Permanent Electric Dipole Moment of the Cesium Atom. An Upper Limit to the Electric Dipole Moment of the Electron*

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An atomic-beam magnetic-resonance apparatus has been employed to search for a linear Stark effect on the flop-in Zeeman transition $(F=4, m_F=-4) \longleftrightarrow (F=4, m_F=-3)$ of the ground state of the cesium atom. Such an effect could be interpreted in terms of a permanent electric dipole moment (EDM) of the cesium atom. The existence of an EDM in a nondegenerate physical system having well-defined angular momentum, such as the cesium atom in its ground state, would be direct evidence of a violation of parity and time-reversal invariance. To detect a possible linear Stark effect, a voltage of 10 kV is applied across two parallel metal plates located between the rf loops of a Ramsey double-hairpin structure. A beam of cesium atoms passes between the metal plates, where it is subjected to an electric field of approximately 5×10^4 V/cm. The flop-in Zeeman transition is observed for a magnetic field of 0.95 G. The signal generator which drives the rf loops is adjusted so that the detector signal corresponds to the point of maximum slope on one side of the central peak of the two-loop interference pattern. The direction of the electric field is reversed every 0.57 sec, and the resonance signal is accumulated in two counters (one for each direction of the electric field), gated synchronously with the electric field switching frequency. Thus, the quadratic Stark effect contributes the same signal to each counter, and a difference between the counters indicates the presence of a resonance shift linear in E. With the apparatus used in the present experiments, resonance shifts as small as 2 or 3 parts in 10^6 of the linewidth (850 cps in cesium) could be observed with about 2 h of integration time. A linear resonance shift which simulates a linear Stark effect can be caused by 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 $\vec{E}$. To separate this motional magnetic field effect from a possible linear Stark effect, the resonance shift linear in E is studied as a function of the angle between $\vec{E}$ and $\vec{H}$, and the angles corresponding to zero resonance shift are obtained in sodium and cesium. A difference between these angles for sodium and cesium would indicate the presence of a linear effect other than $\vec{v}\times\vec{E}$. The magnitude of this difference can be used to set an upper limit on the linear Stark effect in cesium. A relativistic argument shows that if the electron possesses an EDM, the EDM of the Cs atom is approximately 119 times as large, whereas that of the sodium atom is 0.3 times as large. These results, together with the measured values of the angles which correspond to zero resonance shifts in Na and Cs, lead to the following upper limits on the EDM's of the Cs atom and the free electron: $\mu_{\mathrm{Cs}}/e<3\times10^{-21}$ cm and $\mu_e/e<2.5\times10^{-23}$ cm.

I. INTRODUCTION

In 1964, Christenson, Cronin, Fitch, and Turlay[1] reported their observation of the CP-violating $K_2^0\to2\pi$ decay. The occurrence of this decay together with the CPT theorem[2,3] implies that time-reversal invariance is violated in this process. This fact has motivated several groups of researchers to conduct direct experimental tests of T invariance. As yet, no direct experimental evidence of a violation of T invariance has been found.

One area where much effort has been expended is in the search for a permanent electric dipole moment (EDM) of the neutron and the electron.[4] An EDM is detected by its interaction with an electric field $\vec{E}$. Assuming that the EDM is parallel to the spin $\vec{\sigma}$, the interaction can be written as $d\vec{\sigma}\cdot\vec{E}$ (d is a constant with the dimensions of charge length that characterizes the strength of the interaction). The interaction is odd under parity, as

$$\vec{E}\xrightarrow{P}-\vec{E} \quad\text{and}\quad \vec{\sigma}\xrightarrow{P}\vec{\sigma}.$$

Furthermore, the interaction is odd under time reversal, as

$$\vec{E}\xrightarrow{T}\vec{E} \quad\text{and}\quad \vec{\sigma}\xrightarrow{T}-\vec{\sigma}.$$

Therefore, the existence of an EDM would be direct evidence for violations of both parity and time-reversal invariance.[5]

At the time of the observation of the CP violation in 1964, an upper limit to the EDM of the neutron[6] of $\mu_N/e < 5\times10^{-20}$ cm had been reported. The best upper limit for the electron[7] was on the order of 4×10^{-16} cm. Significant improvements on the electron-moment upper limits have come about for two reasons: (i) the development of highly sensitive methods of measuring small shifts due to an electric field in atomic rf resonances,[8] and (ii) a theoretical interpretation by Sandars[9,10] who showed that electric field (Stark shift) measurements on neutral atoms could lead to information on the EDM of the electron.

Sandars and elsewhere Salpeter[11] have shown that as a result of relativistic effects, an assumed EDM in the electrons of an atom will manifest itself as an EDM of the whole atom. In particular, the ratio

$$R \equiv \text{atomic EDM/electronic EDM}$$

has been calculated by different methods[9,10] with the result that for the heavier more relativistic alkalies there is an enhancement of the effects of an electronic EDM, e.g., one finds for cesium $R=119$. A list of the enhancement factors for the alkalies appears later in Table III.

The experiments with atoms involve a search for energy changes (frequency shifts) linear in an externally applied electric field. The presence (or absence) of such "linear Stark shifts" may then be interpreted with Sandars's model as statements about the EDM of the electron. It should be emphasized however, that a linear Stark shift in a nondegenerate atomic system, though evidence for a P and T violation, may arise from causes other than an electron EDM.

Experiments designed to search for a linear Stark shift in the ground state of the Cs atom were first reported in 1964.[8] An atomic-beam magnetic resonance apparatus was used to monitor the transition between the levels $(F=4, m_F=-4)$ and $(4, -3)$ in Cs. The transition was induced in a Ramsey double-hairpin structure in a weak uniform magnetic field $\vec{H}$. A set of parallel electric field plates was located between the rf loops. The rf signal generator was tuned so that the beam detector signal corresponded to the point of maximum slope on one side of the central peak of the Ramsey interference pattern. This assured that a maximum change in the detector signal would be produced by a resonance shift.

The resonance shift $\Delta\nu$ could be written as a function of the applied electric field $\vec{E}$ in the following way:

$$\Delta\nu = k_1 E + k_2 E^2 .$$

The k_2 term represents the quadratic Stark effect and the k_1 term represents a possible linear Stark effect.

An electric field of the form $\vec{E} = \vec{E}_{ac}\sin\omega t + \vec{E}_{dc}$ was applied.

The resonance shift could then be written

$$\Delta\nu = k_1(E_{ac}\sin\omega t + E_{dc}) + k_2[\tfrac{1}{2}E_{ac}^2(1-\cos 2\omega t) + 2E_{ac}E_{dc}\sin\omega t + E_{dc}^2] .$$

A lock-in detector was used to separate the ω and 2ω components.

With E_{dc} set to zero, a small resonance shift was observed at ω which was linear in the applied field. If this effect were attributed entirely to an EDM of the cesium atom, the magnitude of the moment would have been

$$\mu_{Cs}/e = (2.2 \pm 0.1)\times10^{-19}\,\text{cm}.$$

However, as was pointed out at the time, the observed effect could have been easily produced by the motional magnetic field $\vec{v}/c\times\vec{E}$. One can show that if $\vec{v}\times\vec{E}$ is not exactly perpendicular to the homogeneous magnetic field $(\vec{H})$ in the resonance region, a resonance shift, linear in E, is produced by the interaction of the atom's magnetic dipole moment with the motional magnetic field.

That the motional magnetic field effect produced the linear shift was verified in later experiments[12] by studying the effect in several alkalies with different magnetic moments and velocities.

These experiments led to an upper limit to the EDM of the cesium atom of

$$\mu_{Cs}/e \leq 1\times10^{-19}\,\text{cm}.$$

The set of experiments described in the present paper was undertaken to: (i) directly measure the motional magnetic field effect and separate it from possible linear Stark effects, and (ii) substantially reduce or eliminate other systematic effects which could have influenced the upper limit on μ_{Cs} stated above.

II. APPARATUS

Much of the atomic-beam magnetic-resonance apparatus used in this experiment has been described elsewhere in the literature.[12,13] However, several important modifications[14] have been made in order to reduce, eliminate, and directly measure instrumental effects which can simulate a linear Stark shift. These improvements in the original apparatus are described below.

A. *C* Magnet

The C magnet used in the present experiments was designed specifically with a provision for rotating the magnetic field with respect to the electric field in order to separate the motional magnetic field effect from a possible linear Stark shift. This was achieved by winding two independent sets of rectangular Helmholtz coils on a Lucite tube ($6\frac{1}{2}$ in. o.d., 6 in. i.d., $17\frac{3}{8}$ in. long). One set of coils (the H_z coils) produces a horizontal magnetic field $H_z\hat{k}$ parallel to the electric field within 1^0. The other set of coils (the H_y coils) produces a vertical magnetic field $H_y\hat{j}$. Adjustment of the current in the latter pair of coils (the current in the H_z coils is held constant) allows one to vary the alignment of $\vec{E}$ and $\vec{H}$ electronically and thus manipulate the $\vec{v}\times\vec{E}$ effect.

The positions of the H_z and H_y coils and the orientation of the fields produced by these coils is shown in the cross-sectional view of the *C* magnet in Fig. 1. Ten-conductor flat-ribbon cable (Spectra-Strip 1024) was used as magnet wire. To facilitate accurate positioning of the coils, Lucite guides were mounted along the length of the Lucite tube. The cross-sectional view of the *C* magnet in Fig. 1 shows one such set of guides. The guides were sections cut from a $6\frac{1}{2}$ in. i.d., 7 in. o.d. Lucite tube.

The coils were wound in such a way that each of the four H_z (H_y) cable lengths shown in Fig. 1 was actually part of one continuous cable carrying a current I_z (I_y) in each of its 10 wires. The four cable lengths of the H_z (H_y) coils were joined at the ends of the coil form in such a way that magnetic-field contributions from the connecting "end" cables tended to cancel out.

The optimum position of the wires on the coil form to obtain the best magnetic field homogeneity at the beam was determined by a computer program. The calculated inhomogeneities for the best configurations determined by the computer were approximately 6×10^{-5} and 2×10^{-5}, respectively, for H_z and H_y over the height of the beam ($\simeq\frac{1}{2}$ in.). The beam width ($\simeq\frac{1}{16}$ in.) was sufficiently small that it was thought that inhomogeneities over the width would be insignificant.

The measured full widths at half-intensity of the central peaks of the interference patterns for Cs and Na were (850 ± 50) cps and (1900 ± 150) cps, respectively. Other data relevant to the resonance region are listed in Table I.

A Princeton Applied Research model TC-602

TABLE I. Data on *C*-magnet region.

rf loop separation	$10\frac{3}{8}$ in.
rf loop slit width	$\frac{1}{16}$ in.
Length of electric field plates	5 in.
Gap between electric field plates	0.075 ± 0.001 in.
Magnetic field at the position of the beam	$\simeq0.95$ G
"Flop-in" Zeeman transition frequencies of Cs	332 kc/sec
"Flop-in Zeeman transition frequencies of Na	664 kc/sec

power/reference source operated in the constant current mode was used as the power supply for the H_z and H_y coils. The H_y coils were supplied with an adjustable fraction of the current in the H_z coils using the arrangement shown in Fig. 2. The current, I_z in the H_z coils was held constant at 745.7 $\pm$0.4 mA during all of the experiments.

In the experiments, three different values of I_y were used to produce three different alignment angles of the electric and resultant magnetic fields. These three values of I_y were set within 1 mA (1 mA corresponds to an angle of approximately 0.08 deg) about the value of I_y for which the linear signal vanished. The current I_y (and therefore the alignment angel) was determined by measuring the voltage across the standard resistor R_s shown in Fig. 2. This measurement was performed with a Rubicon model No. 2745 potentiometer.

In order to minimize the effect of fluctuations in the ambient magnetic field the *C*-magnet coils are surrounded by three concentric magnetic shields. The shields (James Millen Mfg. Co., Inc., Malden, Mass.) are Hypernom tubes $8\frac{1}{2}$, $10\frac{1}{2}$, and $12\frac{1}{2}$ in. in diameter and are capped at each end. The two inner shields have $\frac{1}{32}$ in. walls. The outer shield has a $\frac{1}{16}$ in. wall. The lengths of the shields are 18, 20, and 22 in., respectively.

B. Electric Field Plates

The electric field plate assembly is shown in Figs. 3 and 4. This structure was centered be-

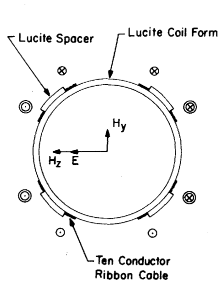

FIG. 1. Cross section of *C* magnet. Coil form o.d. $=6\frac{1}{2}$ in. i.d. $=6$ in. The single circles are nearest the cables which produce the H_z (horizontal) field. The double circles are nearest the cables which produce the H_y (vertical) field. The sense of the current in each coil is indicated in the usual fashion by dots and crosses.

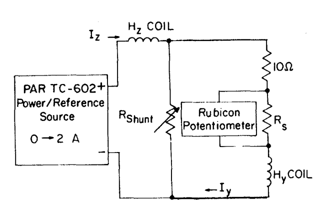

FIG. 2. Current supply for the *C*-magnet coils.

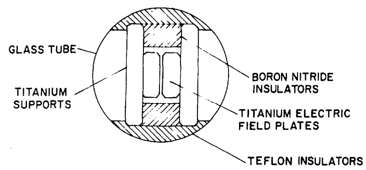

FIG. 3. Electric field plate structure – cross section. Electric field plate length = 5 in., height = 1 in., width = $\frac{3}{8}$ in.

tween the rf loops in the resonance region. The field plates ($5\times1\times\frac{3}{8}$ in.), made of titanium, were surface ground to minimize the possibility of arcing.

Four titanium bars are screwed onto the back of each field plate. These bars are used to attach the field plates to $\frac{1}{2}$ in. o.d. boron nitride spacers. The gap between the field plates is 0.075 ± 0.001 in. The field plate assembly fits tightly inside Teflon spacers.

C. Ovens

Schematic diagrams of the oven design used in the present experiments are shown in Figs. 5 and 6. The ovens were made of copper and produced stable beams of cesium, sodium, and rubidium.[15] The oven slit ($\frac{1}{4}\times\frac{3}{64}$ in.) was kept at a higher temperature than the oven body by winding the heater coil (0.020-in.-diam tantalum wire) in the two channels closest to the slit with three times as many turns per unit length.

Cs and Na beams were formed simultaneously in the same oven by using a mixture of pure Na metal and cesium chloride. $\frac{3}{8}$-in.-diam cylindrical plugs of sodium were cut into disks about $\frac{1}{8}$ in. thick and these disks were distributed uniformly in a charge of CsCl. About 12g of CsCl and 3g of Na metal at a temperature of 350°C was found to produce Na and Cs beams of roughly equal intensities (full beam $\cong 2\times10^{-8}$ A) which lasted for more than 24 h.

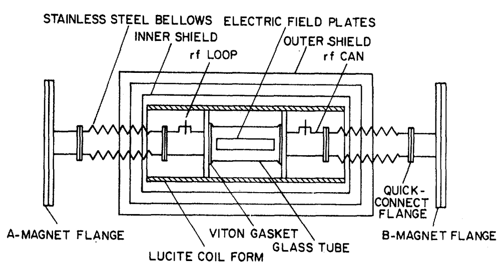

FIG. 4. *C*-magnet region – side view. Distance between rf loops = $10\frac{3}{8}$ in.; length of the electric field plates = 5 in.

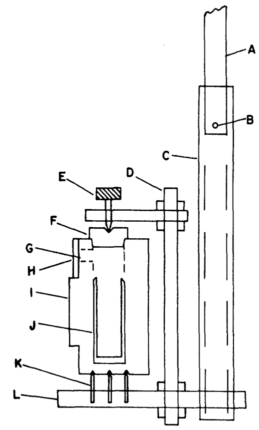

FIG. 5. High-temperature oven and oven support – side view. A. Stainless-steel rod, (can be rotated from outside of the oven vacuum chamber). B. Set screw which fastens the solid shaft to tube, C. Stainless-steel tube ($\frac{1}{8}$-in.-thick wall). D. $\frac{5}{16}$ in. threaded rod. E. Knurled screw used to hold oven cap down on the oven body and to hold the oven in place on the tapered tungsten pins, *K*. F. Oven cap. G. $\frac{1}{4}$-in.-diam beam channel which leads from the oven well (in which an element is placed) to slit, *H*. H. Oven slit (slit width adjustable from 0 to $\frac{3}{32}$ in.). I. Oven body. J. Crucible in which an element is placed. K. Tapered tungsten pins. L. Oven support platform.

D. Detection Electronics

A block diagram of the detection electronics is shown in Fig. 7. The frequency synthesizer (Schlumberger FS-30) which drives the rf loops is adjusted so that the detector signal corresponds to the point of maximum slope on one side of the cen-

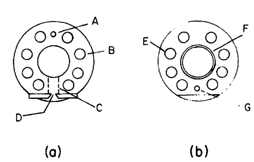

FIG. 6. High-temperature oven: top and bottom views. A. Channel for the thermocouple. B. Heater channel (0.253 in. diameter), lined with alumina tubes (Alsimag 614, American Lava Corporation), extends entire length of oven body, approximately 3 in. long and has $\frac{1}{64}$-in. shoulder at bottom of oven body to prevent alumina insulators from falling out. C. $\frac{1}{4}$-in.-diam beam channel which leads from the oven well to the oven slit. D. Oven slit. E. Bottom of the heater channel; diameter of the opening is $\frac{7}{32}$ in. F. Circular *V*-shaped groove to position oven body on tungsten pins. G. Channel for thermocouple which ends just under the beam channel, *C*.

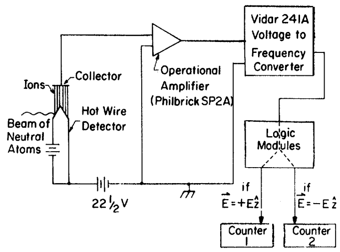

FIG. 7. Block diagram of digital lock-in system.

tral peak of the Ramsey interference pattern. The direction of the electric field is reversed every 0.57 sec and the resonance signal is accumulated in the two counters (one for each direction of the electric field) which are gated synchronously with the electric field. The magnitude of the electric field is held constant while its direction is reversed. Thus, the quadratic Stark effect contributes the same signal to each counter and a difference between the counters indicates the presence of a resonance shift linear in E. Resonance shifts as small as two or three parts in 10^6 of the linewidth (850 cps in Cs) could be observed after about 2 h of integration time.

The timing sequence, generated by logic modules, is shown in Fig. 8. The time between successive time marks is 26 msec. There is a period of 78 msec after the electric field is reversed during which both counters are off. This delay assures that the electric field is at equilibrium when the counters are accumulating data. Since $\partial \vec{E}/\partial t$ is zero during this time, possible systematic effects due to displacement current flowing between the plates are eliminated.

The switching pattern of the counters and of the electric field is arranged as shown in Fig. 8 instead of in a strictly alternating pattern to reduce the contribution to the linear resonance shift signal by a drift in the beam intensity. The arrangement in Fig. 8 will eliminate the effect of a beam intensity which is drifting linearly in time over one complete counting cycle. Fluctuations in the beam intensity are often to a good approximation linear in time over a 1-sec period, the approximate time for one complete counting cycle.

E. High-Voltage Network

The voltage applied to the electric field plates is supplied by two Northeast Scientific Co. (Acton, Mass.) model RE-10010 regulated high-voltage supplies. A Hipotronics, Inc. oil-filled 20-kV DPDT relay is used to reverse the electric field. The maximum switching time of the relay used is about 27 msec. The high-voltage switching signal is obtained from the logic circuitry of the digital lock-in system.

III. EXPERIMENTAL PROCEDURE

The atomic-beam magnetic-resonance apparatus is used to monitor the flop-in[16] Zeeman transition

$$(F=I+\tfrac{1}{2},\ m_F=-I-\tfrac{1}{2}) \longleftrightarrow (I+\tfrac{1}{2},\ -I+\tfrac{1}{2})$$

in the ground state of the alkali atoms. This transition is induced in a magnetic field ($\vec{H}$) of 0.95 G. Voltages of up to 10 kV are applied to the field plates (plate separation is 0.075 in.) centered between the rf loops.

The quadratic Stark signal is eliminated by using the electric-field switching procedure described in Sec. IID (see Fig. 8). Calibration is accomplished by frequency modulation of the synthesizer frequency by a known amount ΔF. This procedure is shown in Fig. 9. The electric field is left on during the calibration in order to account for any distortion of the resonance due to the quadratic Stark effect.

Data were taken on opposite slopes of the resonance curve in order to eliminate any resonance-independent contributions to the detected signal.

From the geometry of the atomic-beam apparatus, it is reasonable to assume that the average velocity $\vec{v}$ of the detected atoms is perpendicular to the average electric field $\vec{E}$ in the resonance region. The resonance shift produced by the motional magnetic field is then given by

$$\Delta\nu(E, \bar{v}, \theta) = (g_F \mu_0 / hc)\bar{v}E\sin\theta \quad , \qquad (1)$$

where $\bar{v}$ is the average velocity of the detected atoms and θ is the effective angle between the electric and magnetic fields.

If $\vec{E}$ and $\vec{H}$ were perfectly aligned ($\theta = 0$), then no motional magnetic field effect would be present. However, the present experiments can detect resonance shifts on the order of 0.005 cps in Na. In order to reduce the contribution from the motional magnetic field effect to less than 0.005 cps, (i.e., within the uncertainty of the present experiments), θ would have to be less than 1.3×10^{-5} rad. The

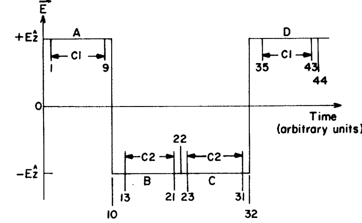

FIG. 8. One complete cycle of digital lock-in system arranged for measuring the linear Stark effect.

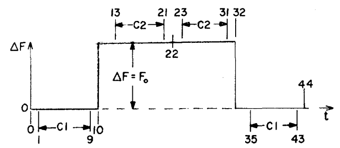

FIG. 9. Frequency modulation pattern.

method used to separate the motional magnetic field effect from a possible linear Stark effect makes use of the fact that the former depends on the alignment angle θ while the latter does not.

In these experiments, the angle θ is kept sufficiently small so that $\sin\theta \sim \theta$. The coils are designed so that

$$\theta = \theta_0 - I_y/I_z, \tag{2}$$

where I_y and I_z are the currents in the vertical and horizontal coils, respectively, and θ_0 is the angle of misalignment between $\vec{E}$ and $\vec{H}$ when $I_y = 0$.

Let δ represent the contribution to the linear signal from other effects, including a possible EDM. Then the total resonance shift per unit electric field, linear in E, can be written

$$k \equiv \Delta\nu(E)/E = m(I_y + I_0) + \delta, \tag{3}$$

where $m = -g_F \mu_0 \bar{v}/hcI_z, \quad I_0 = -I_z\theta_0.$

The signal (k) is studied as a function of I_y (the angle between $\vec{E}$ and $\vec{H}$) and the point of zero signal (I_y axis intercept $\equiv I$) is determined for different elements. The importance of making comparisons between different elements stems from the fact that the initial misalignment (θ_0) of $\vec{E}$ and $\vec{H}$ is not known *a priori*. If only one element is studied, this information is required to determine whether linear effects other than $\vec{v} \times \vec{E}$ are present. In a comparison, however, the initial misalignment subtracts out. If the intercepts are not the same for different elements, then an interaction other than $\vec{v} \times \vec{E}$ (such as an EDM) is also contributing to the signal.

At a fixed value of I_y, data are taken continuously for 20 of the counting cycles shown in Fig. 8. This process is then repeated for approximately 20 min, spent equally on both sides of the resonance curve. The data are then averaged and corrected by the "filling factor"[17] to take into account the fact that the electric field plates do not extend the entire distance between the rf loops.

IV. RESULTS

A typical plot of k versus I_y for the elements sodium and cesium is shown in Fig. 10. Seventeen comparison measurements (11 of which were independent) such as that illustrated in Fig. 10 were made. The results of these measurements are summarized in Table II. A comparison was made between successive Na and Cs runs only if a short time (< 20 min) elapsed between experiments. This was done to minimize effects produced by changes in environmental conditions over the time taken to make a comparison (2 h).

The intercept difference is computed for each Cs-Na comparison and the average of these values is $\langle \Delta I\rangle_{av} = \langle I_{Cs} - I_{Na}\rangle_{av} = 0.076 \pm 0.015$ mA. The error stated above is one standard deviation of the mean based on the 11 independent Cs-Na comparison measurements.

The errors stated for I and m in Table II are standard deviations obtained from a "least-squares" fit of a straight line to each plot of k versus I_y. The magnitudes of these errors for a given run were frequently a reflection of the quality of that run. Thus, if the beam were exceptionally noisy during a run, there was likely to be a larger scatter of the three points about the "best line" for that run. However, there is always the possibility with only three points per run, that a relatively poor run will accidentally place the three points very nearly on a straight line, giving rise to small errors for I and m. For this reason, the stated errors in Table II are not used to weight the runs in obtaining a value for the upper limit on the EDM of the cesium atom. The errors are listed in Table II merely as an indication of how well the points in each run fit a straight line. The distribution of the individual values of ΔI and m is used, rather than the errors on I and m stated in Table II, to assign an error to the upper limit on the EDM of cesium.

In Sec. III, it was shown that m satisfies the equation

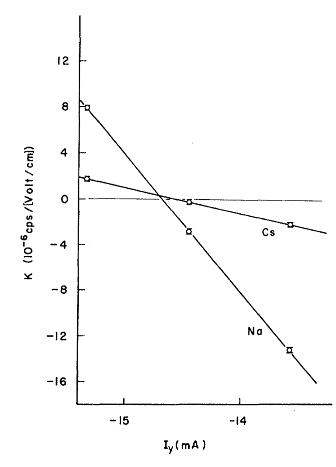

FIG. 10. k versus I_y for Na and Cs. A change of 1 mA in I_y produces a change in the angle between $\vec{E}$ and $\vec{H}$ of approximately 0.08°.

TABLE II. Cs–Na comparison experiments[a]

Element	Run	I (mA)	m [10^{-5} cps/(V/cm) mA]
Cs[a]	101D	−14.560 ± 0.094[b]	−0.2334 ± 0.0213[b]
Na	203D	−14.674 ± 0.020	−1.161 ± 0.0240
Cs	102D	−14.576 ± 0.054	−0.2225 ± 0.0116
Na	204D	−14.682 ± 0.002	−1.183 ± 0.0028
Cs	103D	−14.653 ± 0.035	−0.2336 ± 0.0083
Na	206D	−14.717 ± 0.018	−1.162 ± 0.0216
Cs	105D	−14.736 ± 0.158	−0.2369 ± 0.0417
Na	207D	−14.745 ± 0.012	−1.180 ± 0.0161
Cs	106D	−14.578 ± 0.003	−0.2345 ± 0.0010
Na	209D	−14.680 ± 0.008	−1.204 ± 0.0130
Cs	107D	−14.644 ± 0.025	−0.2290 ± 0.0077
Na	210D	−14.691 ± 0.0004	−1.177 ± 0.0007
Na	211D	−14.696 ± 0.010	−1.121 ± 0.0145
Cs	108D	−14.588 ± 0.037	−0.2581 ± 0.0130
Cs	109D	−14.507 ± 0.084	−0.2247 ± 0.0264
Na	212D	−14.710 ± 0.012	−1.104 ± 0.0170
Cs	110D	−14.604 ± 0.062	−0.1842 ± 0.0154
Na	213D	−14.687 ± 0.030	−1.070 ± 0.0427
Cs	111D	−14.628 ± 0.031	−0.2578 ± 0.0108
Na	214D	−14.709 ± 0.024	−1.189 ± 0.0332
Cs	112D	−14.646 ± 0.0003	−0.2308 ± 0.0001
Na	215D	−14.704 ± 0.020	−1.202 ± 0.0289

[a]Comparisons were made only between those runs connected by brackets.

[b]Errors on I and m are standard deviations obtained from a "least-squares" fit of a straight line to each plot of k versus I_y.

$$m = -g_F \mu_0 \bar{v}/hcI_z .$$

Since $\bar{v}$ is approximately proportional to $(T/M)^{1/2}$, where M is the mass of the element being studied, and T is the temperature of the oven, one would expect the ratio of the experimental m values for Na and Cs to be approximately

$$[g_F(\mathrm{Na})/g_F(\mathrm{Cs})](M_{\mathrm{Cs}}/M_{\mathrm{Na}})^{1/2} \cong 4.8.$$

The ratio of the m values for Na and Cs in each comparison measurement is reasonably close to this value. A detailed discussion of the relationship between the average velocity of atoms in the beam and the value of m is presently being prepared.[18]

V. INTERPRETATION OF DATA

From Eq. (3), we see that the difference between the I_y axis intercepts of two different elements is given by

$$\Delta I = I_2 - I_1 = \delta_1/m_1 - \delta_2/m_2. \qquad (4)$$

If we assume that all systematic effects contributing to δ_1 and δ_2 have been accounted for, then these experiments, which measure ΔI and m_1 and m_2, yield the relative linear Stark shifts between the two atoms. With the further stipulation that δ_1 and δ_2 represent the EDM's of the atoms due to an electron EDM, we may, in a comparison of sodium and cesium, neglect $(\delta_{\mathrm{Na}}/m_{\mathrm{Na}})$ in comparison to $(\delta_{\mathrm{Cs}}/m_{\mathrm{Cs}})$ for the following reasons: (i) Cesium ($Z = 55$) is much more relativistic than sodium ($Z = 11$). Schiff,[19] Salpeter,[11] and Sandars[9,10] have shown that EDM's on electrons only manifest themselves in atoms through relativistic effects. Therefore, one would expect δ_{Na} to be much less than δ_{Cs}. (ii) In the specific cases of Na and Cs Sandars has calculated the effects due to an electron EDM and shown that $\delta_{\mathrm{Cs}} \sim 187\delta_{\mathrm{Na}}$. The results of his calculations of the electric dipole enhancement factors, D_A/D_e = atomic EDM/electron EDM for the alkali atoms, are shown in Table III. (iii) The results of the experiments summarized in Table II indicate that $m_{\mathrm{Na}} \sim 5m_{\mathrm{Cs}}$.

For these reasons, the data will be presented so as to place a value on the quantity δ_{Cs}, the differential linear Stark effect in the cesium atom, given by

$$\delta_{\mathrm{Cs}} = m_{\mathrm{Cs}}(I_{\mathrm{Na}} - I_{\mathrm{Cs}}). \qquad (5)$$

TABLE III. Calculated values of the ratio D_A/D_e for alkali-atom ground states.[a]

Element	D_A/D_e
Li	4.3×10^{-3}
Na	3.18×10^{-1}
K	2.42
Rb	24
Cs	119

[a]Reference 10.

The value of δ_{Cs} as given by Eq. (5) is computed for each Cs-Na comparison and the average of these values is

$$\delta_{Cs} = (1.71 \pm 0.34) \times 10^{-7}\ \text{cps/Vcm}^{-1}$$

The error stated above is one standard deviation of the mean based on 11 independent Cs-Na comparison measurements.

This value for δ_{Cs} is uncorrected for any of the systematic effects discussed in Sec. VI.

VI. SYSTEMATIC EFFECTS

A. Leakage Currents

Small currents produced by the application of high voltage to the electric field plates can flow across the boron-nitride insulators which separate the field plates, or from one or both of the field plates to ground. The sense of these currents is reversed when the electric field is reversed. These currents set up small magnetic fields which are switched synchronously with the electric field. This causes a synchronous modulation of the Zeeman interaction which simulates a linear Stark effect.

To measure these leakage currents, a battery powered electrometer (RCA WV-84C) was placed in series with the cables leading to the electric field plates and the voltage across the plates was set at 10 kV, the maximum voltage used in these experiments. A leakage current of 6×10^{-9} A was measured. From a "worst case" analysis, it was concluded that this leakage current would account for at most 10% of the observed intercept difference.

B. Magnetic Pickup Effects

Any changing magnetic field in the C region which is synchronous with the switching pattern of the counters can simulate a linear Stark effect. Possible sources of such magnetic field changes are: (i) Ambient magnetic field changes due to the synchronous switching of the current which drives the high-voltage relay; (ii) ambient magnetic field changes arising from the synchronized switching of currents in the digital lock-in circuitry; (iii) modulation of the C-magnet power supply due to changes in the line voltage produced by the synchronous switching of the other electronics.

In order to reduce the magnetic field produced by the switching of the high-voltage relay, the relay was mounted in an iron tube having a wall thickness of $\frac{3}{8}$ in. and the ends of the cylinder were covered with $\frac{1}{4}$-in.-thick iron plates. No change in the magnetic field just outside the iron container could be detected when the relay was switched on. A Bell 120 Gaussmeter was used to measure field changes and a change of 0.01 G could have been detected. The relay was placed as far from the C-magnet region as possible.

Two tests for the presence of magnetic pickup effects were conducted: The resonance shifts in sodium and cesium were measured as a function of the electric field with the alignment angle held constant. A magnetic pickup effect would result in a nonzero resonance shift at $E = 0$. Figures 11 and 12 show the results of this test for sodium and cesium, respectively. A small effect was observed in Na. In the second test, experiments were performed on potassium with the electric field turned off. Again, a small effect was observed. The results of both types of tests are summarized in Table IV.

As sodium and potassium have the same g_F values, the agreement between the electric field independent resonance shifts observed in these elements supports the conclusion that they are due to a magnetic effect. For cesium, the same magnetic field change will only produce half as large a resonance shift as that in Na or K. This fact, together with the rather large uncertainty serves to make the cesium test consistent with the Na and K data.

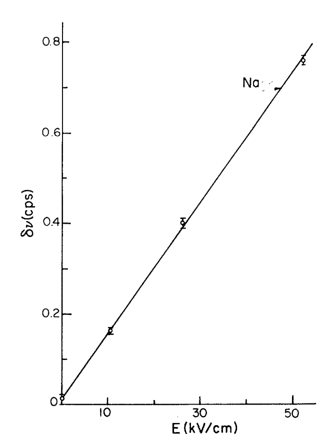

FIG. 11. $\Delta\nu$ versus E for fixed I_y in Na.

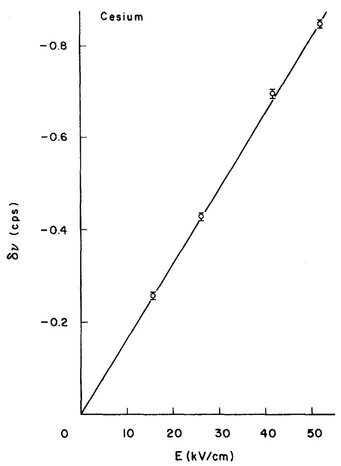

FIG. 12. $\Delta\nu$ versus E for fixed I_y in Cs.

The measurement in K with $E=0$, as it is independent of the instabilities and uncertainties introduced by the application of the high voltage, is used to calculate what fraction of the intercept difference between Na and Cs is due to a magnetic pickup effect. This leads to the following corrected value of δ_{Cs}:

$$\delta_{Cs} = (1.19 \pm 0.39)\times 10^{-7} \text{cps/Vcm}^{-1}.$$

C. Quadratic Mixing Effect

The digital lock-in system makes a measurement of

$$\Delta \equiv \tfrac{1}{2}[\Delta\nu(E_1) - \Delta\nu(E_2)],$$

where $\Delta\nu(E_1)$ is the resonance shift produced by the field $E_1\hat{z}$ and $\Delta\nu(E_2)$ is the resonance shift produced by the field $E_2\hat{z}$ (the field obtained by switching the high-voltage relay). Ideally, $\vec{E}_2 = -\vec{E}_1$ and

$$\Delta = k_1 E_1 .$$

If, however, there is a slight difference in the magnitude of the electric field when its direction is reversed, this difference can couple with the quadratic Stark effect (QSE) to give rise to a signal which is not subtracted out.

An approximate expression ($\Delta E \ll E_1$) for Δ is

$$\Delta = k_1 E_1 + k_2 E_1 \Delta E, \tag{6}$$

where $\Delta E \equiv |E_1| - |E_2|$. If $E_2 \neq -E_1$, Eq. (6) implies that in addition to the linear resonance shift k_1E_1 there will be a term proportional to the QSE which contributes to the signal measured by the digital lock-in system.

A search for the presence of a quadratic mixing effect was made by comparing the I_y axis intercepts in the following cesium transitions: $(F, m_F) = (4,-4) \leftrightarrow (4,-3)$, the "flop-in" Zeeman transition, and the unresolved hfs transitions, $(4,+1) \leftrightarrow (3,0)$ and $(4,0) \leftrightarrow (3,+1)$. The ratio of the QSE on the hfs transitions to that on the Zeeman transition is about 200.[13] The hfs transitions are thus much more sensitive to quadratic mixing than the Zeeman transition. However, the magnetic dependence of the hfs and Zeeman transitions is very nearly identical, so any difference between the intercepts must be due to electric effects.

Two comparison experiments of the type described above were performed. Assuming that any difference between the intercepts associated with the above hyperfine and Zeeman transitions was due to quadratic mixing, these experiments lead to correction terms which must be included in Eq. (4). These experiments lead to the following corrected values, respectively, (including the magnetic pickup effect) for the linear Stark shift (δ) of the cesium atom:

$$\delta_1 = (0.90 \pm 0.40)\times 10^{-7} \text{ cps/V cm}^{-1},$$

$$\delta_2 = (1.38 \pm 0.39)\times 10^{-7} \text{ cps/V cm}^{-1},$$

where subscripts 1 and 2 refer to the two comparison experiments.

These values are to be compared with the value of δ corrected only for the magnetic pickup effect (Sec. VI B),

$$\delta_{Cs} = (1.19 \pm 0.39)\times 10^{-7} \text{ cps/V cm}^{-1}.$$

We see that in each case, quadratic mixing effects are within the experimental error.

D. Trajectory Effects

In Sec. III it was shown that I_0 is proportional to the initial misalignment angle θ_0 between $\vec{E}$ and $\vec{H}$. θ_0 is the effective angle between $\vec{E}$ and $\vec{H}$ when there is no current in the vertical field coil. It is assumed that I_0 (and therefore θ_0) is constant, independent of which element is being studied.

If, however, there were a difference between the

TABLE IV. Results of magnetic pickup tests.

Element	Cs	Na	K
$\Delta\nu(E=0)$ cps	(-0.00421 ± 0.0118)	(0.0130 ± 0.0065)	(0.0088 ± 0.0032)
g_F	$\frac{1}{4}$	$\frac{1}{2}$	$\frac{1}{2}$

initial misalignment angles of Na and Cs of

$$\theta_{0_{Cs}} - \theta_{0_{Na}} \sim 6\times10^{-3} \text{ deg},$$

this would produce an apparent linear Stark effect of the same magnitude as the observed effect. Two conditions must be fulfilled in order for such an effect to occur.

(1) There must be sufficient inhomogeneities in the electric and/or magnetic fields in the resonance region which the beams traverse.

(2) The beams must follow different sets of trajectories through these inhomogeneities in such a way that the effective alignment angle of the fields is different for both elements.

Several tests were made for the presence of such trajectory effects. In one set of tests, comparison measurements were run between K and Na (for which relative EDM effects should be negligible) in different oven ports. These ports were separated by $9\frac{1}{8}$ in. along the beam axis of the apparatus. The first set of comparisons yielded the following weighted mean for the K–Na intercept difference,

$$\Delta I = -0.063 \pm 0.006 \text{ mA}.$$

A second set of comparisons with the ovens interchanged yielded

$$\Delta I = 0.017 \pm 0.009 \text{ mA}.$$

These experiments indicated that intercept differences measured using separate ovens are influenced by trajectory effects arising from different source locations.[20]

Another set of experiments was performed to search for a correlation between the vertical position of the K oven and the intercept. A small intercept change was found when the oven was moved $\frac{1}{4}$ in. above its original position.[20] The difference between intercepts measured in these experiments was 0.048 ± 0.016 mA.

The two tests described above provided the motivation for running both Cs and Na simultaneously from the same oven.

Several experiments were conducted on Na and Cs beams formed in the same oven, to determine whether a change in the I_y axis intercept could be caused by rotating the oven slightly about its symmetry axis. It was felt that such a rotation might produce a small change in the spatial distribution of velocities of atoms in the C magnet. Such a change might be expected to produce an intercept change if trajectory effects were playing an important role in the present experiments.

Comparisons (1–11) were performed with the oven oriented to produce the maximum resonance signal in both Na and Cs. Comparisons (12–14) and (15–17) correspond to clockwise and counterclockwise oven rotations, respectively, which reduced the resonance amplitude by about 20%. The results of these experiments are summarized in Table V. No measurable changes were found in the intercept differences when the oven was rotated. There were also no observable changes in the intercepts of Na and Cs themselves.

If rotating the oven had produced a change in the Cs-Na intercept differences, this would have indicated that trajectory effects might be influencing the present upper limit on the EDM of the Cs atom. However, the fact that no such change was observed does not rule out the possibility that trajectory effects were at least partially responsible for the Cs-Na intercept difference. Thus, for example, it is possible that there existed a difference in the spatial distribution of velocities of the two beams due to the different deflecting forces on each of them, and that the oven was not rotated far enough to change this difference. In the present experiments, further rotation was not feasible due to the further loss of signal. A much more detailed study of possible trajectory effects is presently being conducted in this laboratory.

E. Hysteresis Effects

It is assumed in the analysis of the data, that the magnetic field H_y produced by the vertical field coil is proportional to the current I_y, which flows in this coil. If, however, there were a small hysteresis effect in the magnetic shields surrounding the C magnet, the magnetic field H_y would not "track" I_y perfectly. This could give rise to an intercept difference in a comparison experiment between two elements simply because the value of H_y which aligns $\vec{E}$ and $\vec{H}$ could be produced by a different value of I_y in a Cs experiment than in a Na experiment.

A study of the resonance frequency as a function of I_y in sodium indicated that such effects were well within the uncertainty in the present experiments.

VII. FINAL RESULTS

A. Upper Limits to the EDM of Cs Atom and Electron

The value of δ_{Cs}, corrected for the magnetic pickup effect, leads to the following value for the EDM of the cesium atom[21]:

$$\mu_{Cs}/e = (-2.0 \pm 1.8)\times10^{-21} \text{ cm},$$

where the error is 3 times the standard deviation of the mean. We feel that in view of the relatively

TABLE V. Summary of oven rotation experiments.

Comparison No.	Oven position	ΔI (mA)	m_{Cs} [10^{-5} cps/(V/cm) mA]	δ_{Cs} [10^{-7} cps/(V/cm)]
1	Maximized	-0.114 ± 0.096[a]	-0.233 ± 0.021[a]	2.66 ± 2.25[a]
2	Maximized	-0.098 ± 0.057	-0.223 ± 0.012	2.19 ± 1.29
3	Maximized	-0.106 ± 0.054	-0.223 ± 0.012	2.36 ± 1.21
4	Maximized	-0.029 ± 0.035	-0.234 ± 0.008	0.68 ± 0.82
5	Maximized	-0.064 ± 0.039	-0.234 ± 0.008	1.50 ± 0.91
6	Maximized	$+0.019 \pm 0.159$	-0.237 ± 0.042	-0.45 ± 3.77
7	Maximized	-0.009 ± 0.159	-0.237 ± 0.042	0.21 ± 1.19
8	Maximized	-0.102 ± 0.009	-0.235 ± 0.001	2.40 ± 0.21
9	Maximized	-0.036 ± 0.026	-0.229 ± 0.008	0.82 ± 0.60
10	Maximized	-0.047 ± 0.025	-0.229 ± 0.008	1.08 ± 0.57
11	Maximized	-0.108 ± 0.038	-0.258 ± 0.013	2.79 ± 0.99
12	Rotated *cw*	-0.203 ± 0.085	-0.225 ± 0.026	4.57 ± 1.97
13	Rotated *cw*	-0.106 ± 0.063	-0.184 ± 0.015	1.95 ± 1.16
14	Rotated *cw*	-0.083 ± 0.069	-0.184 ± 0.015	1.53 ± 1.27
15	Rotated *ccw*	-0.081 ± 0.039	-0.258 ± 0.011	2.09 ± 1.01
16	Rotated *ccw*	-0.063 ± 0.024	-0.231 ± 0.0001	1.46 ± 0.55
17	Rotated *ccw*	-0.058 ± 0.020	-0.231 ± 0.0001	1.34 ± 0.47

[a]Standard deviation obtained from a "least-squares" fit of a straight line to each plot of k versus I_y.

limited study of trajectory effects, this result cannot be regarded as definite evidence for the existence of an EDM of the Cs atom but should be used to set the following upper limit:

$$\mu_{Cs}/e < 3\times10^{-21} \text{ cm}.$$

This upper limit together with the enhancement of an electronic EDM calculated by Sandars leads to an upper limit of

$$\mu_e/e < 2.5\times10^{-23} \text{ cm}$$

to the EDM of the electron.

B. Upper Limit to EDM of Cs Atom Based Only Upon Measurements on Cesium

The upper limit on the EDM of Cs stated in Sec. VII A is based upon comparison measurements between Cs and Na and a theoretical calculation by Sandars. It is also possible by using only the data obtained for a single element to determine an upper limit on the linear Stark effect in that element.

The technique used in these experiments involves measuring the intercept for a given element as a function of velocity (proportional to the slope in a k-versus-I_y plot). Consider as an example, the case where two runs are made, resulting in the slopes m_1 and m_2 and in the I_y axis intercepts I_1 and I_2, respectively. From Eq. (3), we see that the intercept difference $I_1 - I_2$ and δ are related by

$$\delta = [m_1 m_2/(m_1 - m_2)](I_1 - I_2) . \quad (7)$$

Thus, if the $\vec{v}\times\vec{E}$ effect is the only linear effect present(and there are no trajectory effects) then

$$I_1 = I_2 \quad \text{and} \quad \delta = 0.$$

The results of the measurements were presented in Table II. In runs Cs-110D and Cs-111D as an example, the errors in the slopes (m) do not cover the difference between the slopes. Recalling that

$$m = -g_F \mu_0 \bar{v}/hcI_z,$$

and that I_z is held constant during all the experiments, the difference between the slopes in runs Cs-110D and Cs-111D must be attributed to a different average velocity $\bar{v}$ in each of these runs. Two possible contributions to a change in $\bar{v}$ are: (i) The oven temperatures may have been different for these experiments. (ii) The oven was intentionally rotated before run Cs-111D was begun. Such a rotation of the oven can produce a change in the average velocity of the detected particles because of velocity selection by the focusing magnets.

The intercept differences in the experiments labeled Cs 108D–Cs 110D and Cs 111D–Cs 110D were used with Eq. (7) above to obtain a value of δ of

$$\delta = (-0.6 \pm 3.2)\times10^{-7} \text{ cps/Vcm}^{-1} .$$

This corresponds to an EDM of the Cs atom of

$$\mu_{Cs}/e = (1.0 \pm 5.3)\times10^{-21} \text{ cm},$$

which is consistent with the value quoted above in Sec. VII A.

C. Purity of Cs Ground State

The result above (Sec. VII A) can be used to set an upper limit to the real part of the probability amplitude of parity mixing[12] in the ground state of the cesium atom. If some p state is present so that the wave function takes the form $|s\rangle + a|p\rangle$, then from our measured limit on the EDM of the Cs atom we deduce that

$$\mathrm{Re}(a) < 10^{-13} \ .$$

VIII. CONCLUSIONS

A search has been made for a linear Stark effect in the ground-state Zeeman transitions in the alkali atoms. Plots of $k = \Delta\nu/E$ versus the current, I_y in the vertical field coils of the C magnet have revealed small differences between the I_y axis intercepts of Cs and Na. Ideally, if the only effect contributing to k in each element were the motional magnetic field effect, the I_y axis intercept would be independent of which element was being studied.

There are several instrumental effects which could have contributed to the observed intercept differences. An independent measurement of one of these instrumental effects (a magnetic pickup effect) has been used to correct the Cs-Na intercept difference. The correction reduced the observed difference by about 30%. Trajectory effects could possibly have been responsible for part or even all of the remaining Cs-Na intercept difference.

Analysis of a number of other possible instrumental effects did not indicate that any of these could have accounted for a significant fraction of the Cs-Na intercept difference.

The results of these experiments have been used to set upper limits to the EDM of the Cs atom and of the electron

$$\mu_{Cs}/e < 3\times10^{-21} \text{ cm},$$

$$\mu_e/e < 2.5\times10^{-23} \text{ cm}.$$

A longer rectangular Helmholtz coil C-magnet region with improved beam collimation is presently being used for experiments in this laboratory. In addition to improving the sensitivity of the experiments,[22] this region should lessen the influence of trajectory effects and allow for a more detailed study of these and other systematic effects.

Note added in proof. It has recently been shown [R. C. Casella, Phys. Rev. Letters 21, 1128 (1968)] that existing data on the CP-nonconserving $K^\circ \to 2\pi$ decays provide direct experimental evidence of a violation of T invariance.

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[21]These experiments are performed in a weak magnetic field so that the total angular momentum F may be considered as a good quantum number. For these transitions ($|\Delta m| = 1$) the relation between δ and μ/e is then given by $\mu/e(\text{cm}) = 300h\delta F/e$ where δ is in units of cps/V/cm), $h = 6.63 \times 10^{-27}$ erg sec, $F = 4$ for the "flop-in" Zeeman transition in Cs, and $e = 4.80 \times 10^{-10}$ esu.

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