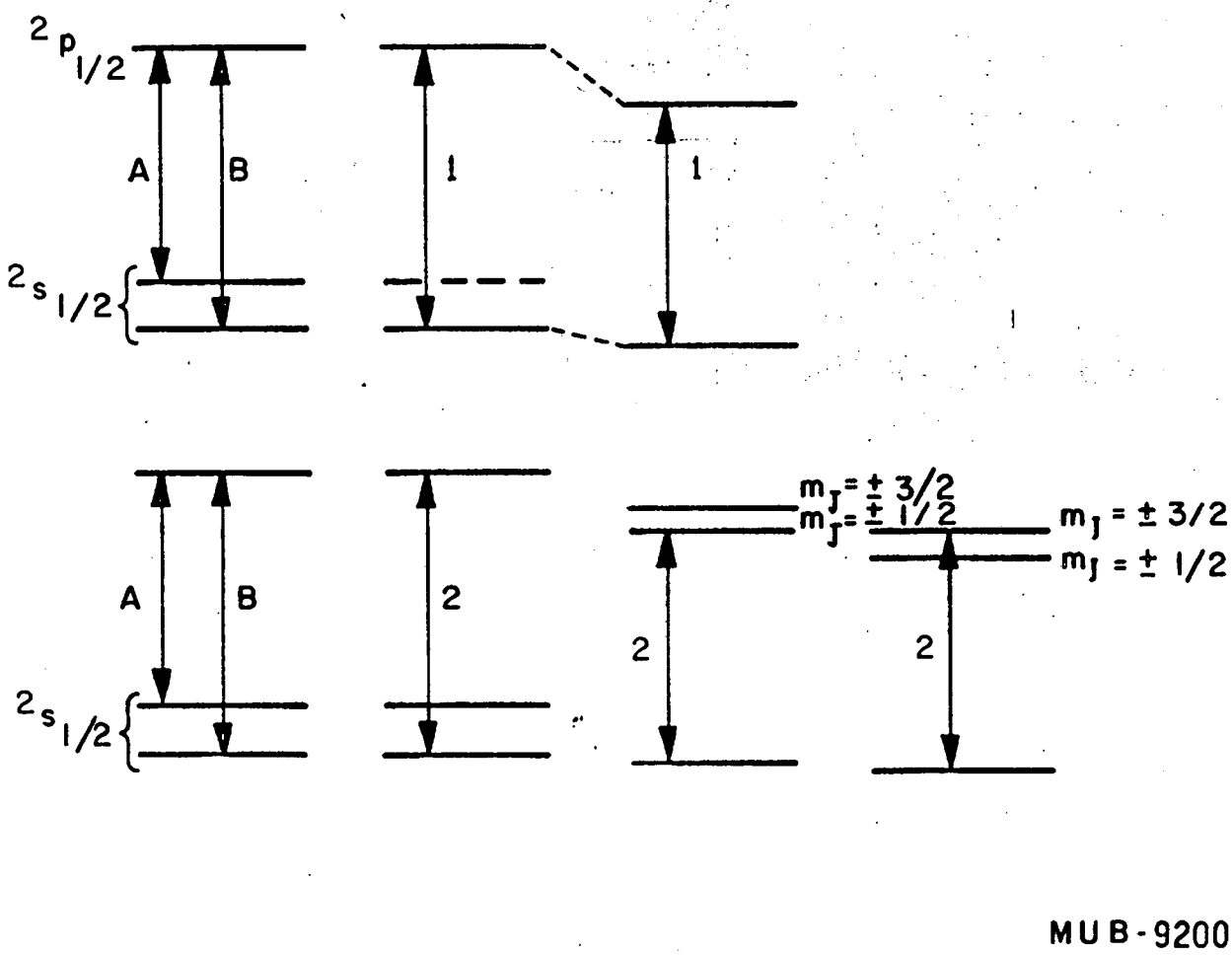
$^2P_{1/2}$
A
B
1
1
$^2S_{1/2}$
A
B
2
2
2
$m_J = \pm 3/2$
$m_J = \pm 1/2$
$m_J = \pm 3/2$
$m_J = \pm 1/2$
$^2S_{1/2}$
MUB-9200

Fig. 2

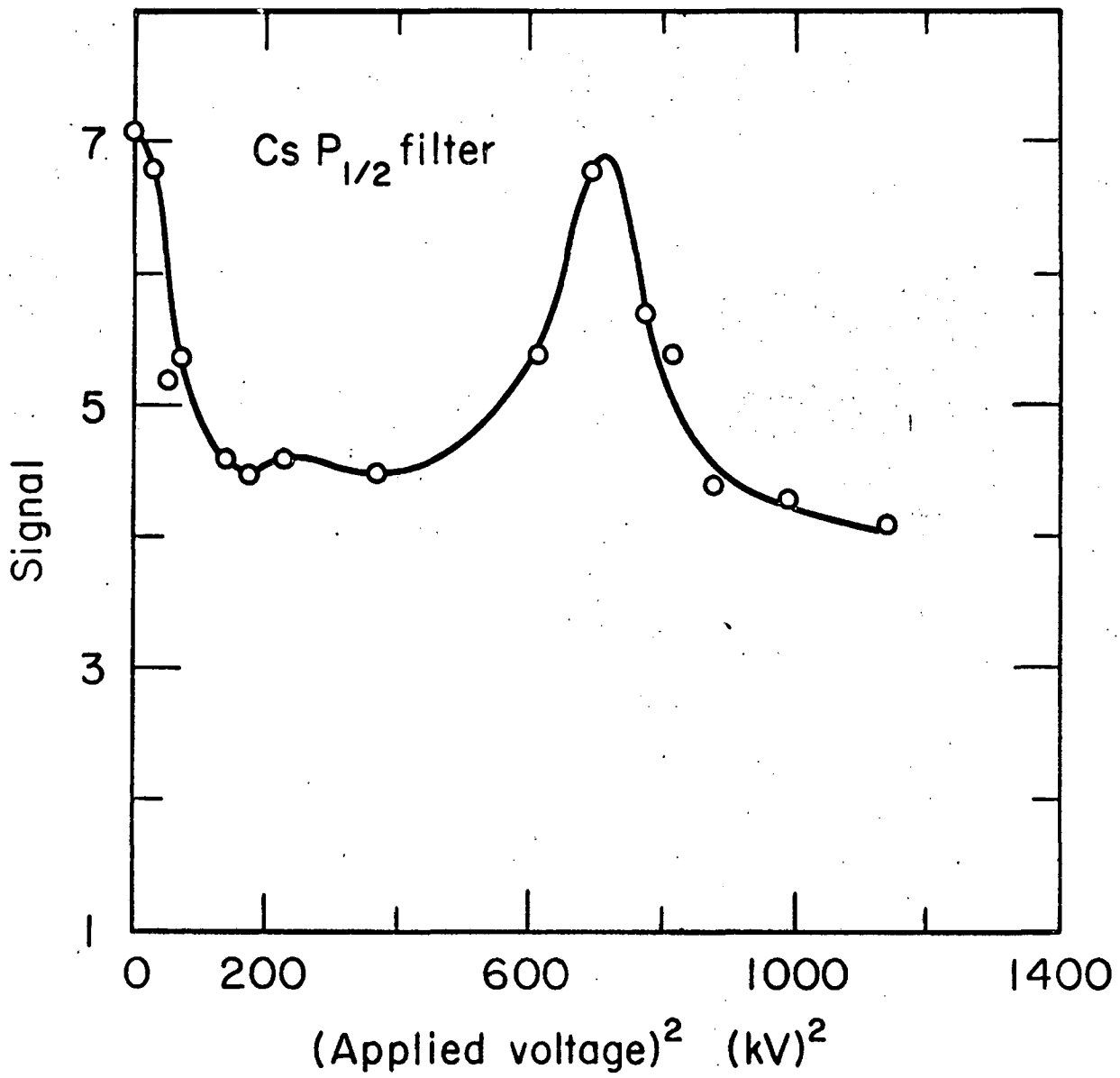

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Fig. 3 A

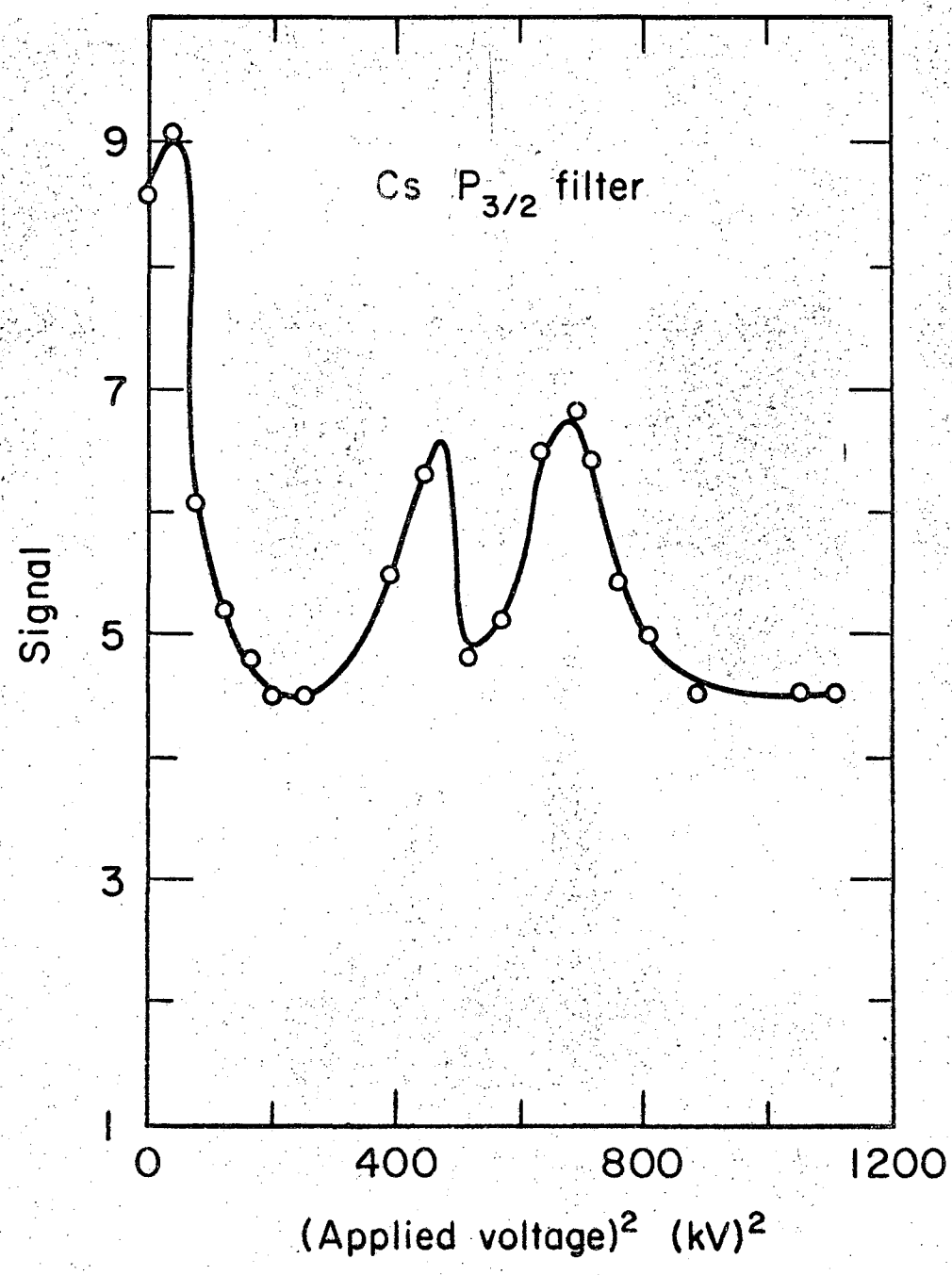

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Fig 3 B

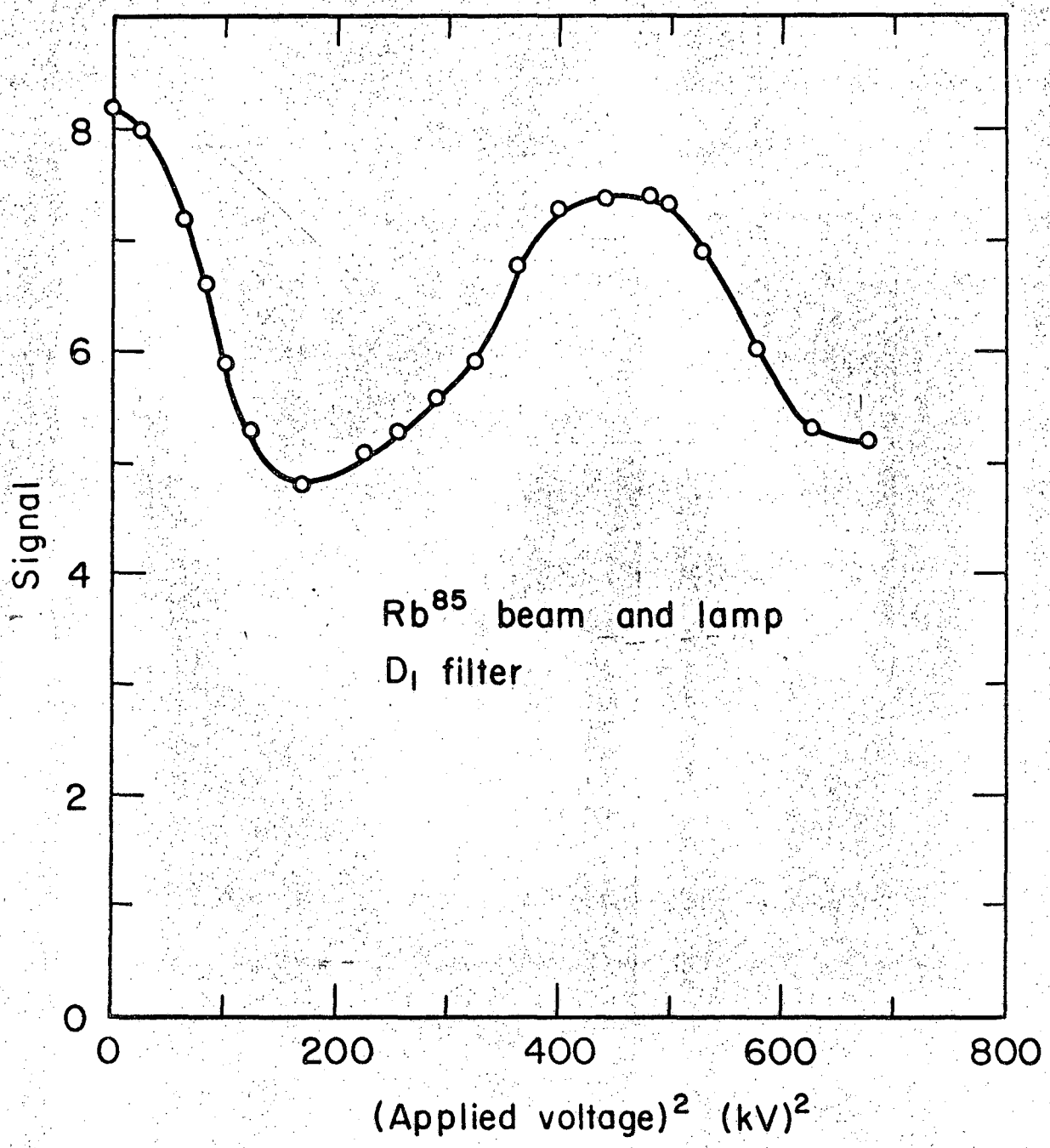

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Fig. 4 A

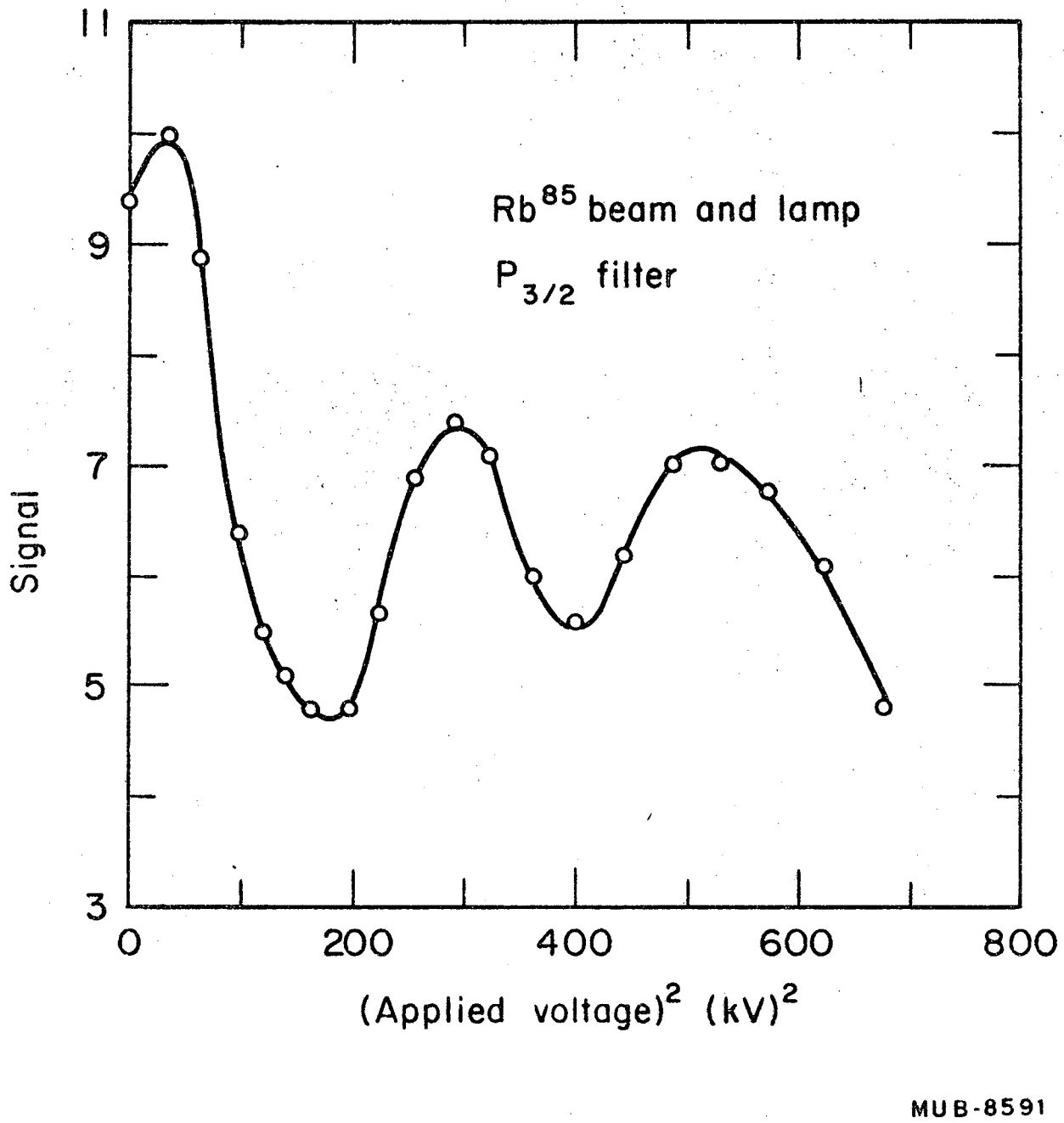
Rb85 beam and lamp
P3/2 filter
Signal
11
9
7
5
3
0
200
400
600
800
(Applied voltage)2 (kV)2
MUB-8591

Fig. 4 B

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