Atomic-Beam Measurement of Isotope Shifts in the D_1 Line of ^{127}Cs, ^{129}Cs, ^{133}Cs, ^{134m}Cs, ^{134}Cs, and ^{137}Cs†

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(Received 31 July 1968)

The atomic-beam method has been applied to the measurement of isotope shifts in the D_1 line of five radioactive isotopes including the isomeric pair ^{134}Cs-^{134m}Cs. The shifts in these lines, relative to the energy in the D_1 line of ^{133}Cs are as follows: ^{127}Cs, +5.9(1.5) mK; ^{129}Cs, +2.8(1.5) mK; ^{134}Cs, +1.8(1.0) mK; ^{134m}Cs, -2.2(1.2) mK; and ^{137}Cs, -6.0(1.5) mK. Here a positive sign means that the frequency of the D_1 line for the indicated isotope is greater than that for ^{133}Cs. Evidence is presented which indicates that the observed shifts arise almost completely from nuclear effects. These shifts must therefore be compared with a calculated shift arising from the normal volume effect of 10 mK/neutron.

I. INTRODUCTION

Historically, the measurement of isotope shifts has always been done by methods in which light is detected. Conventional optical spectroscopy, optical scanning methods,[1] and laser methods,[2] all have this feature in common. Associated with the light-detecting experiments are certain inherent problems which have long inhibited the study of radioactive isotopes. In particular, carrier-free samples of the radioactive isotopes under study must be employed, for otherwise the weak light from the small number of radioactive isotopes under study will be swamped by the intense light of the carrier atoms. In the case of neutron-produced isotopes, this implies that a very expensive and time-consuming mass separation must be performed. In the case of cyclotron-produced isotopes a chemical separation must be performed. Both of these put stringent limits on the half-lives of the isotopes that can be studied. Additionally, there is the problem of wall interaction of the isotopes with the walls of the container in which they are placed. Although there has been some progress in extending the range of radio isotopes studied,[3] it is clear that the fundamental problems still remain, particularly as regards isotopes with half-lives of a few hours or less.

Recently a method employing the atomic-beam technique has been reported for investigating isotope shifts.[4] With the atomic-beam technique as applied to radioactive isotopes, detection is directly on the radioactivity of the isotopes under study, and the basic problems besetting the light-detecting experiments are circumvented. As a result, the atomic-beam technique has enjoyed a great deal of success in the study of hyperfine structures of radioactive isotopes to the extent that spins and moments of several hundred radioactive isotopes have, by now, been successfully measured.[5] Hence the atomic-beam technique would seem to offer fair hope of substantially extending the range of isotope-shift measurements in radioactive isotopes.

In this paper we report the first measurement of isotope shifts on a number of radioactive isotopes of the same element, cesium. From many points of view, cesium is an almost ideal element for a first study of this kind. From a purely experimental point of view, the experimental methods for producing and detecting a large number of cesium isotopes are already known from earlier atomic-beam hyperfine-structure studies.[6] In addition, cesium lamps producing intense D-line radiation are easy to make, and finally the Stark shifts of the D_1 line are sufficiently large[7] that the line separations between the different isotopes are easily spanned. Also, from the point of view of nuclear physics, the cesium isotopes offer interesting possibilities. The neutron-deficient cesium isotopes around ^{127}Cs are believed to be strongly deformed, while ^{137}Cs has a magic number of 82 neutrons and is well described by the shell model. Hence a study of the isotope shifts over a large change of neutron number should determine the systematics of the change in the mean value of $\langle r^2 \rangle$ in going from highly deformed nuclei to spherical nuclei.

II. APPARATUS

The basic apparatus employed is a conventional atomic-beam machine with flop-in magnet geometry.[8] The only new features are (1) a pair of electric field plates capable of sustaining fields up to nearly 10^6 V/cm and (2) hardware associated with the cesium-resonance radiation incident on the C region. We describe each of these separately.

(1) *Electric-field plates.* The most important requirement on our electric field plates is that they can sustain an electric field sufficiently intense to Stark shift the D line through an amount greater than the hyperfine structure of the cesium ground state. This implies fields of the order of several hundred thousand V/cm. In addition we ask for the best homogeneity possible over a region equal in height to the beam height and as long as the light irradiation length (about 0.040 in. by 2 in.). We have been able to achieve about 0.5% at the highest fields.

A schematic of the basic design is shown in Fig. 1. With ordinary metal electrodes, the highest fields we could produce were about 3×10^5 V/cm. This limiting field arises from high-field emission of electrons from the cathode surface. In order to improve on this, we use a technique employed in the electromagnetic beam separators

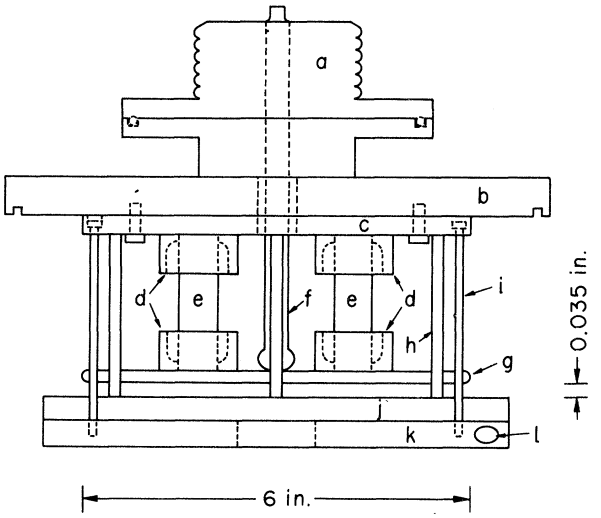

FIG. 1. Schematic diagram of the electric field plate construction.

a, ceramic high-voltage insulator;
b, vacuum flange;
c, backing plate;
d, discharge prevention cups;
e, alumina insulator;
f, high-voltage feed-through rod;
g, anode;
h, spacers;
i, support rods;
j, glass cathode;
k, aluminum backing plate for cathode;
l, hole for heating element.

used at the Bevatron[9]; that is, to substitute a cathode made from soft glass which is heated to about 60°C. With an anode made from hard stainless steel and a gap width of 0.035 in., fields of almost 10^6 V/cm can be maintained. Insulation of the anode from ground is accomplished by 2-in.-long alumina spacers. These spacers are, in turn, surrounded by stainless-steel cups to prevent charge layers from building up on the alumina. To achieve the best homogeneity possible, the surfaces of the glass cathode, the anode, the alumina spacers, and the ground plate on which the alumina spacers rest are all ground as parallel and flat as possible. The proper gap width is now achieved by six stainless-steel spacers which are held between the ground plate and the glass cathode. These spacers are also ground flat and parallel and to the same length. The high voltage is introduced onto the anode by means of a Ceramaseal 50-kV high-voltage feed-through.

An interesting feature of the electric field plates is that because of the narrow gap and the length (about 6 in.), they act as a state selector in the atomic-beam apparatus. With reference to Fig. 2, it can be seen that by rotating the plates with respect to the beam axis, either the trajectory with $m_J = +\frac{1}{2}$ in the A magnet or the trajectory with $m_J = -\frac{1}{2}$ in the A magnet can be blocked out. With reference to a Breit-Rabi diagram[10] for $J = \frac{1}{2}$, it is observed that essentially all atoms with $m_J = +\frac{1}{2}$ in the A magnet arise from the upper hyperfine level ($F = I + \frac{1}{2}$), whereas all atoms with $m_J = -\frac{1}{2}$ in the A magnet arise from the lower hyperfine level ($F = I - \frac{1}{2}$). This feature is very

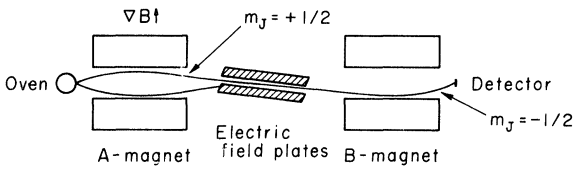

FIG. 2. Schematic diagram (exaggerated) showing how the electric field plates can select either of the two trajectories.

useful and has been employed extensively in our experiment.

(2) *High-voltage system.* The high-voltage supply for these experiments is a Sames 50-kV electrostatic generator, type R. This supply is highly regulated and provides a very stable and easily reproducible voltage. Voltage measurement was achieved with a model Park high-voltage divider. The output of this is fed into a Vidar 240 voltage-to-frequency converter, and the frequency is read on a Hewlett-Packard 524C frequency counter. In this way a fast, accurate, and stable voltage readout is obtained.

(3) *Optics.* The cesium lamp employed is a Varian model X49-609 spectral lamp. The D-line filters were obtained from Spectrolab, Inc. (12484 Gladstone Ave., Sylmar, California 91342).

III. METHOD AND RESULTS

The basic method employed results from the idea of a tuning experiment; this idea has already been used successfully in the study of the isotope shifts in the mercury isotopes.[1] In those experiments a magnetic field was used to tune the emission lines of lamp atoms with the absorption lines of the isotopes under study. However, in atoms, such as cesium, which exhibit considerable hyperfine structure (hfs), a magnetic field is not a suitable tuning mechanism. The reason is that the magnetic field will lift the Zeeman degeneracy, which gives rise to a complicated splitting pattern. In the case of ^{133}Cs, for example, with a nuclear spin of $I = \frac{7}{2}$, there are 16 Zeeman sublevels in the ground state alone. Moreover most of them are connected by optical matrix elements to six excited states, and the resultant tuning signals are too complicated to be interpretable.

The D_1 transition $6^2p_{1/2} \rightarrow 6^2s_{1/2}$ of cesium has, however, a property that makes the employment of electric fields an almost ideal tuning mechanism; namely, that both states involved in the transition have electronic angular momentum $J = \frac{1}{2}$. To understand the special nature of states with $J = \frac{1}{2}$, consider the second-order Stark matrix element determining the energy shift:

$$\Delta W(n^2l_J IFm_F) = \sum_{\Psi} \frac{|\langle n^2l_J IFm_F | -e\vec{r}\cdot\vec{E}|\Psi\rangle|^2}{\Delta E(\Psi; n^2l_J IFm_F)}, \quad (1)$$

where ΔW is the induced energy shift in the state with the indicated quantum numbers, $\vec{r}$ is the di-

pole operator, and $\vec{E}$ is the applied electric field. It can be shown[7] that for the case where $J = \frac{1}{2}$, the matrix element squared,

$$|\langle n^2 l_J I F m_F | - e\vec{r}\cdot\vec{E}|\Psi\rangle|^2 ,$$

is independent of all the angular momentum quantum numbers for the state. This means that the only mechanism by which states of different F can have a differential shift results from the fact that in the denominator of (1) different hyperfine levels, F belonging to the same multiplet (n, I, J, l), have slightly different energies. However, this is clearly a very small effect, being of order (hfs/optical energy separation) $\approx 10^{-5}$. If the possibility is now considered of inducing a differential shift in states of the same F, but different m_F (lifting of Zeeman degeneracy), it is seen that the expression (1) gives no mechanism for doing so. Since lifting of Zeeman degeneracy must occur in a higher order of perturbation theory, it is reasonable to expect that such an effect is even smaller than the differential shift of the hyperfine levels. Thus the largest shift is the gross Stark shift which affects all of the hyperfine states equally. Experimental values of the gross Stark shift,[7] differential hyperfine shift,[11] and lifting of the Zeeman degeneracy[12] all exist in the literature, and in Table I we indicate the magnitude of these effects at an electric field of 0.5×10^6 V/cm, the largest field employed so far in these experiments. In order to understand the significance of these effects for the Stark-tuning experiments reported here, the magnitude of the latter two effects need to be compared with the linewidth expected in the experiment. The smallest possible linewidth is the natural linewidth of the D_1 transition, about 10 MHz. This is substantially larger than either of these effects. Hence we conclude that for purposes of a tuning experiment the only significant effect of an electric field is to shift all hyperfine states belonging to an atomic level by an equal amount, all other effects being negligible.

Unlike the Zeeman-tuning experiments, where the frequency shift induced for a given applied magnetic field is accurately calculable from the well-understood Zeeman effect, the Stark effect is not well known. Hence any experiment employing an electric field as a tuning mechanism must have a built-in mechanism for determining the Stark effect; i.e., for obtaining a calibration of frequency shift versus applied voltage.[2] The method for doing this has been reported in Ref. 4, so we give here only a brief review.

The basic apparatus is shown in Fig. 3. Light from a cesium discharge lamp is passed through

TABLE I. Experimental Stark effects in cesium at a field of 0.5×10^6 V/cm.

Effect	Magnitude at 0.5×10^6 V/cm
Gross Stark shift	2.56×10^4 MHz
Differential hyperfine shift	0.56 MHz
Lifting of Zeeman degeneracy	0.003 MHz

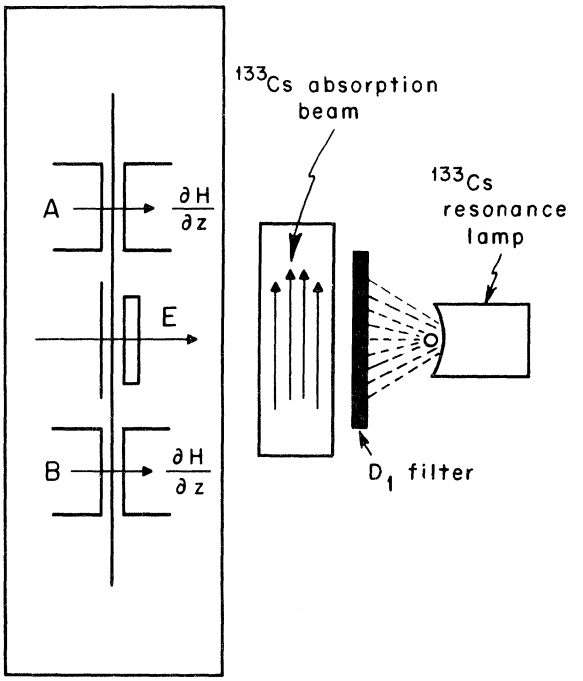

FIG. 3. Schematic diagram of the apparatus.

a filter which transmits only the D_1 line. This line consists, in the lamp, of two resolved components which are separated by the ground-state hfs. The width of each of these components is about 1500 MHz, the principal sources of this width being the hfs of the excited $6^2p_{1/2}$ state and the Doppler width. In order to improve the precision of the experiment, the light is now passed through a cell containing an optically dense cesium absorption beam. Such a beam will absorb the light in two lines from each of the components emitted by the lamp. (See the energy-level diagram of Fig. 2.) The separation of the two lines is equal to the hfs of the excited $6^2p_{1/2}$ state. The width of the lines is characteristic of the beam collimation and is about 150 MHz. Within the lines we have been able to achieve almost complete absorption (>50%) for several hours. The light so filtered and structured is now allowed to fall incident between the plates of an electric field apparatus located in the C region of the atomic-beam apparatus.

Consider now the behavior of a ^{133}Cs beam in the C region of the atomic-beam apparatus under the irradiation of this light. At zero electric field the absorption lines (see Fig. 4) of the atoms in the atomic-beam apparatus coincide with the absorption lines of cesium atoms in the absorption cell, and a minimum in the intensity curve is observed. However, as the electric field is turned on, the Stark effect decreases the frequency of the absorption lines of atoms in the beam apparatus, and the observed signal increases until the electric field is sufficient to shift the frequency by an amount equal to the hfs of the excited $6^2p_{1/2}$ state when a second intensity minimum is

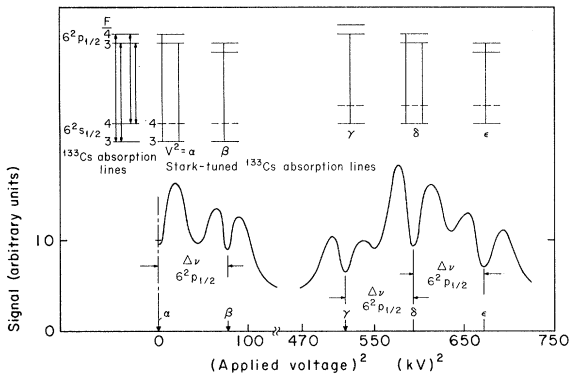

FIG. 4. Observed ^{133}Cs signal versus square of applied voltage. The position of the Stark-shifted absorption lines relative to those in the absorption cell for each of the observed minima is indicated directly above the minima. The separation between α and δ corresponds to a shift equal to the ground-state hyperfine separation and serves as a calibration.

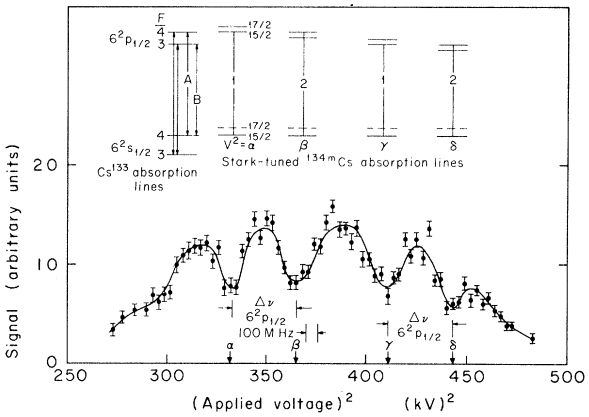

FIG. 5. Observed ^{134m}Cs signal versus square of applied voltage. The minimum α occurs when beam absorption line 1 coincides with ^{133}Cs absorption line A; β occurs when line 2 coincides with A; γ occurs when 1 coincides with B; and δ occurs when 2 coincides with B.

observed. At higher electric fields the frequency is shifted by an amount equal to the ground-state hfs, and the beam absorption line is brought into resonance with the second lamp emission line. Here, three intensity minima can be observed corresponding to the overlap positions indicated in the energy-level diagram. These three minima are equally spaced and correspond to a shift by an amount equal to the hfs of $6^2p_{1/2}$. The unlabeled minima correspond to structure present in the lamp line. As can be seen from the energy-level diagram, the separation between the two minimum points labeled α and δ corresponds to a Stark shift of the energy levels equal to 9192 MHz, the hfs of the ^{133}Cs ground state. It is this that we use as a calibration.

This same method, applied to atomic beams of other isotopes, can be used to determine the isotope shifts. Consider now the case of a ^{134m}Cs beam. (Energy-level diagram shown in Fig. 3.) Here the hfs of both $6^2s_{1/2}$ and $6^2p_{1/2}$ is smaller than for ^{133}Cs, so that Stark tuning can bring about only one overlap of the beam-absorption lines with the lamp emission lines. However, there are four possible overlap positions of the Stark-shifted energy levels of ^{134m}Cs beam atoms with the unshifted levels of ^{133}Cs atoms in the absorption cell. As seen in Fig. 5, these overlap positions correspond to minima in the observed intensity pattern. From the energy-level diagram, it is clear that the separations between the minima α and β and between γ and δ correspond to the hfs of the $6^2p_{1/2}$ state of ^{134m}Cs. These are experimentally seen to be equal. Moreover, the separations between α and γ and between β and δ should correspond to the hfs of the $6^2p_{1/2}$ state of ^{133}Cs. This also agrees with our observations. From the absolute positions of the four minima and a knowledge of the hfs of the ground and excited states, the isotope shift can be deduced. We find

$$\text{isotope shift} = -1.4(1.5)\times 10^{-3}\ \text{cm}^{-1},$$

where the negative sign indicates that the energy of the D_1 line in ^{134m}Cs is smaller than that in ^{133}Cs.

Similar data have been obtained for all the other isotopes reported here. Results are given in Table II.

IV. DISCUSSION

In Table I are given the results to date on all cesium isotope-shift measurements for the D_1 line. In the case of ^{131}Cs and ^{132}Cs the measurements are actually made for the D_2 line, but we infer the D_1 isotope shift from these results and the ratio of D_2 to D_1 isotope shift given by Hühnermann and Wagner[13] for ^{134}Cs. These measurements form a rather complete picture for the isotopes in the range ^{127}Cs to ^{137}Cs.

There are several striking features to the data. First we note that there seems to exist a small but finite isomer shift between ^{134m}Cs and ^{134}Cs, the size of the effect being larger than two standard deviations if one employs Hühnermann and Wagner's value for ^{134}Cs. This is very strong evidence that the observed shifts are due primarily to nuclear effects since mass effects would not give rise to an isomer shift. Also bearing on this point are recent calculations by a nonrelativistic Hartree-Fock approximation of Bauche[14] on the specific mass effect in cesium. Bauche's result is a shift of -0.77 mK between ^{132}Cs and ^{134}Cs, where the minus sign indicates the shift is in the direction opposite to the normal (reduced) mass shift. The normal mass shift can be calculated exactly and is equal to $+0.69$ mK between the same two isotopes, and the two mass effects substantially cancel each other out. Although there is certainly an error to be associated with Bauche's calculation, we believe that the main conclusion

TABLE II. Measured isotope shifts in the cesium isotopes (10^{-3} cm^{-1}).

Isotope	127	129	131	132	134	134m	135	137
This work	5.9(1.5)	2.8(1.5)			1.8(1.0)	−2.2(1.2)		−6.0(1.5)
Otten and Ullrich[20]			−0.31(5)	1.6(5)				
Hühnermann and Wagner[13]			0.39(9)		1.17(5)		−1.23(7)	−4.81(6)

that mass effects are small relative to the measured shifts is well supported by the ^{134}Cs–^{134m}Cs isomer shift.

If it is accepted that the observed shifts are indeed nuclear effects, then there are several qualitative comments that can be made. First we note that the observed shifts are only a small fraction of the shifts predicted by the normal volume effect. The prediction of the normal nuclear effect can be calculated from the usual expressions[15] using for $|\Psi^2(0)|$ the value determined from the ^{133}Cs ground-state hfs and the NMR value[16] for the nuclear moment. It is found that $\delta(\Delta E) = 10$ mK/neutron. The observed shifts are seen to be almost an order of magnitude smaller over the entire range of measurement. A similar situation has been found to exist in the measured isotope shifts in the xenon[17] isotopes ($z = 54$) and in the barium[18] isotopes ($z = 56$).

There would appear to be at least two possible explanations for the smallness of these shifts. One possibility is that in the approach to a closed shell of neutrons, the size of the normal volume effect should be somewhat diminished. Another possible explanation is suggested by the fact that the cesium isotopes on the neutron-deficient side of ^{133}Cs seem to have increased deformations. Recent measurements[19] of the quadrupole moments of ^{131}Cs and ^{132}Cs confirm this point. Hence the deformation effect may offset the volume effect on the neutron-deficient side of ^{133}Cs.

†Work supported by the U. S. Atomic Energy Commission.

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