Electric Dipole Moments of Alkali Atoms. A Limit to the Electric Dipole Moment of the Free Electron *

J. P. Carrico,† E. Lipworth, P. G. H. Sandars,‡ T. S. Stein,§ and M. C. Weisskopf
Brandeis University, Waltham, Massachusetts
(Received 19 January 1968; revised manuscript received 17 July 1968)

The observation of an electric dipole moment (EDM) in an atomic system of well-defined angular momentum would be direct evidence for violations of both parity and time-reversal invariances. A search has been made for an EDM in the cesium atom, using an atomic-beam magnetic-resonance technique. A large 25-cps voltage is applied across two parallel metal plates situated between the loops of a Ramsey double-hairpin structure, in an atomic-beam apparatus adjusted to observe "flop-in" transitions. A beam of cesium atoms passes between the plates and is there subject to an electric field of about 5×10^4 V/cm. The "flop-in" transition $(4, -3) \leftrightarrow (4, -4)$ is observed for a low magnetic field with the rf oscillator adjusted so that the signal at the detector corresponds to the point of maximum slope on either side of the central peak of the Ramsey pattern. If the atom possessed an EDM, it would produce a 25-cps component in the signal at the detector when 25-cps ac voltage alone is applied to the electric field plates. Using phase-sensitive detection techniques, it is possible to separate out such a component from the expected 50-cps component due to the quadratic Stark interaction. The width at half-intensity of the central Ramsey peak is about 2 kc/sec, and shifts of the order of a few parts in 10^5 of this are easily detected. A 25-cps component of the detector signal is observed. However, it is likely that this signal is caused by one or more instrumental effects which can simulate an EDM signal. Such instrumental effects are discussed in detail. Also, measurements are performed on other alkali atoms, and comparisons are made between their signals. The results of these comparisons show that the interaction between the atom's magnetic dipole moment and the magnetic field $[(\vec{v}/c) \times \vec{E}\,]$ due to the atom's motion through the electric field is responsible for most of the linear signals. On the basis of these comparisons and subject to the limitations discussed in the text, our limit to the EDM of the Cs atom is $(5.1 \pm 4.4) \times 10^{-20} e$ cm. This result can be used: (1) to set a limit to the EDM of the free electron. If the electron possesses an EDM, then from a relativistic argument it can be shown that the EDM of the Cs atom would be approximately 133 times larger. Using this enhancement factor, our limit to the EDM of the free electron is $(3.8 \pm 3.3) \times 10^{-22} e$ cm. (2) to set a limit to the purity of the Cs ground state. Writing the Cs ground state as $|s\rangle + a|p\rangle$, we find $\mathrm{Re}\,a \leq 1.4 \times 10^{-12}$, which is of the order of magnitude expected from a parity violation in the weak interactions.

I. INTRODUCTION

Several years ago Lee and Yang[1] pointed out that the existence of an electric dipole moment (EDM) in an elementary particle would imply the violation of both parity (P) and time-reversal (T) invariances. Since at that time it was thought that T invariance was exact, the search for EDM's was not pursued very vigorously. The limits which were *then* available are given in Table I.

The situation changed dramatically in 1964 when Christenson, Cronin, Fitch, and Turlay[2] reported their discovery of the CP-violating $K_2{}^0 \rightarrow 2\pi$ decay. Through the CPT theorem, this violation of CP invariance implies that there must be a corresponding violation of T invariance. However, in spite of strenuous effort, no direct experimental evidence for T violation has yet been found. It seems very likely that the best evidence concerning T violations can come from sensitive experiments on the EDM's of elementary particles.

This interesting situation has stimulated considerable experimental activity in the search for EDM's. Ramsey and his group have enormously improved the precision of their neutron-beam-resonance technique to obtain a limit on the EDM of the neutron $D \lesssim 3 \times 10^{-22} e$ cm.[3] Similar precision has been obtained by Shull and Nathans,[4] who found by means of a very elegant neutron scattering experiment $D \lesssim 3 \times 10^{-22} e$ cm. In a neutron-beam-resonance experiment Cohen *et al.*[5] determined a neutron EDM limit which is consistent with the smaller limits of the experiments by Shull and Nathans[4] and by Ramsey and his group.[3] Rand[6] has reported a limit $D \lesssim 2 \times 10^{-16} e$ cm for the EDM of the electron at high momentum transfer. He deduced this limit from an analysis of 180° scattering of 200-MeV electrons in the Stanford linear accelerator.

Evidence concerning P and T violations can also be obtained from sensitive experiments searching for a linear Stark effect (LSE) in atoms. The argument of Lee and Yang[1] can be used to show that the observation of a LSE in an atom would be direct evidence for violations of both P and T invariances. Since LSE experiments involve the measurement of effects of an applied electric field on transitions between different atomic levels, the quantity that is determined is the limit to a differential LSE between the atomic levels under investigation. Because linear Stark interactions can be represented by parameters having the dimensions of EDM, a common practice is to use EDM in referring to the limit to a LSE in an atom.

TABLE I. Upper limits on some EDM's prior to the discovery of *CP* violation.

Particle	EDM (*e* cm)	Ref.
Electron	1.2×10^{-15}	a
Electron	$4\ \times10^{-16}$	b
Muon	0.6×10^{-17}	c
Neutron	2.4×10^{-20}	d
Proton	1.3×10^{-13}	e
Neutrino	$2\ \times10^{-20}$	f

[a] J. Goldemberg and Y. Torizuka, Phys. Rev. 129, 2580 (1963).
[b] D. T. Wilkinson and H. R. Crane, Phys. Rev. 130, 852 (1963).
[c] G. Charpak, F. J. M. Farley, R. L. Garwin, T. Muller, J. C. Sens, and A. Zichichi, Nuovo Cimento 22, 1043 (1961).
[d] J. H. Smith, E. M. Purcell, and N. F. Ramsey, Phys. Rev. 108, 120 (1957).
[e] R. M. Sternheimer, Phys. Rev. 113, 828 (1959).
[f] S. Rosendorff, Nuovo Cimento 17, 251 (1960).

In 1962, Ensberg reported results of a search for an EDM of the Rb^{85} atom.[7] Using an optical-pumping experiment, he obtained a limit $D \lesssim 10^{-18}e$ cm. This work has since been published in detail.[8]

In 1963, experiments were begun to search for an EDM of the Cs atom.[9] The method involved searching with high-sensitivity atomic-beam magnetic-resonance techniques for an EDM in the ground state of the Cs atom. The transition between the levels (4, − 4) and (4, − 3) of Cs was induced in a Ramsey double-hairpin structure in a weak uniform magnetic field $\vec{H}$. A set of parallel electric field plates was situated between the rf loops. As previously reported,[10] a small shift, linear in the applied electric field ($\vec{E}$), was observed. This effect could possibly have been interpreted as due to the interaction of an EDM of the Cs atom with this field. Stated as a limit to the Cs-atom EDM, the result of these experiments was $D \lesssim (2.2\pm0.1)\times10^{-19}e$ cm.

As was pointed out at the time, it was possible that a $\vec{v}\times\vec{E}$ effect was causing the linear shift. As the atom moves with velocity $\vec{v}$ through the electric field it experiences a motional magnetic field $\vec{v}\times\vec{E}$. A resonance shift, linear in $\vec{E}$, due to the interaction of the atom's magnetic dipole moment with this motional magnetic field, results if $\vec{E}$ is not parallel to $\vec{H}$. The experiments are quite sensitive to this, since a misalignment of 0.01 rad would explain the above result. This possibility has been investigated by comparing shifts in various alkali atoms.[11] In this paper, we report the details of the experiments and the results of the comparisons.

As mentioned earlier, the observation of a LSE would constitute direct evidence for violations of *P* and *T*. Sandars[12] has discussed general ways in which such violations can occur and has shown that a LSE in the ground state of an alkali atom can be expressed in terms of three independent parameters having the dimensions of an EDM. A number of different models of *P* and *T* violations can contribute to these parameters and sensitivity of a particular experiment ot each model depends on the nature of the resonance transition under investigation. In this paper, we report a limit to the differential LSE between the levels (4, − 3) and (4, − 4) of the ground state of Cs. Because our experiments involve observations of electric field effects on transitions between Zeeman sublevels, our search for a LSE involves LSE contributions which affect each Zeeman sublevel differently. As mentioned earlier, it is common to use EDM in referring to the limit to a LSE in an atom. This presents no problem as long as it is realized that an atom can have several EDM's, owing to the possibility of several contributions to the LSE. We shall adopt the convention in which a *limit to the EDM of the atoms means a limit to the differential* LSE *between Zeeman substates*.

Our limit to the EDM of the Cs atom can be interpreted in terms of a limit to the EDM of the free electron. At first sight, this approach is not promising. Schiff[13] has given a very powerful theorem which implies that an atom cannot have an EDM due to an electron EDM. However, it has been shown elsewhere[14] that this theorem is only true in the nonrelativistic limit. If a full relativistic calculation is carried out, one finds that the ratio $S=D_{\text{atom}}/D_{\text{elec}}$ is nonzero and in favorable cases can even be greater than unity. In Ref. 14, the values of the shielding factor *S* have been calculated for the ground states of the alkali atoms. The results are reproduced in Table II. These values are, of course, only approximate since the full many-body problem in atoms cannot be solved exactly. However, the single-particle approximation which was used is likely to be a good approximation for the alkali atoms and the errors in the values in Table II are not expected to be large. In particular, the trend of *S* with atomic number is likely to be correct.

The important results from Table II are that the shielding factor *S* increases rapidly with atomic number and that for Cs it is 133. This large value of *S* is produced by the favorable combination of high atomic number ($Z=55$) and enormous polarizability ($\alpha=48\times10^{-24}$ cm^3).[15] This enhancement factor together with some new high-sensitivity atomic-beam techniques contribute to the great improvement in sensitivity of the result reported here.

II. PRINCIPLE OF THE EXPERIMENT

A method has been developed which enables us to make measurements of resonance shifts as small as a few parts in 10^5 of the linewidth. Experiments are conducted on an atomic-beam magnetic-resonance apparatus. The atomic beam passes between a pair of parallel metal plates which are situated between the loops of a Ramsey double-hairpin structure.[16,17] Figure 1 shows the Breit-Rabi diagram for Cs. The transition between the levels (4, − 4) and (4, − 3) is induced in a low magnetic field with the rf oscillator frequency adjusted so that the signal at the hot-wire detector corresponds to the point of maximum slope on

TABLE II. Values for the ratio of the EDM of some atomic ground states to the EDM of the free electron.[14]

Atom	Z	$S=D_{\text{atom}}/D_{\text{elec}}$
Li	3	4.5×10^{-3}
Na	11	3.3×10^{-1}
K	19	2.65
Rb	37	27.5
Cs	55	133

either side of the central peak of the Ramsey interference pattern.

Our method makes use of the fact that the resonance signal is some function $S(f-\nu)$ of the difference between the oscillator frequency ν and the Bohr resonance frequency $f=(W_2-W_1/h$. On applying the electric field $\vec{E}$, the separation between energy levels changes, and hence f changes from f_0 to $f_0+\delta f$, say. For small δf, the resultant change in signal intensity is given by

$$\delta S=S(f_0+\delta f-\nu_0)-S(f_0-\nu_0)$$
$$=(\partial S/\partial f)_{f_0-\nu_0}\delta f+\tfrac{1}{2}(\partial^2S/\partial f^2)_{f_0-\nu_0}(\delta f)^2+\cdots \,. \tag{1}$$

As shown experimentally elsewhere[18] for the small shifts that we are interested in, the curvature terms $\partial^2S/\partial f^2$, etc., can be neglected, and the signal change is proportional to the change in resonance frequency

$$\delta S=g\delta f\,, \tag{2}$$

with $g=(\partial S/\partial f)_{f_0-\nu_0}$. The constant g is clearly a function of $(f_0-\nu_0)$, and it can be maximized by working at the point of maximum slope and by obtaining as narrow a resonance line as possible.

To determine the value of g, we make a slight change in the oscillator frequency (with the electric field off). The resulting change in detector signal is given by

$$\Delta S=S(f_0-\nu_0+\delta\nu)-S(f_0-\nu_0)$$
$$=(\partial S/\partial\nu)_{f_0-\nu_0}\delta\nu\,. \tag{3}$$

But $\partial S/\partial\nu=-\partial S/\partial f$, (4)

and hence $\Delta S=-g\delta\nu$. (5)

Thus the slope can be calibrated by frequency modulation of the oscillator and measurement of the resulting detector signal with a phase-sensitive detector.

To determine a possible EDM effect, the shift in the resonance by an electric field $\vec{E}$ is written as

$$\delta f=k_1\vec{E}^2+k_2\vec{E}\,, \tag{6}$$

where k_1E^2 describes the quadratic Stark interaction and k_2E represents a possible EDM effect. The quadratic effect has been observed, and the result, $k_1=(-127\pm20)\times10^{-10}$ cps/(V/cm)^2, is reported elsewhere.[18]

If simultaneously an ac voltage is applied to one of the electric field plates and a dc voltage to the other, i.e.,

$$E=E_0\sin\omega t+E_{\text{dc}}\,,$$

then $\delta f=k_1E_{\text{dc}}^2+\tfrac{1}{2}k_1E_0^2+2k_1E_0E_{\text{dc}}\sin\omega t$

$$-\tfrac{1}{2}k_1E_0^2\cos2\omega t+k_2E_0\sin\omega t+k_2E_{\text{dc}}\,. \tag{7}$$

Using a phase-sensitive detector, k_2 can be determined in two ways: (a) by observing the component of the signal oscillating at ω, where

$$\delta f(\omega)=(2k_1E_{\text{dc}}+k_2)E_0\sin\omega t\,. \tag{8}$$

A measurement of the slope of the line obtained by plotting δf against E_{dc} (at fixed E_0) determines k_1. If at $E_{\text{dc}}=0$, the curve does not pass through the origin; the intercept with the δf axis can be used to measure k_2. (b) If $E_{\text{dc}}=0$, then the component at ω is

$$\delta f(\omega)=k_2E_0\sin\omega t\,. \tag{9}$$

Consequently, k_2 is the slope of the curve δf versus E_0 taken with zero dc field.

During an experiment, observations are taken by moving from one side of the central resonance peak to the other. It is easily seen that in moving from side to side, the phase of the signal at the detector changes by 180°. Thus the signal as measured by the phase-sensitive detector is the difference between the signals that are measured on each side. Since the same remark is true for the calibration (slope measuring) signal, any difference in the

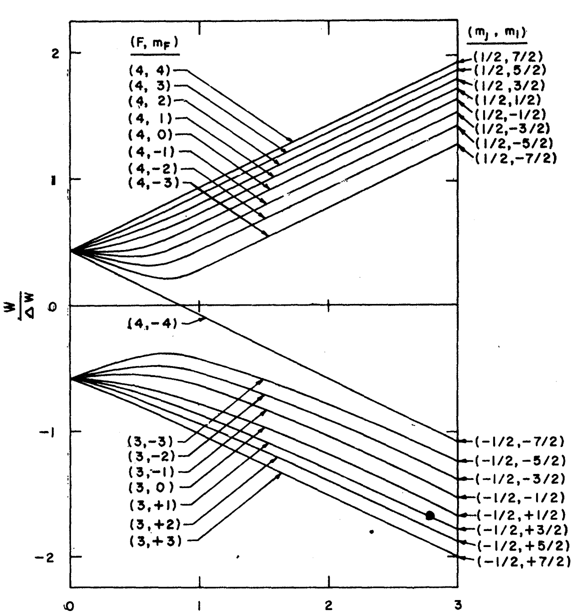

FIG. 1. Breit-Rabi diagram for Cs.

slope on the two sides is not important. On the other hand, any spurious amplitude modulation of the rf oscillator produced by the frequency-modulating circuit, which could produce a signal of the same phase at the two sides of the resonance, is automatically subtracted out. Also, spurious pickup at the detector is subtracted out. Thus, any change in the dc output of the phase-sensitive detector in going from one side of the resonance to the other can be attributed only to a resonance-dependent phenomenon.

III. APPARATUS

A. General

A schematic of the atomic-beam apparatus is presented in Fig. 2, which shows the source, deflecting magnets (A and B fields), uniform magnet (C field), and the surface ionization detector. A rf Ramsey double-hairpin structure is located in the C field. Situated between the rf loops is a set of parallel electric field plates. The details of the C-field region are shown in Fig. 3 and an exploded view of the electric field plates is given in Fig. 4. This apparatus has been described in more detail elsewhere.[18]

B. Electronics

The signal from the hot wire is amplified by a Tektronix (122) amplifier and then detected by a phase-sensitive amplifier (Princeton Applied Research JB-4). The dc output of the lock-in is displayed on a recorder (Bausch & Lomb VOM 5). The signal is also monitored by a Keithley (603) electrometer amplifier, used to observe the dc level of the beam and to adjust to the side of the central peak of the Ramsey pattern. The detection scheme is shown in Fig. 5.

The ac high voltage is supplied to the field plates using the scheme shown in Fig. 6. The ac high voltage is monitored with an oscilloscope using a well-calibrated resistance divider chain across the transformer output.

The modulation frequency is derived from an audio oscillator (Hewlett-Packard 201 CR). A modulation frequency of 25 cps is used to avoid possible complications from subharmonics of the line frequency and to minimize effects due to the finite beam transit time.

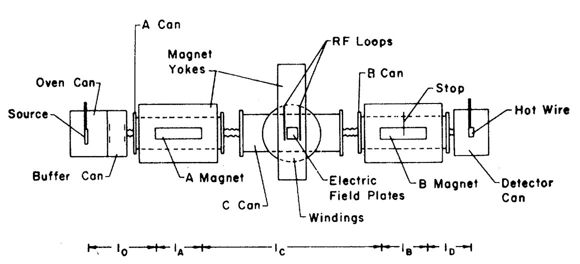

FIG. 2. Schematic of the atomic-beam magnetic-resonance apparatus. $l_0 = 16\frac{1}{2}$ in., $l_A = l_B = 12$ in., $l_C = 44$ in., $l_S = 6$ in., and $l_D = 11\frac{1}{2}$ in.

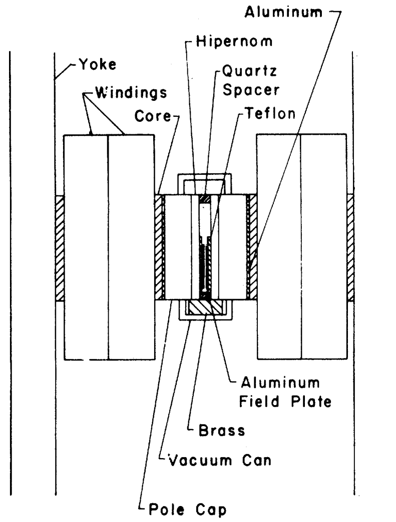

FIG. 3. C magnet.

The rf loops are driven by a General Radio unit oscillator (1211–C) which is modified for frequency modulation. Frequencies are monitored by a General Radio digital frequency meter (1130–A).

A General Radio frequency meter and discriminator (1142–A) is used to measure the amplitude of the frequency modulation. The details are given in Fig. 7. The unit oscillator which drives the Ramsey loops is heterodyned with a local oscillator (General Radio 1211–C) in a mixer (General Radio 874–MR) to yield a 150-kc/sec signal, which is then fed into the discriminator. A sinusoidal frequency modulation of the oscillator which drives the loops results in a modulation of the 150-kc/sec beat frequency, which is measured as an ac voltage superimposed on the dc output of the discriminator. The sensitivity is such that a frequency modulation of 100-cps amplitude produces a signal of 10 mV. The discriminator's output is measured using a phase-sensitive detector.

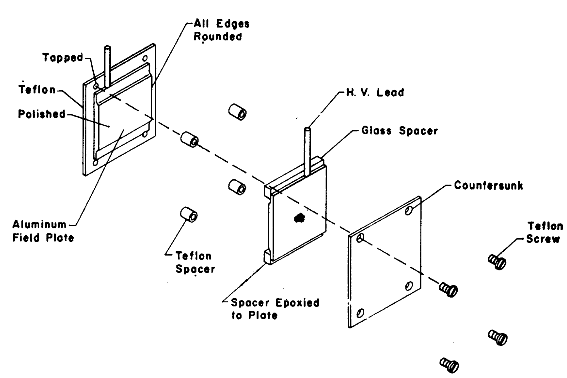

FIG. 4. Electric field plates. Spacing = 0.0675 ± 0.0005 in., plate length = plate width = 3 in., plate thickness = $\frac{5}{32}$ in., and Teflon thickness = $\frac{1}{8}$ in.

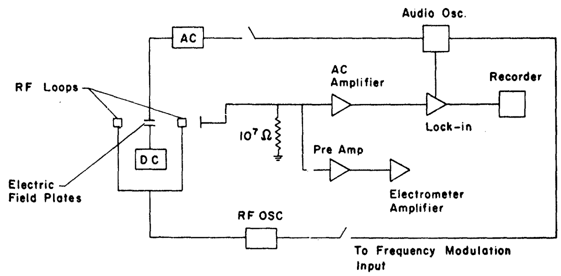

FIG. 5. Detection scheme.

IV. RESULTS

A typical Ramsey pattern is shown in Fig. 8. The full width at half-intensity of the central peak is about 2 kc/sec for Cs. Typical full beam strengths are about 5×10^{-8} A. At resonance, the recovered beam is about 25×10^{-10} A. The phase-sensitive detector signal (obtained by moving from side to side of the central resonance peak) is shown in Fig. 9. This signal corresponds to a shift in the resonance of about 5×10^{-4} of the linewidth of the central peak. The integration time is approximately 1 sec.

In Fig. 10, the shift in the resonance is plotted as a function of E_0E_{dc} (with E_0 held constant and E_{dc} varied). It is seen that a finite signal remains when $E_{dc}=0$. This signal was further investigated by keeping $E_{dc}=0$ and observing the component of the resonance signal modulated at ω as a function of E_0. A typical result of this is shown in Fig. 11. For values of E_0 less than 2×10^4 V/cm the shift is linear in E_0. However, as noted in Fig. 11, curvature is present for values of E_0 greater than about 2×10^4 V/cm. From Eq. (9) such curvature is unexpected. However, if harmonics of the fundamental modulating frequency are present in the HV transformer output, then even though there is no dc bias on the field plates (i.e., $E_{dc}=0$), a signal occurring at ω can still arise from the quadratic Stark interaction in the following way.

With harmonics present, the field between the plates is expressed as

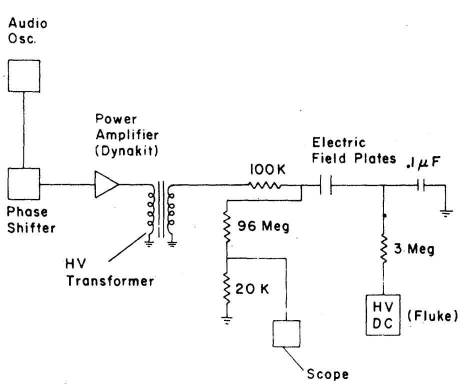

FIG. 6. High-voltage scheme.

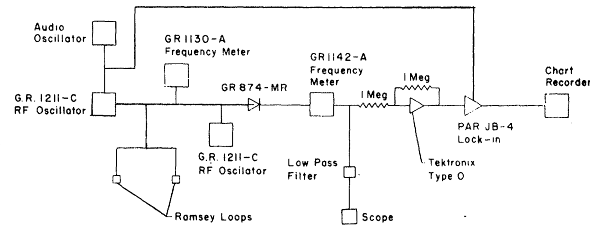

FIG. 7. Scheme for the measurement of the amplitude of frequency modulation.

$$E=E_0\sin\omega t+E_2\sin(2\omega t+\alpha)+E_3\sin(3\omega t+\alpha')+\cdots. \tag{10}$$

Substituting this into Eq. (6), the component of the shift modulated at ω with $\vec{E}_{dc}=0$ is

$$\delta f(\omega)=k_1[E_0E_2\cos(\omega t+\alpha)+E_2E_3\cos(\omega t+\alpha'-\alpha)+\cdots]+k_2E_0\sin\omega t. \tag{11}$$

Second and third harmonics in the output of the HV transformer were measured with a phase-sensitive detector and the results are shown in Fig. 12.

Within the range of the values of E_0 used in the present experiments, $E_2<0.1\,E_0$ and $E_3<E_2$, so that with less than 10% error

$$\delta f(\omega)\sim k_1E_0E_2\cos(\omega t+\alpha)+k_2E_0\sin\omega t. \tag{12}$$

From a detailed analysis of the phases of the voltage applied to the plates and the resonance signals, using a phase-sensitive detector, it was found that $\alpha\sim90°$, so that the amplitude of the shift modulated at ω is

$$\delta f\sim -k_1E_0E_2+k_2E_0 \tag{13}$$

or $$\delta f/\vec{E}_0\sim -k_1E_2+k_2. \tag{14}$$

The constant k_2 is found in the following manner. $\delta f/E_0$ is plotted against E_2 (see Fig. 13). The slope of this line determines k_1 and the intercept with the $\delta f/E_0$ axis determines k_2. The average value of k_2 found in this manner is 1.04×10^{-5} cps/(V/cm) based on 12 experiments. The value of k_2 obtained directly from the data and Eqs. (2), (5), and (14) must be multiplied by $\frac{4}{3}$ to take into account the fact that the electric field plates extend only $\frac{3}{4}$ of the distance between the centers of the rf loops.[17] This leads to a so-called filling factor correction of $\frac{4}{3}$. We have already included this factor of $\frac{4}{3}$ in the average value 1.04×10^{-5} cps/(V/cm) for k_2. In Fig. 13, the plot of $\delta f/E_0$ versus

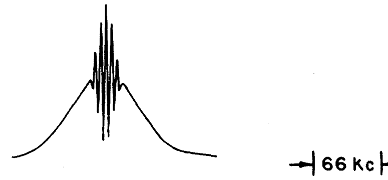

FIG. 8. Ramsey pattern.

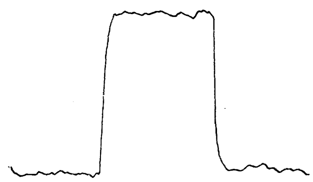

FIG. 9. Signal produced by a Stark shift of the resonance.

E_2 is shown for the data of Fig. 11. The plot is seen to be linear, which indicates that the approximations leading to Eq. (14) are reasonable.

In the course of the experiments, it was found that the harmonic content changed in time. Fortunately, however, the value k_2 obtained in the way just described is not very sensitive (at least within the accuracy of the present experiments) to the exact amount of E_2. On the other hand, this method of data reduction does not lead to reliable values of k_1, because there is a large uncertainty in the knowledge of E_2.

As mentioned earlier, the intercept with the δf axis in a plot of δf versus $E_{\rm dc}$ can also be used to determine k_2. However, this method was not used because its accuracy is limited by the uncertainties in $E_{\rm dc}$ and E_2.

V. DISCUSSION OF RESULTS

As noted earlier, the shift of the resonance frequency caused by the interaction of the atom with an electric field E is expressed by

$$\delta f = k_1 E^2 + k_2 E, \qquad (15)$$

where k_1E^2 is the quadratic Stark effect and k_2E is a possible EDM effect. If $E = E_0 \sin\omega t + E_{\rm dc}$, the component of the shift modulated at ω is

$$\delta f(\omega) = (2k_1 E_{\rm dc} + k_2) E_0 \sin\omega t. \qquad (16)$$

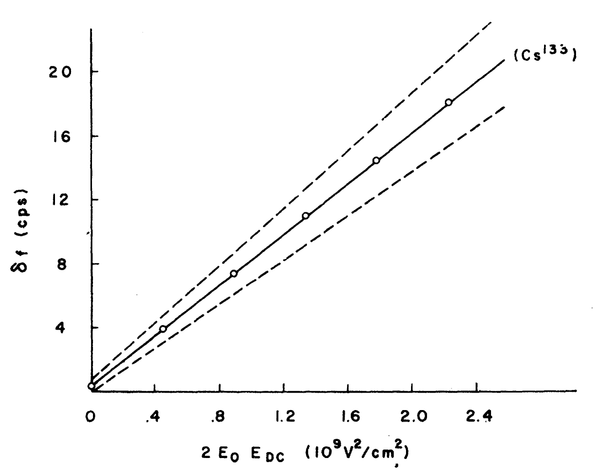

FIG. 10. Quadratic Stark effect in cesium observed at ω with E_0 held constant and $E_{\rm dc}$ varied. The data are uncorrected for the filling factor. The dashed lines indicate the error as calculated from estimated uncertainties in the calibrations.

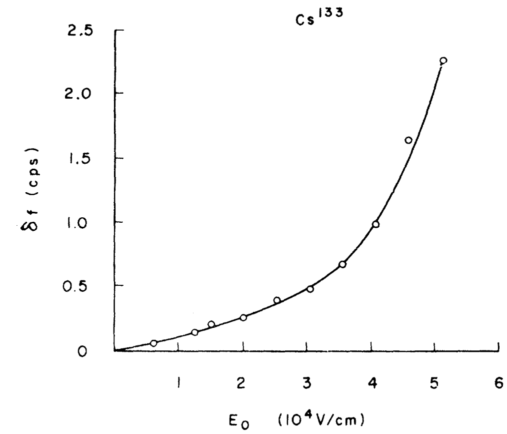

FIG. 11. Shift in Cs observed at ω with $E_{\rm dc} = 0$. The data are uncorrected for the filling factor.

This expression implies that if the atom has an EDM, the signal due to its interaction with an electric field must satisfy the following conditions: (a) The signal at ω must not vanish when $E_{\rm dc} = 0$; (b) the signal at ω when $E_{\rm dc} = 0$ must be linear in E_0; and (c) the signal at ω when $E_{\rm dc} = 0$ must be in phase or 180° out of phase with the quadratic Stark signal modulated at $\omega(E_{\rm dc} \neq 0)$. These conditions were investigated for values of E_0 less than 2×10^4 V/cm so as to minimize interference from the quadratic Stark effect arising because of harmonics

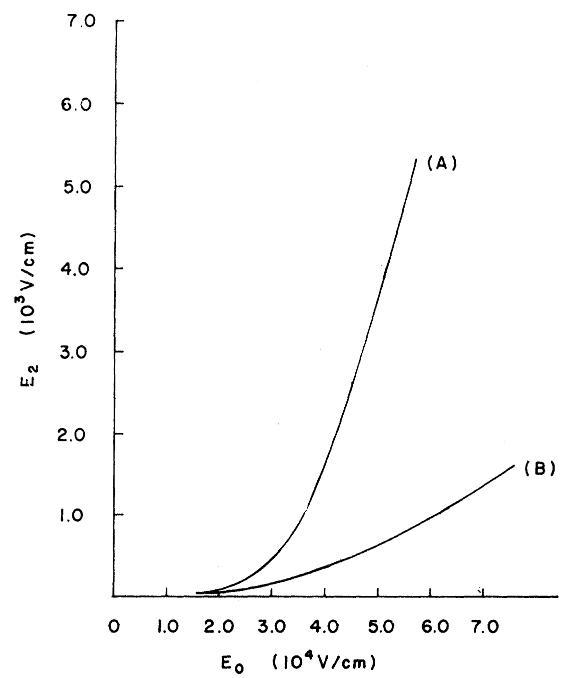

FIG. 12. Harmonic content in the output of the high-voltage transformer. Curves A and B indicate the second and third harmonics, respectively.

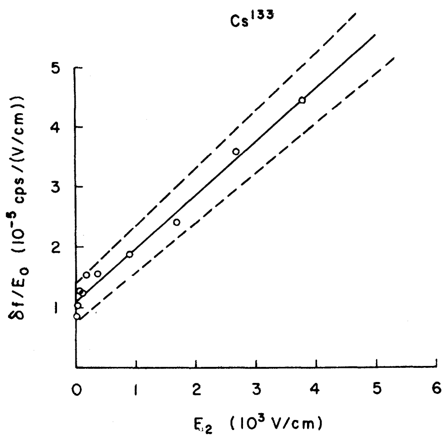

FIG. 13. $\delta f/E_0$ is plotted versus E_2 for Cs. The data are uncorrected for the filling factor. The dashed lines indicate the error as calculated from estimated uncertainties in the calibrations.

in the HV transformer output [see Eq. (13)]. Typical results for Cs are presented in Figs. 10 and 11. From Fig. 10 it is seen that the signal does not vanish when E_{dc} goes to zero; from Fig. 11 it is seen that the signal at ω with $E_{dc}=0$ is a linear function of the electric field (for values of $E_0 < 2\times10^4$ V/cm. Also, the signal modulated at ω when $E_{dc}=0$ was found to be in phase with the signal when $E_{dc}\neq 0$. The dashed lines in Figs. 10 and 13 indicate the error as calculated from estimated uncertainties in the calibrations. The standard deviation associated with each datum point is much too small to be indicated on the graph. A discussion of errors is presented elsewhere.[18]

On the basis of 12 measurements, the average value of k_2 is 1.04×10^{-5} cps/(V/cm) (filling factor included). The standard deviation of the mean is 0.07×10^{-5} cps/(V/cm). This result can be directly used to set a limit to the EDM of the Cs atom in the following way. Assume the Cs atom possesses an EDM $\mu_{\vec{E}}$ and also assume the observed signal linear in $\vec{E}$ (i.e., k_2E) is completely due to this EDM. Then

$$\mu_{\vec{E}} = eD = k_2hF , \qquad (17)$$

where F is the total spin of the atom, h is Planck's constant, e is the electronic charge in esu, and D is the distance in cm. For Cs, $F=4$ and we find $D \lesssim 2\times10^{-19}$ cm. However, we cannot at this point assume that the signal linear in $\vec{E}$ is due entirely to an EDM of the atom because of the following possible alternative explanations for the signal: (a) It is possible that the signal is due to an instrumental effect which simulates an EDM signal or (b) it is possible that the signal is the vector resultant of a true EDM signal combined with an instrumental signal.

The most likely causes of electric dipole simulating signals are (a) the interaction of the atom's magnetic dipole moment with a magnetic field whose origin is in the high voltage applied to the electric field plates and (b) an instrumental effect which in some way is caused by the quadratic Stark interaction. Concerning the latter, an example has already been given; the mixing effects of the transformer harmonics had to be taken into account in analyzing the Cs data.

Effects which could produce dipolelike signals might possibly arise from (a) a displacement current (the field plates comprise a condenser), (b) a motional magnetic field, (c) magnetic effects due to transformer harmonics, (d) leakage currents, (e) motion of the magnet pole pieces, (f) magnetic pickup, and (g) spurious dc voltages. These effects are discussed in detail below. In order to learn whether any of these effects are responsible for the linear signal, other alkali atoms are studied, and comparisons are made between their linear signals. In this way, it is possible to determine if the linear signals are caused by (a) the quadratic Stark effect, (b) a magnetic effect which is independent of the atom's velocity, or (c) a magnetic effect which depends on the atom's velocity.

Typical results of measurements of linear signals in other alkali atoms are shown in Fig. 14 for Rb^{85} and Rb^{87} and in Fig. 15 for Na and K. Final average values of k_2 for each alkali atom are presented in Table III (filling factor included), where the uncertainty is the standard deviation of the mean (the number of experiments performed on each alkali is also indicated).

Values of k_2 for Rb^{85} and Rb^{87} are obtained in the same manner as for Cs. However, because the quadratic Stark shifts[18] in Na and K are much smaller than in Cs and Rb^{87} and there is no evidence on their resonance signals of the effects of transformer harmonics, k_2 for each of these elements is obtained directly from the slope of the line $\delta f = k_2E_0$.

We now discuss the instrumental effects mentioned earlier.

A. Displacement Current

The electric field plates form a capacitor into which a displacement current flows when the oscillating high voltage is applied. This displacement

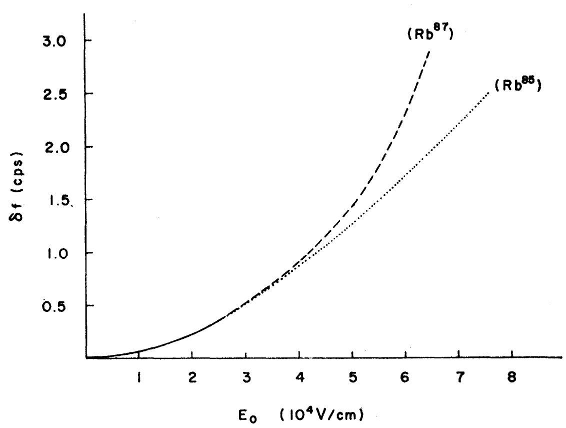

FIG. 14. Linear shifts observed in Rb^{85} and Rb^{87}. The data are uncorrected for the filling factor.

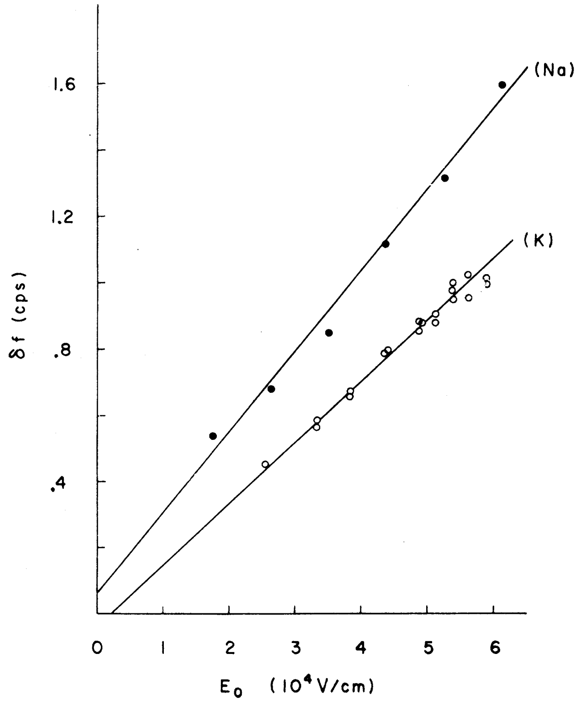

FIG. 15. Linear shifts observed in Na and K. The data are uncorrected for the filling factor.

current results in a magnetic field which could modulate the Zeeman levels. If $E = E_0 \sin\omega t$, then, since $i = C\partial V/\partial t$, this magnetic field can be written as

$$H_1 = a\omega E_0 \cos\omega t \ . \tag{18}$$

Here i is the displacement current, C is the capacitance between the plates, and a is a geometric constant. For circular plates, a can easily be calculated. We have $a = r/2c$, where r is the radius at which $\vec{H}_1$ is calculated. To obtain an estimate to the signal produced by $\vec{H}_1$, assume $r \sim 10$ cm (the length and height of the plates), $\omega = 2\pi \times 25$ cps, $E_0 \sim 10^5$ V/cm. We find $H_1 \sim 10^{-5}$ G, which, if $\vec{H}_1$ were parallel to $\vec{H}_0$, would produce a 4-cps shift in the Cs resonance. In fact, $\vec{H}_1$ is probably inclined at some angle γ to $\vec{H}_0$. The separation of Zeeman levels is then proportional to $\vec{H}_R$, where

$$H_R = [H_0^2 + H_1^2 + 2H_0H_1\cos\gamma]^{1/2} \ . \tag{19}$$

Because $H_1 \ll H_0$ ($H_1 \sim 10^{-5}$ G and $H_0 \sim 10$ G), we have

$$H_R \sim H_0 + a\omega E_0 \cos\omega t \cos\gamma \ . \tag{20}$$

It is very difficult to calculate an appropriate average for γ since the angle between the plates and $\vec{H}_0$ is not known. However, the angle between the plates and $\vec{H}_0$ is obviously close to $\frac{1}{2}\pi$. Neglecting effects from lead wires, symmetry considerations show that $\vec{H}_1$ is probably inclined to $\vec{H}_0$ at an angle close to $\frac{1}{2}\pi$, i.e., $\gamma \sim \frac{1}{2}\pi$. This means that the resonance shift at ω (with $E_{dc} = 0$) due to $\vec{H}_1$ should be very much smaller than the 4 cps quoted earlier.

It is seen from Eq. (18) that a signal caused by a displacement current must satisfy the following conditions: (a) The signal (with $E_{dc} = 0$) must be linear in the modulation frequency ω, (b) the signal must be linear in E_0, and (c) the signal must be 90° out of phase with the quadratic Stark signal modulated at $\omega(E_{dc} \neq 0)$. Using a phase-sensitive detector, the signal (with $E_{dc} = 0$) was found to be in phase with the quadratic Stark signal modulated at $\omega(E_{dc} \neq 0)$. Also, the signal was found to be independent of ω. Therefore, it is concluded that $\gamma \sim \frac{1}{2}\pi$, so that a displacement current effect was negligible in the present experiment. The fact that a displacement current was present was substantiated by the measurement of an ac current of 10^{-5} A to the plates, which is consistent with the capacitance of the plates (20 pF).

TABLE III. Final average values of k_2.[a]

Element	$k_2 [10^{-5}$ cps/(V/cm)]	No. of expts.
Cs	1.04 ± 0.07[b]	12
K	2.79 ± 0.21	10
Na	3.84 ± 0.21	11
Rb^{85}	1.43 ± 0.20	6
Rb^{87}	2.15 ± 0.16	14

[a]These values have been corrected for the filling factor.

[b]The uncertainty is the standard deviation of the mean.

B. Motional Magnetic Field

As the atom travels between the electric field plates, it experiences a motional magnetic field $\vec{H}_1 = (\vec{v}/c) \times \vec{E}$, where $\vec{v}$ is the velocity of the atom. $\vec{H}_1$ combines with $\vec{H}_0$ to produce a resultant field $\vec{H}_R$ with which the atom interacts. The part of $\vec{H}_R$ modulated at ω (with $\vec{E}_{dc} = 0$) is

$$H_R(\omega) = v/c)\sin\gamma \sin\theta \vec{E}_0 \sin\omega t \ , \tag{21}$$

where γ is the angle between $\vec{v}$ and E and θ is the angle between $\vec{E}$ and $\vec{H}_0$. If the signal at ω is assumed to be due entirely to the motional magnetic field and $\gamma \sim \frac{1}{2}\pi$, then θ is found to be about $\frac{1}{2}°$. It is quite reasonable to expect such a misalignment between $\vec{E}$ and $\vec{H}_0$.

Attempts were made to run Cs beams at different velocities using an oven with a heated snout in order to determine the velocity dependence of the signal and substantiate the existence of the $\vec{v} \times \vec{E}$ effect. However, these attempts were unsuccessful because of a considerable increase in beam noise associated with the beam effusing from the heated snout. In Sec. VI B an independent method of showing that the linear signal is quite likely due to a $\vec{v} \times \vec{E}$ effect is discussed.

C. Magnetic Effects Due to Transformer Harmonics

In the discussion of possible magnetic effects which could simulate an EDM interaction, the fact that the HV transformer output contained harmonics of the fundamental modulation frequency must be considered. Such harmonics have been considered previously in context with their giving rise to a signal at ω through the quadratic Stark interaction. To consider how harmonics can cause a signal modulated at ω through a magnetic field interaction, write the resultant field $\vec{H}_R$ with which the atom's magnetic dipole moment interacts as

$$H_R = [(H_0 + H_1\cos\phi)^2 + (H_1\sin\phi)^2]^{1/2} \sim H_0 + H_1\cos\phi + \tfrac{1}{2}(H_1\sin\phi)^2/(H_0 + H_1\cos\phi) , \qquad (22)$$

where $\vec{H}_0$ is the steady C field, $\vec{H}_1$ is the magnetic field which has arisen in some way from the HV applied to the plates, and ϕ is the angle between $\vec{H}_0$ and $\vec{H}_1$.

Writing $H_1 = aE$, where

$$E = E_0\sin\omega t + E_2\sin(2\omega t + \alpha) + \cdots , \qquad (23)$$

the component of the resultant field modulated at ω is

$$H_R(\omega) \sim a\cos\phi\,\sin\omega t\, E_0 + \tfrac{1}{2}(a^2E_0E_2/H_0)\sin^2\phi\cos(\omega t + \alpha) . \qquad (24)$$

As noted earlier (Sec. IV) $\alpha \sim \frac{1}{2}\pi$, so that

$$H_R(\omega) \sim [a\cos\phi - \tfrac{1}{2}(a^2E_2/H_0)\sin^2\phi]E_0\sin\omega t . \qquad (25)$$

If the effects of harmonics are important, then, since E_2 is not a constant independent of E_0 (see Fig. 12), curvature can be expected in the plot of frequency shift as a function of E_0 for the Na and K runs. (Remember that Na and K have very small quadratic Stark effects.[18]) However, as evidenced in Fig. 15, none is present.

Also, if, for example, $\vec{H}_1$ is the motional magnetic field, i.e., $H_1 = v' \times E_0/c$, where $v' = v\sin\gamma$ (γ is the angle between $\vec{E}$ and $\vec{v}$), then

$$H_R(\omega) = \left[\frac{v'}{c}\cos\phi - \frac{1}{2}\frac{v'^2}{c^2}\frac{\sin^2\phi}{H_0}E_2\right]E_0\sin\omega t , \qquad (26)$$

where ϕ is the angle between $\vec{H}_1$ and $\vec{H}_0$. Using typical values, $E_0 \sim 5\times10^4$ V/cm, $E_2 \sim 0.1\vec{E}_0$, $H_0 \sim 10$ G, and $v \sim 5\times10^4$ cm/sec, the shift expected from the E_2E_0 contribution is approximately 1/1000 of the effect actually observed, and therefore is of no serious consequence.

D. Leakage Current

A resistance leak, either across the insulators separating the electric field plates or from a plate to ground (see Figs. 3 and 4), would permit a conduction current to flow when the voltages are applied to the plates. This current can cause a magnetic field which would affect the resonance.

The various pieces comprising the electric field plate system were carefully cleaned before assembly to minimize the possibility of leakage. At the highest dc operating voltage (10 kV), the leakage current to the plates was less than 10^{-7} A. A simple calculation indicates that the maximum resonance shift modulated at ω (with $E_{dc} = 0$), which is produced by a current of this magnitude in the present apparatus, is 0.1 cps.

It is shown in Sec. VI A that signals in different alkali atoms do not normalize to a constant when divided by g_F alone. This shows that a conduction current alone is not responsible for the signal at ω.

E. Motion of the Magnet Pole Pieces

The electric force on the field plates is proportional to the square of the electric field. If the plate structure is elastic, then plate oscillation can occur at 2ω, when $E_{dc} = 0$ and also at ω when $E_{dc} \neq 0$. This could lead to chopping of the beam. Since data are taken by moving from one side of the resonance to the other, beam-chopping effects are subtracted out and thus do not affect the results. However, the resonance can be affected by such plate motion if the motion is transmitted to the magnet pole pieces; the resulting change in the magnetic field is then detected by a change in the Zeeman splitting. A rough calculation shows that pole piece displacements of a few microinches can cause a resonance shift in Cs of the order of 10 cps or more. Evidence that plate motion was, in fact, occurring was obtained in two ways: (a) Small signals at ω (with $E_{dc} \neq 0$) and at 2ω were observed on the full beam; this indicates that the beam was being chopped by plate motion. (b) Resonance signals were observed at 2ω in various alkali atoms with $E_{dc} = 0$ and the same ac voltage applied to both plates. These signals were found to be proportional to the square of the ac voltage; also, the ratios of signals for various alkali atoms were found to be in the ratios of their respective g_F values. These signals are believed to have been produced by pole-piece motion caused either by electrostrictive effects in the Teflon sheets or glass spacers in the plate structure, or by electrostatic forces on the field plates or pole pieces. The plate structure is wedged tightly between the pole pieces (see Fig. 3), and any small expansion or contraction of the structure when the ac voltage was applied could have been transmitted to the pole pieces. Attempts were made to reduce the effect by changing the spacers and the insulators and making the entire plate-magnet assembly more rigid. The effect was changed but never completely eliminated.

A small linear signal at ω was also observed when $E_{dc} = 0$ and the same ac voltage was applied to both plates. However, it is assumed that this signal was caused by either a conduction current or a motional magnetic field effect which arose in some way from the fringing electric field. Effects arising from the harmonic content in the output of the high voltage transformers are excluded as a cause of this signal. The reason for this is that the observed signal is linear in the applied voltage, whereas if harmonic effects are important the signal would exhibit nonlinearity because E_2 is not constant (see Sec. V C).

Although pole-piece motion could have affected the quadratic Stark measurements,[18] it is difficult to see how pole-piece motion could produce an effect at ω when $E_{dc}=0$ because the plate motion depends quadratically on the applied voltages. One possibility is that the harmonics in the high voltage are mixed by the quadratic electric force to give a component of pole-piece motion at ω. However, this effect is not important for the following reason. The signals observed in Na and K were found to be linear in the applied electric field. If harmonic effects are important, the signals would be nonlinear because E_2 is not constant (see Fig. 12).

A completely new plate structure situated in a Helmholtz coil C-field region is being developed which should eliminate problems associated with plate motion in subsequent experiments.

F. Magnetic Pickup Effects

At an early stage of the experiments, magnetic fields from the high-voltage transformer were observed to modulate the resonance at ω. The transformer was moved sufficiently far away so that this effect was eliminated. Also, extensive tests were made to check for other possible sources of pickup on the resonance. None were found. Although several sources of pickup in the detection electronics were discovered and eliminated, the method of taking data, which involves moving from one side of the resonance to the other and observing the difference signal, cancels out any pickup of this type.

Tests were also made to check whether the electric field in some way causes spurious frequency modulation of the rf oscillator. No such effects were found.

G. Effects of Spurious dc Voltage

A spurious dc bias across the electric field plates produced perhaps by some rectification of the high-voltage ac could have given rise to a linear shift in two ways. As seen from Eq. (16), the presence of a spurious E_{dc} would cause a quadratic Stark shift signal modulated at ω. However, such an effect would be largest in Cs and smallest in Na and K,[18] whereas the linear shifts are actually smallest for Cs and Rb^{85} and largest for Na and K.

Another way in which the effect of a spurious E_{dc} might have caused a linear shift can be seen from Eq. (22). Substituting $H_1=aE$, where $E=E_0\sin\omega t+E_{dc}$, the component of the resultant magnetic field modulated at ω is then

$$H_R(\omega)\sim\left[a\cos\phi+\frac{a^2\sin^2\phi}{H_0}E_{dc}\right]\sin\omega t\,. \qquad (27)$$

Attempts to measure spurious E_{dc} were made by observing the dc voltage drop across a resistor placed in series with the electric field plate when ac was applied to the plate. We established easily that $E_{dc}<0.005E_0$. (Contact potential effects are considered to be even less important.) Therefore

$$\frac{a^2\sin^2\phi}{H_0}E_0E_{dc}<\frac{a^2\sin^2\phi}{H_0}(0.005E_0^2). \qquad (28)$$

The magnitude of $a^2\sin^2\phi$ can be determined indirectly by making use of some results mentioned in Sec. V C. We note that the term

$$\frac{a^2\sin^2\phi}{H_0}E_0E_{dc}$$

is of similar form to the term

$$\frac{a^2\sin^2\phi}{2H_0}E_0E_2$$

of Eq. (25) with the difference that $\frac{1}{2}E_2$ replaces E_{dc}. As mentioned in Sec. V C, no evidence of the E_0E_2 term was observed for Na and K. A rough estimate of its upper limit for Na is

$$\frac{\mu_0 g_F a^2\sin^2\phi}{2hH_0}E_0E_2<0.05\text{ cps}. \qquad (29)$$

In the range of high voltages applied to the plates in the present experiments (10^4 V/cm $\lesssim E_0\lesssim 5\times10^4$ V/cm) the corresponding range of magnitude of E_2 was $0.01E_0\lesssim E_2\lesssim 0.1E_0$ (see Fig. 12). If we choose $E_2\sim 0.01E_0$, to make a worst-case argument in the following discussion, we have

$$\frac{\mu_0 g_F a^2\sin^2\phi}{2hH_0}(0.01E_0^2)\lesssim\frac{\mu_0 g_F a^2}{2hH_0}\times\sin^2\phi E_0E_2<0.05\text{ cps}. \qquad (30)$$

Comparison of Eqs. (28) and (30) shows that

$$\frac{g_F\mu_0 a^2\sin^2\phi}{2hH_0}E_0E_{dc}<\frac{\mu_0 g_F a^2}{2hH_0}\times\sin^2\phi E_0E_2<0.05\text{ cps}. \qquad (31)$$

Consequently, magnetic effects at ω due to spurious dc are less than those produced by harmonic mixing, and the latter were found to be negligible.

VI. COMPARISONS BETWEEN DIFFERENT ALKALIES

A. Normalization Against g_F

The values of k_2 for each of the alkali atoms are presented in Fig. 16 (filling factor not included). They are analyzed for a possible magnetic effect in the following way. A shift δf in the resonance caused by the interaction of the magnetic dipole moment of the atom with a magnetic field ($\vec{H}$) is

$$\delta f=g_F\mu_0 H/h\,.$$

Assume that the linear shifts are entirely magnetic in origin and that the magnetic field is proportional to the electric field with no dependence

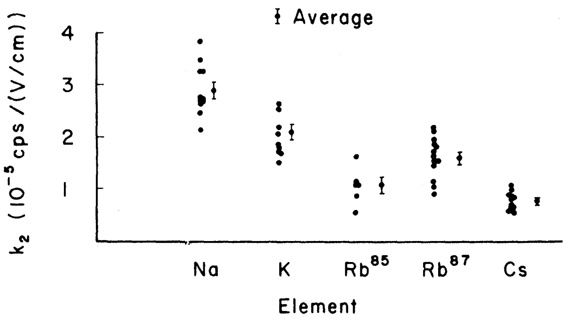

FIG. 16. Plot of values of k_2 for alkali atoms. The data are uncorrected for the filling factor. The error bars indicate the standard deviation of the mean.

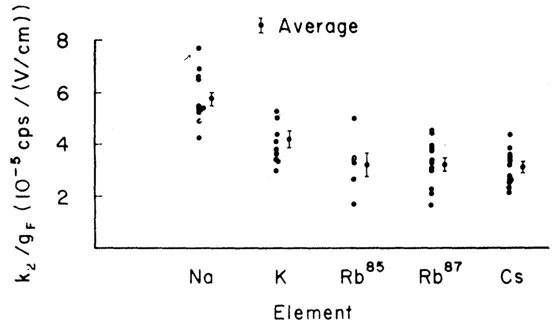

FIG. 17. Plot of values of k_2/g_F for alkali atoms. The data are uncorrected for the filling factor. The error bars indicate the standard deviation of the mean.

of the magnetic field on the velocity of the atom. Then the ratio of shifts for elements 1 and 2 is

$$\frac{\delta f(1)}{\delta f(2)} = \frac{g_F(1)}{g_F(2)} = \frac{k_2(1)}{k_2(2)},$$

so that

$$\frac{k_2(1)}{g_F(1)} = \frac{k_2(2)}{g_F(2)}. \qquad (32)$$

In Fig. 17 the values of k_2 divided by g_F for each of the alkali atoms are shown together with the final averaged values (filling factor not included). The error is the standard deviation δf the mean. If a magnetic interaction, in which the magnetic field is proportional to the electric field with no dependence on the atom's velocity, is entirely responsible for the linear shifts, then Eq. (32) would hold and the values of k_2/g_F for the alkali atoms would agree with each other. As evidenced in Fig. 17, the agreement is poor.

B. Normalization Against g_F and Velocity

The possibility that the linear shifts are due only to a motional magnetic field is now examined. The motional magnetic field is

$$H_1 = (vE/c)\sin\gamma\ , \qquad (33)$$

where $\gamma \sim \frac{1}{2}\pi$. Since $E = E_0 \sin\omega t$, the resulting shift in the resonance is

$$\delta f = (g_F\, \mu_0/hc) v E_0 \sin\gamma \cos\alpha\ , \qquad (34)$$

where α is the angle between $\vec{H}_1$ and $\vec{H}_0$. Taking $v \propto (T/m)^{1/2}$, where T is the temperature in °K and m is the mass of the atom, the ratio of the shifts for two elements is

$$\frac{\delta f(1)}{\delta f(2)} = \frac{g_F(1)}{g_F(2)} \frac{(T_1/m_1)^{1/2}}{(T_2/m_2)^{1/2}} = \frac{k_2(1)}{k_2(2)}\ ,$$

so that

$$\frac{k_2(1)}{g_F(1)}\left(\frac{m_1}{T_1}\right)^{1/2} = \frac{k_2(2)}{g_F(2)}\left(\frac{m_2}{T_2}\right)^{1/2}\ . \qquad (35)$$

Figure 18 shows the values $(k_2/g_F)(m/T)^{1/2}$ for each of the alkali atoms (filling factor not included). These values are listed in Table IV (filling factor included). The uncertainty indicated by the straight error bars is the standard deviation of the mean. The curly error bars are for presentation purposes only and represent the change expected in $(k_2/g_F)(m/T)^{1/2}$ for a temperature variation of 100°C (the actual temperature uncertainty in the present experiments is estimated to be about 20°C). It is seen that the data conform better to a horizontal straight line here than when velocity independence is assumed (see Fig. 17). This agreement is especially good for Na and K for which EDM interactions are expected on theoretical grounds to be smallest, i.e., assuming an EDM of the atom is due to that of the electron – (see Table II and Sec. VII A). Also, as noted earlier, the Na and K data are not complicated by harmonic mixing effects. Because of the excellent agreement between the Na and K results using the motional magnetic field analysis, the discussion that

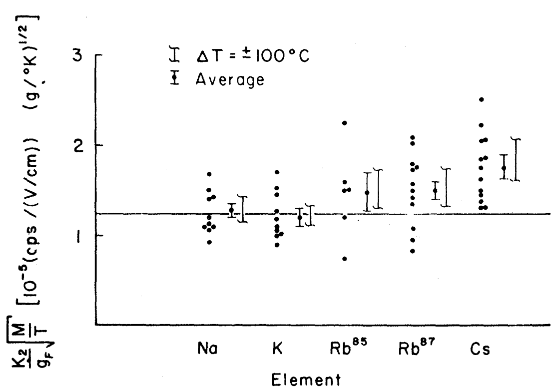

FIG. 18. Plot of values of $(k_2/g_F)(m/T)^{1/2}$ for alkali atoms. The data are uncorrected for the filling factor. The straight error bars represent the standard deviation of the mean. The curly error bars indicate the change in $(k_2/g_F)(m/T)^{1/2}$ for a temperature change of 100°C.

TABLE IV. Values of $(k_2/g_F)\,(m/T)^{1/2}$ for each alkali atom and their deviations from the Na – K average.[a]

Element	$(k_2/g_F) \times (m/T)^{1/2}$ [b]	$(k_e/g_F) \times (m/T)^{1/2}$ [b, c]	k_e [d]
Cs	2.32 ± 0.16[e]	0.71 ± 0.63[f]	0.31 ± 0.27[f]
K	1.60 ± 0.12	0.03 ± 0.52	0.04 ± 0.77
Na	1.69 ± 0.09	0.07 ± 0.57	0.15 ± 1.29
Rb^{85}	1.97 ± 0.28	0.35 ± 0.60	0.25 ± 0.43
Rb^{87}	2.01 ± 0.15	0.39 ± 0.60	0.41 ± 0.64

[a] These values have been corrected for the filling factor.
[b] In units of 10^{-5}cps $(\text{V/cm})^{-1}(g/{}^\circ\text{K})^{1/2}$.
[c] k_e is defined by

$$\frac{k_e}{g_F}\left(\frac{m}{T}\right)^{1/2}(\text{element}) \equiv \frac{k_2}{g_F}\left(\frac{m}{T}\right)^{1/2}(\text{element}) - \left\langle \frac{k_2}{g_F}\left(\frac{m}{T}\right)^{1/2}\right\rangle_{\text{Na}-\text{K}} \text{ average.}$$

[d] In units of 10^{-5}cps/(V/cm).
[e] The uncertainty is the standard deviation of the mean.
[f] The uncertainty is the standard deviation of the mean obtained from a calculation which includes the standard deviation of the mean of the element under consideration and the standard deviation of the mean of the Na – K average.

follows is based on the assumption that the only observable instrumental effect present is that caused by a motional magnetic field.

VII. INTERPRETATION OF RESULTS

A. Limits to the EDM's of Alkali Atoms

The results of the previous section (VI B) can be used to set limits to alkali EDM's due to an electron EDM.[19]

The EDM interactions in Na and K are assumed to be zero (on the scale of sensitivity achieved in the present experiments) – see Table II. Their $(k_2/g_F)(m/T)^{1/2}$ values are then averaged together. Deviations of the $(k_2/g_F)(m/T)^{1/2}$ values for each of the alkali atoms from this average are obtained and are listed in Table IV under the heading k_e (filling factor included). The error is the standard deviation of the mean obtained from a calculation which includes the standard deviation of the mean of of the element under consideration and the standard deviation of the mean of the Na–K average.

As suggested by the values of k_e for Rb and Cs, these elements appear to exhibit effects which cannot be taken completely into account by the simple $\vec{v}\times\vec{E}$ analysis performed here. For the purpose of presentation, these differences are assumed to be due to an EDM in the Cs and Rb atoms producing observable effects [much in the manner outlined earlier in Sec. V; see Eq. (17)]. The results of this analysis are presented in Table V. If these numbers are taken as a measure of an upper limit to the EDM of the Rb and Cs atoms, the following reservations must be kept in mind. The differences in k_e for Cs and Rb could be due to one or more of the following: (a) The manner in which the $\vec{v}\times\vec{E}$ effect was taken into account was not adequate, (b) the measured linear signal may have been the vector resultant of a $\vec{v}\times\vec{E}$ produced signal combined with another instrumentally produced signal of unknown origin or with one previously discussed but not adequately estimated, and (c) a cancellation between a true EDM-produced signal and some instrumentally produced signal. Experimental accuracy was not sufficiently good in the present experiments to pursue these matters in more detail. A new C-field region is under development which should allow us to undertake a careful analysis of the $\vec{v}\times\vec{E}$ effect and eliminate the effects of pole-piece motion.

Note that on the basis of our assumption of a zero EDM interaction in Na and K, the EDM limits for Rb and Cs cannot be larger than the limits quoted in Table V. The reason for this is that the phases of the linear signals were carefully measured and all were found to have the same sign. Therefore, assuming zero EDM interactions in Na and K, whatever the source of the signal producing effects, they all had to be of the same sign; otherwise the signals would have been out of phase with each other or the final averaged values of $(k_2/g_F)(m/T)^{1/2}$ for Rb and Cs would have to be smaller than the Na–K mean (see Fig. 18). Thus, in using k_e to determine a limit to the EDM of the Cs (Rb) atom it was correct to take the *difference* between the Cs (Rb)$(k_2/g_F)(m/T)^{1/2}$ value and the Na–K mean (not the sum).

B. EDM of the Free Electron

From Table II we see that the calculations of Ref. 14 predict that the EDM of the Cs atom should be ~133 times greater than the EDM of the free electron. If we interpret the result in Table V as a possible EDM in the Cs atom and assume the validity of the calculated enhancement factor for Cs, we obtain for the EDM of the free electron

$$D \sim (3.8\pm3.3)\times10^{-22}\ \text{cm}.$$

This value is some six orders of magnitude lower than that quoted in Table I.

Our new limit on the EDM sets a very stringent restriction on any violation of P and T in the

TABLE V. Proposed limits on the EDM's of some alkali atoms.[a]

Element	$D(10^{-20}\ \text{cm})$
K	$0.0\pm\ 6.3$[b]
Na	0.0 ± 12.1
Cs	$5.1\pm\ 4.4$
Rb^{85}	$3.1\pm\ 5.3$
Rb^{87}	$3.8\pm\ 5.8$

[a] These limits are based on the assumption that the assumed EDM effect arises from that of the electron.
[b] The uncertainty is the standard deviation of the mean obtained from the standard deviation of the mean of k_e.

electromagnetic interactions of electrons. On dimensional grounds the EDM of the electron resulting from such a P and T violation should be of order $e\times$ (Compton wavelength), i.e., $e\times(2\times10^{-10}$ cm). Our limit is smaller than this by more than ten orders of magnitude.

On the other hand, the limit is not quite sensitive enough to be a useful test for T violations involving the weak interactions. Here dimensional arguments would suggest a value for the EDM of order $Gm_e e \sim 10^{-22} e$ cm, where G is the weak-coupling constant, m_e is the mass of the electron, and we have used units of $\hbar = c = 1$. A theory which predicts a value of this magnitude has been suggested by Arbuzov and Filippov.[20] A second model which predicts an EDM for the electron of this order of magnitude has been put forw rd by Salzman and Salzman.[21] It is based on the assumption that the P and T violations are due to the electromagnetic interactions of the intermediate vector boson. Wu[22] has argued that such a P- and T-violating interaction follows from the assumption that the interaction should be minimal. The value of the electron EDM to be expected on the basis of this model is $\sim 2\times10^{-24} e$ cm.

C. Purity of the Cs Ground State

An alternative way of looking at our results is in terms of the purity of the Cs ground state. If some p state is present so that the wave function takes the form $|s\rangle + a|p\rangle$, the EDM of the atom is given by

$$\mu = eD = -e\,|\langle s|\vec{r}\,|p\rangle|\,(a+a^*)\,,$$

where we have adopted the phase connection in which the matrix element of $\vec{r}$ is real. We assume that any admixture is due to an internal interaction in the atom. Such an interaction must violate parity in order to admix a p state into an s atate. It must also violate time-reversal invariance in order to ensure that the admixture coefficient a has a nonvanishing real part; otherwise the dipole moment will vanish.

We obtain a rough value for $|\langle s|\vec{r}\,|p\rangle|$ by writing the polarizability α as

$$\alpha \sim 2e^2|\langle s|\vec{r}\,|p\rangle^2|/\Delta W\,,$$

where ΔW is the energy separation between the ground state and the first excited p state. Then

$$\mathrm{Re}\,a \lesssim \tfrac{1}{2} D(2e^2/\alpha\Delta W)^{1/2}\,.$$

Attributing the admixture to an interaction between an electron EDM and the central field of the atom, we can use our determined limit on the EDM of the Cs atom (see Table IV) to deduce that

$$\mathrm{Re}\,a \lesssim 1.4\times10^{-12}\,,$$

where we have used[15]

$$\alpha = 48\times10^{-24}\ \mathrm{cm}^3$$

and also[23]

$$\Delta W = 11\,500\ \mathrm{cm}^{-1}\,.$$

The weak interactions can produce admixtures of the same order of magnitude as our result. In particular, if Carhart's[24] calculation for the admixture of some $2p$ state into the s state of hydrogen is applied to Cs, we find $|a|\sim 10^{-12}$. Carhart's theory is time-reversal invariant, so that a is imaginary and $a+a^*=0$; thus the EDM is zero. However, the fact that a parity violation in the weak interactions can produce admixtures of the order of the upper limit determined in the present experiments suggests that if time-reversal invariance is violated in the weak interactions, an EDM might result whose magnitude could be measured by an atomic-beam experiment.

VII. CONCLUSION

As mentioned earlier, steps are being taken both to minimize instrumental effects and to permit exploration of them in more detail than was accomplished in the experiments described here. In particular, a longer resonance region and refinements in the detection electronics and modulation scheme involving switched dc are being developed.[25] The new C field is comprised of two pairs of rectangular Helmholtz coils mounted at right angles to each other on a cylindrical form enveloping the electric field plates and loops. Adjustment of the current through one pair of coils allows us to vary electronically the angle between the C field and the electric field and thus manipulate the $\vec{v}\times\vec{E}$ effect. The entire resonance region is magnetically shielded by three concentric Hipernom cylinders to minimize noise due to fluctuations in the ambient field. By going over to this type of region, it will be possible to avoid the problems associated with pole-piece motion.

By using digital signal-processing techniques which involve reversing periodically the electric field and accumulating the resonance signal in two counters gated synchronously with the switching frequency, problems associated with harmonic content in the output of the HV transformer are avoided. Also, by introducing the appropriate delays into a counting cycle, a steady electric field is insured during the time data are taken. This eliminates displacement-current effects.

Aside from reducing the magnitude of instrumental effects, we hope to improve the experiments by increasing the sensitivity. This can be done by decreasing the linewidth obtained by increasing the rf loop separation and using a longer integration time.

A reasonable estimate of what sensitivity we might approach can be made. Experiments running with resonance heights corresponding to 10^{-8} A or 10^{11} atoms/sec have been performed with little or no indication that beam noise was worse than that present in the experiments described here.

If we assume a reasonable counting time (integration time) of a few hours and a resonance linewidth of a few hundred cps, then we should expect a sensitivity of the order of 10^{-3} cps or better. At 10^5 V/cm this would correspond to an EDM upper-limit sensitivity of $10^{-22}e$ cm for Cs or, using the Cs enhancement factor, $10^{-24}e$ cm or better for the electron.

The problem, of course, with this argument is that the noise is not random, because of low-frequency detector, beam, and resonance noise. Successful runs at 10^{-8} A resonance heights were accomplished using 25 cps sinusoidally modulated HV and ac phase-sensitive techniques. We do not know to what extent low-frequency noise will bother us when we go over to the switched dc technique. However, in a recent paper[26] describing measurements using the above-described new system (rf loop separation ~ 10 in. giving a linewidth of ~ 850 cps, typical resonance heights of ~ 10^{-9} A, and an integration time of about 7 h), a new upper limit $(2.0 \pm 0.6) \times 10^{-21}e$ cm to the EDM of the Cs atom was reported; the new upper limit to the EDM of the electron is then $(1.7 \pm 0.5) \times 10^{-23}e$ cm. The details of these more recent experiments will be published shortly.[27] Several other methods have also been used to take $\vec{v}\times\vec{E}$ into account in EDM experiments on the Cs atom. Thornburg and King,[28] using a square-wave phase-modulation technique to eliminate $\vec{v}\times\vec{E}$, found a Cs EDM limit of $(0.8 \pm 8.0) \times 10^{-20}e$ cm. In a recent experiment by Angel *et al.*, $\vec{v}\times\vec{E}$ was taken into account by comparing results from beams in opposite directions.[29] They found a limit $(9 \pm 10) \times 10^{-21}e$ cm for Cs.

Experiments are also being conducted on other elements. In particular, Li is being investigated because the $\vec{v}\times\vec{E}$ effect is larger in Li than in other alkali atoms. (Li is much lighter, and the temperature at which a Li beam is formed is higher.) Also, as seen from Table II, its shielding factor is much smaller. Thus Li would serve as a better element for measuring instrumental effects as experimental sensitivity is improved; if the electron posseses an EDM, then the EDM of Cs is expected to be approximately 30 000 times larger than that of Li. Tl is also being investigated because of the possibility that it may have a larger S. Also, it has a P ground state which may make it a candidate for possessing an EDM.[30]

ACKNOWLEDGMENTS

The authors wish to acknowledge the contributions of the Brandeis University machine shop, under the supervision of Arthur Larsen, in the construction of the apparatus, and of Dr. J. Fisher of the American Dynamics Co. (Cambridge, Mass.) for the generous loan of Princeton Applied Research equipment and for hours of discussion concerning phase-sensitive detection techniques. One of us (J. P. C.) would like to thank the Bendix Research Laboratories for making available their facilities during the final stages of preparation of this paper.

---

*Work supported by National Science Foundation

† Holder of Woodrow Wilson National Fellowship. Present address: General Science and Technology Laboratory, Bendix Research Laboratories, Southfield, Mich.

‡ Permanent address: Clarendon Laboratory, Oxford University, England.

§ Present address: Department of Physics, University of Washington, Seattle, Wash.; Holder of National Defense Educational Act Fellowship.

[1] T. D. Lee and C. N. Yang, Brookhaven National Laboratory Report No. BNL-443 (F91), 1957 (unpublished).

[2] J. H. Christenson, J. W. Cronin, V. L. Fitch, and R. Turlay, Phys. Rev. Letters 13, 138 (1964).

[3] P. D. Miller, W. B. Dress, J. K. Baird, and N. F. Ramsey, Phys. Rev. Letters 19, 381 (1967).

[4] C. G. Shull and R. Nathans, Phys, Rev. Letters 19, 384 (1967).

[5] V. W. Cohen, E. Lipworth, R. Nathans, N. F. Ramsey, and H. B. Silsbee, to be published.

[6] R. E. Rand, Phys. Rev. 140, B1605 (1965).

[7] E. S. Ensberg, Bull. Am. Phys. Soc. 7, 534 (1962).

[8] E. S. Ensberg, Phys. Rev. 153, 36 (1967).

[9] P. G. H. Sandars and E. Lipworth, Bull. Am. Phys. Soc. 9, 91 (1964).

[10] E. Lipworth and P. G. H. Sandars, Phys. Rev. Letters 13, 716 (1964); P. G. H. Sandars and E. Lipworth, *ibid.* 13, 718 (1964).

[11] E. Lipworth, A. Adler, J. P. Carrico, T.S. Stein, and P. G. H. Sandars, Bull. Am. Phys. Soc. 11, 403 (1966).

[12] P. G. H. Sandars, Proc. Phys. Soc. (London) B1, 499 (1968).

[13] L. I. Schiff, Phys. Rev. 132, 2194 (1963).

[14] P. G. H. Sandars, Phys. Letters 14, 194 (1965); 22, 290 (1966). See also E. E. Salpeter, Phys. Rev. 112, 1642 (1958); M. Sachs and S. Schwebel, Ann. Phys. (N. Y.) 6, 244 (1959); 8, 475 (1959).

[15] G. E. Chamberlain and J. C. Zorn, Phys. Rev. 129, 677 (1963).

[16] N. F. Ramsey, Molecular Beams (Oxford University Press, London, 1956).

[17] R. D. Haun, Jr., and J. R. Zacharias, Phys. Rev. 107, 107 (1957).

[18] J. P. Carrico, A. Adler, M. Baker, S. Legowski, E. Lipworth, T. S. Stein, M. C. Weisskopf, and P. G. H. Sandars, Phys. Rev. 170, 64 (1968).

[19] Because of the lack of a priori knowledge concerning the size of the $\vec{v}\times\vec{E}$ effect, the alternative to this approach would be to determine limits to the differences between atomic EDM's. This is not pursued here.

[20] B. A. Arbuzov and A. T. Filippov, Phys. Letters 20, 537 (1966).

[21] F. Salzman and G. Salzman, Nuovo Cimento 41, 443 (1966).

[22] T. T. Wu, Phys. Rev. Letters 15, 715 (1965).

[23] C. E. Moore, Atomic Energy Levels, Natl. Bur. Std. (U. S.) Circ. No. 467, (1958).

[24] R. A. Carhart, Phys, Rev. 132, 2337 (1963).

[25] T. S. Stein, J. P. Carrico, E. Lipworth, and M. C. Weisskopf, Bull. Am. Phys. Soc. 12, 508 (1967).

[26] T. S. Stein, J. P. Carrico, E. Lipworth, and M. C. Weisskopf, Phys. Rev. Letters 19, 741 (1967).

[27] T. S. Stein, J. P. Carrico, E. Lipworth, and M. C. Weisskopf, to be published.

[28] C. O. Thornburg, Jr., and J. G. King, Bull Am. Phys. Soc. 11, 329 (1966).

[29] J. R. P. Angel, P. G. H. Sandars, and M. H. Tinker, Phys. Letters 25A, 160 (1967).

[30] M. Sachs and S. Schwebel, Ann. Phys. (N. Y.) 8, 475 (1959); M. Sachs, *ibid.* 6, 244 (1959); Phys. Letters 14, 302 (1965).