OPTICAL DETECTION OF THE CESIUM HYPERFINE TRANSITION*

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The argon spectral line at 8521.4A has been used in the detection of the cesium hyperfine transition at 9192.63 mc. The argon line was Zeeman split to overlap one hyperfine component of the cesium resonance line at 8521A. A glass bulb containing cesium vapor diffusing in a buffer gas was elluminated by the argon light source and the scattered light was observed. Transitions from the upper ground state hyperfine sublevel to the excited state were produced, and the atoms then returned by spontaneous emission to both ground state sublevels. There was thus a pumping of atoms out of the upper hyperfine level and into the lower one. This depletion of the population of the upper level caused a decrease in the amount of scattered light. Microwaves were then applied to the bulb by a horn antenna. At resonance the difference in population of the hyperfine levels was reduced and an increase in the scattered light was seen. Shifts in the resonance frequency of the $(4, 0) \longrightarrow (3, 0)$ line with buffer gas pressure were observed for He, N_2, Ne, Ar and Kr, the smallest being -200 cycles/mm for argon and +580 cycles/mm for Ne[1]. An upper limit of 10 cycles/cm of argon was found for the expected pressure-dependant line breadth caused by cesium-argon collisions. The microwave frequencies were obtained from an Atomichron by feeding in an accurately known variable frequency at the 6.315920 mc stage in the synthesizer. The basic 5 mc frequency for the multiplication chain was supplied by the 5 mc output from another Atomichron which was operating normally.

* This work was performed at the U. S. Naval Research Laboratory, using the unique precision frequency facilities of the Radio Techniques Branch.

1 Similar results have been obtained by M. Arditi and T. R. Carver.

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The use of optical methods to orient atoms was suggested in 1950 by A. Kastler[1] and later, independently, by R. H. Dicke. Since then the term "optical pumping" has come to be applied to any process in which optical resonance radiation of selected polarization, direction, and/or wavelength is used to produce differences in population between the magnetic or hyperfine sublevels of a given atomic level. Changes in the population of the sublevels can be detected optically by observing the resulting changes in polarization or intensity of the scattered or transmitted light.

The observation of narrow hyperfine absorption lines in alkali metal vapors was made possible by a method proposed by Dicke[2] for reducing the doppler width. It was pointed out that if the atoms were restricted by collisions with a buffer gas or with the walls of the apparatus so that the average displacement during the time necessary for the transition was much less than for free atoms, the doppler width should be much smaller also. The collisions must not, of course, change the internal state of the atom appreciably, and it is the finite amount of disorientation actually taking place which limits the narrowness of the lines that can be obtained.

About a year ago hyperfine absorption lines as narrow as 70 cycles at 6834 megacycles were reported by Carver[3] for rubidium-87 in an argon buffer gas. The resonance was detected by microwave power absorption and optical

pumping was used only to increase the population difference between the levels and thus the signal-to-noise ratio. Optical detection of the field-independent component of a hyperfine transition in a vapor was first achieved by Arditi and Carver[4], and soon thereafter by Bell and Bloom[5]. Sodium was used and the original linewidths were 100 cycles or so at 1771 mc. More recently, the field-independent transition in cesium at 9192 mc. has been observed optically by Arditi[6] and by us[7] with somewhat different pumping methods. All of the above optical detection experiments fall under the heading of what we might call "hyperfine pumping".

Fig. 1 gives the energy level diagram for cesium. The excited state hyperfine splittings are much smaller than that for the ground state and will be ignored. By hyperfine pumping we mean any scheme which uses differences in intensity of the hyperfine components of the optical lines to pump atoms out of one ground state hyperfine sublevel and into the other. Atoms from one hyperfine sublevel are raised to the excited state more frequently than those from the other, and if the transition probabilities back to the two ground state sublevels are nearly equal, one gets a resulting difference in population of the ground state sublevels. The amount of light scattered or transmitted can be used to monitor the difference in level populations, since the absorption coefficient will be different for atoms in the two sublevels if the hyperfine optical line intensities are different. If microwaves of the right frequency to produce the hyperfine transitions are then introduced, one can observe a change in the scattered or transmitted light intensity.

The experiments of Arditi and Carver and of Bell and Bloom on sodium and of Arditi on cesium made use of the difference in absorption coefficient of the gas for the two optical hyperfine components to produce a difference in intensity in the rear part of the sample. The transmitted light was monitored. We have also used this method with the modifications of using another absorption cell before the sample to do the filtering and of looking at the scattered light. However, most of our measurements have been made with an argon light source instead of the usual alkali metal discharge light source.

The wavelengths of the relevant optical lines are given at the bottom of Fig. 1. The hyperfine components of the cesium D_2 line are at 8521.0 and 8521.2 A. Nearby is the argon line at 8521.4 A. It can be split in a field of about 5000 gauss so that one of the components overlaps the cesium hyperfine component at 8521.2 A. This provides a light source which has much more intensity at the wavelength of one cesium hyperfine component than at the other and thus makes efficient hyperfine pumping possible.

The experimental arrangement is shown in Fig. 2. Light from an argon discharge is passed through a bulb containing cesium vapor in a buffer gas and the scattered light is monitored by a photomultiplier. The output current is observed as the microwave frequency fed in by the horn is varied through resonance. The circuits of an Atomichron were used to provide the necessary frequency. The internally synthesized 6.315920 mc. signal in the Atomichron was replaced by the output of an extremely precise variable frequency synthesizer. The basic 5 mc. frequency was obtained from another Atomichron which was operating normally. In this way the frequency could

be swept through resonance in very fine steps and the line shape obtained. The Atomichrons, synthesizer, and other frequency measuring equipment were provided by the NRL Radio Techniques Branch.

In Fig. 3 one of the earlier lines obtained is shown. 30 mm. of helium was used as the buffer gas and some power broadening occured. The signal-to-noise ratio for this line was not good, although part of the apparent irregularity is due to the fact that the synthesizer frequency was swept in discrete steps, and during switching at the end of a dial the frequency was momentarily considerable off. More recently we have achieved linewidths of about 40 cycles, or Q's of over 200,000,000, with voltage signal-to-noise ratios of about 400. Better optical filtering of the light source should provide a further reduction in noise.

In the near future we are also planning to try another form of hyperfine pumping which should be very efficient for observing the rubidium-87 hyperfine transition. This method, which has been suggested independently by Carver and Alley in a somewhat different form, consists of using a rubidium-85 filter in front of the rubidium-87 cell to get the desired difference in intensity of the hyperfine components. The isotope shift[8] is sufficient so that one of the rubidium-87 lines is much closer to the corresponding rubidium-85 line than the other one, and thus a large difference in intensity of the transmitted hyperfine components can be produced with little loss of light intensity. It is hoped that Maser action with rubidium-87 will be possible with this method[9].

With the system as described we have made measurements of the frequency and width of the observed line as a function of the buffer gas chosen, of the pressure of the buffer gas, and of the temperature of the bulb. The

frequency shifts have been measured also by Arditi and will be described by him in more detail. Our results were +1050, +890, +580, -190, and -1300* cycles per mm. of buffer gas for helium, nitrogen, neon, argon, and krypton. The main limitations in accuracy for helium, neon, and argon came from uncertainties in the pressures of the bulbs. 500 milliliter bulbs were generally used and no allowance has been made for absorption by the walls. A 75 per cent argon, 25 per cent neon mixture gave very small shifts at room temperature, but the residual shift was found to be quite temperature dependent, the coefficient being about 5 cycles per degree per cm. of buffer gas mixture. This indicated a different fractional change in shift with temperature for the two gases. A check of the temperature dependence for helium, neon, and argon gave roughly +15 cyles/cm/deg, +1 cycle/cm/deg, and -7 cyles/cm/deg. The smallness of the temperature coefficient for neon is quite striking.

Line width measurements gave considerably narrower lines for neon and helium than for argon. With neon the width at half power was 40 cycles from 1 cm to 10 cm, and about twice this width at 3mm and 50 cm with much weaker signals. For helium 40 cycle linewidths were found at 1 and 3 cm. For neon the calculated reduced doppler width at 3 mm was about 50 cycles, while the extra width at 50 cm is consistant with a contribution from buffer gas collisions of one to two cycles/cm. The line width for argon, however, appears to be about 120 cycles at 1 cm and 4 cm, and then to start going up at about 15 cm. Light intensity, magnetic field inhomogeniety, and cesium-cesium collisions do not appear to be responsible for the residual linewidth, but short term fluctuations in the frequency could contribute. The lines observed were Lorentz shaped.

The value for krypton quoted in Reference 7 is incorrect.

We have also made measurements of the Zeeman transitions in the earth's magnetic field. These are the field dependent $\Delta F = 0$, $\Delta M = 1$ transitions shown in Fig. 4. To get away from stray field disturbances we took the apparatus to the Fredericksburg Magnetic Observatory in Fredericksburg, Virginia. The work there was done in collaboration with Mr. Thomas L. Skillman of the Coast and Geodetic Survey and included an absolute measurement of the earth's magnetic field[10]. The method introduced by Dehmelt[11,12] of transmission monitoring with circularly polarized light was used. Linewidths at half power of less than 20 cycles were observed for rubidium and cesium at pressures of a few cm. of argon or neon. For rubidium-87 it was possible to resolve the second order splittings which are about 46 cycles in the earth's field, although the $2 \rightarrow 1$ transition was much stronger than the others. Measurements of linewidths at higher buffer gas pressures gave results consistent with those obtained for the hyperfine transition. In particular, for cesium we found a collisional contribution to the linewidth of about 4 cycles/cm at half voltage for argon and a factor 3 or so less for neon. Measurements of the signal decay time after reversing the magnetic field, as described by Dehmelt[11], were also carried out for cesium in argon, and the measured time was found to be about a factor 5 longer than would correspond to the observed linewidths. This indicates that the most important transitions in the relaxation process are ones for which the change in M is small. The reason for the residual linewidth at low buffer gas pressures for the Zeeman transitions is not known.

From our results so far it has become clear that one can obtain a Q of 200,000,000 for the hyperfine transition in cesium and still have an excellent signal-to-noise ratio and no temperature shift. Wall absorption effects could cause undesired drifts, but none have been seen yet. It is hoped that methods such as the use of buffered walls, which has been discussed by Robinson, Ensberg and Dehmelt[13,14], in the future will lead to still narrower lines that are obtained with a buffer gas. However, it is of interest that we are in a position right now to get precisions better than 1 part in 10^{10} over extended periods with simple apparatus. This should make possible intriguing experiments such as using a Dicke type clock in a satellite to check on the expected shift in rate of a clock in a region of different gravitational potential.

We'd like to express our thanks to the Naval Research Laboratory for permission to use the excellent facilities of the Radio Techniques Branch, and to Mr. H. F. Hastings and the members of his group for their extensive aid with the experiments. We would also like to thank the U. S. Coast and Geodetic Survey for permission to conduct some of the measurements at the Fredericksburg Magnetic Observatory, and Mr. Thomas Kelsall for assisting with the experiments. This work was directly stimulated by the experiments of H. G. Dehmelt and of T. R. Carver, and we have benefitted greatly from discussions with them.

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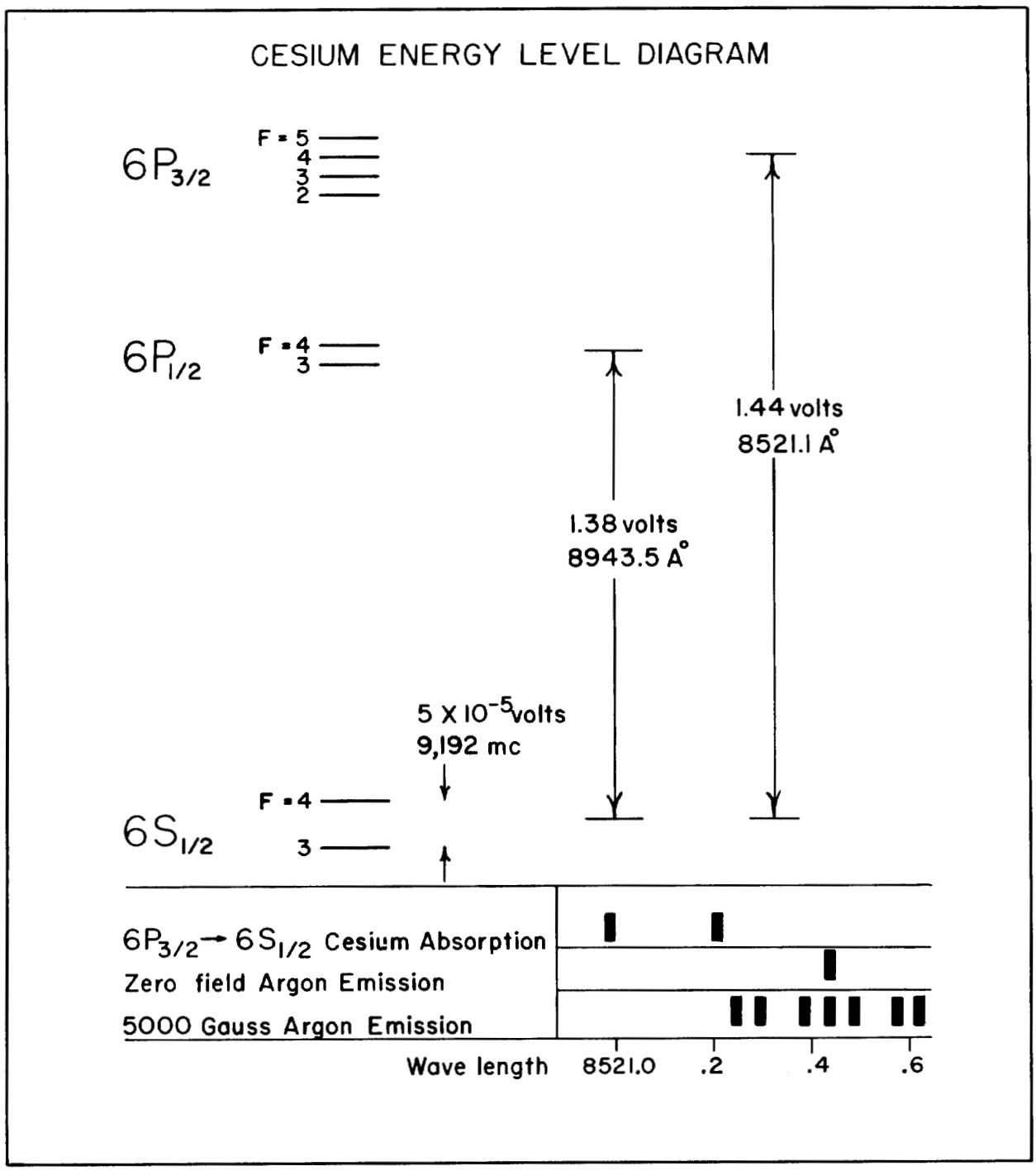
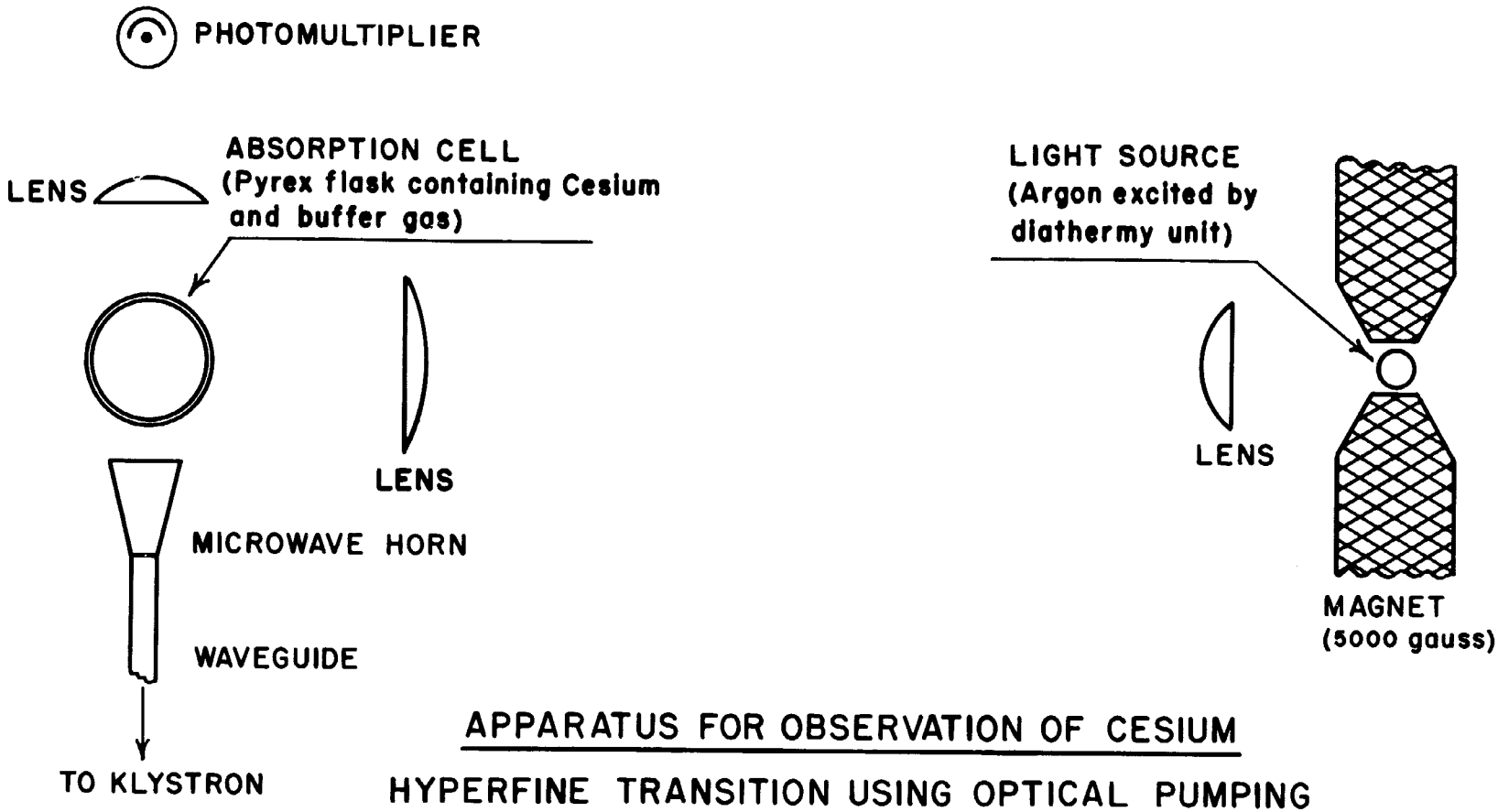

CESIUM ENERGY LEVEL DIAGRAM
6P3/2
F = 5
4
3
2
6P1/2
F = 4
3
1.44 volts
8521.1 A°
1.38 volts
8943.5 A°
5 X 10-5 volts
9,192 mc
F = 4
6S1/2
3
6P3/2 → 6S1/2 Cesium Absorption
Zero field Argon Emission
5000 Gauss Argon Emission
Wave length
8521.0
.2
.4
.6

Figure 1



APPARATUS FOR OBSERVATION OF CESIUM HYPERFINE TRANSITION USING OPTICAL PUMPING

Figure 2

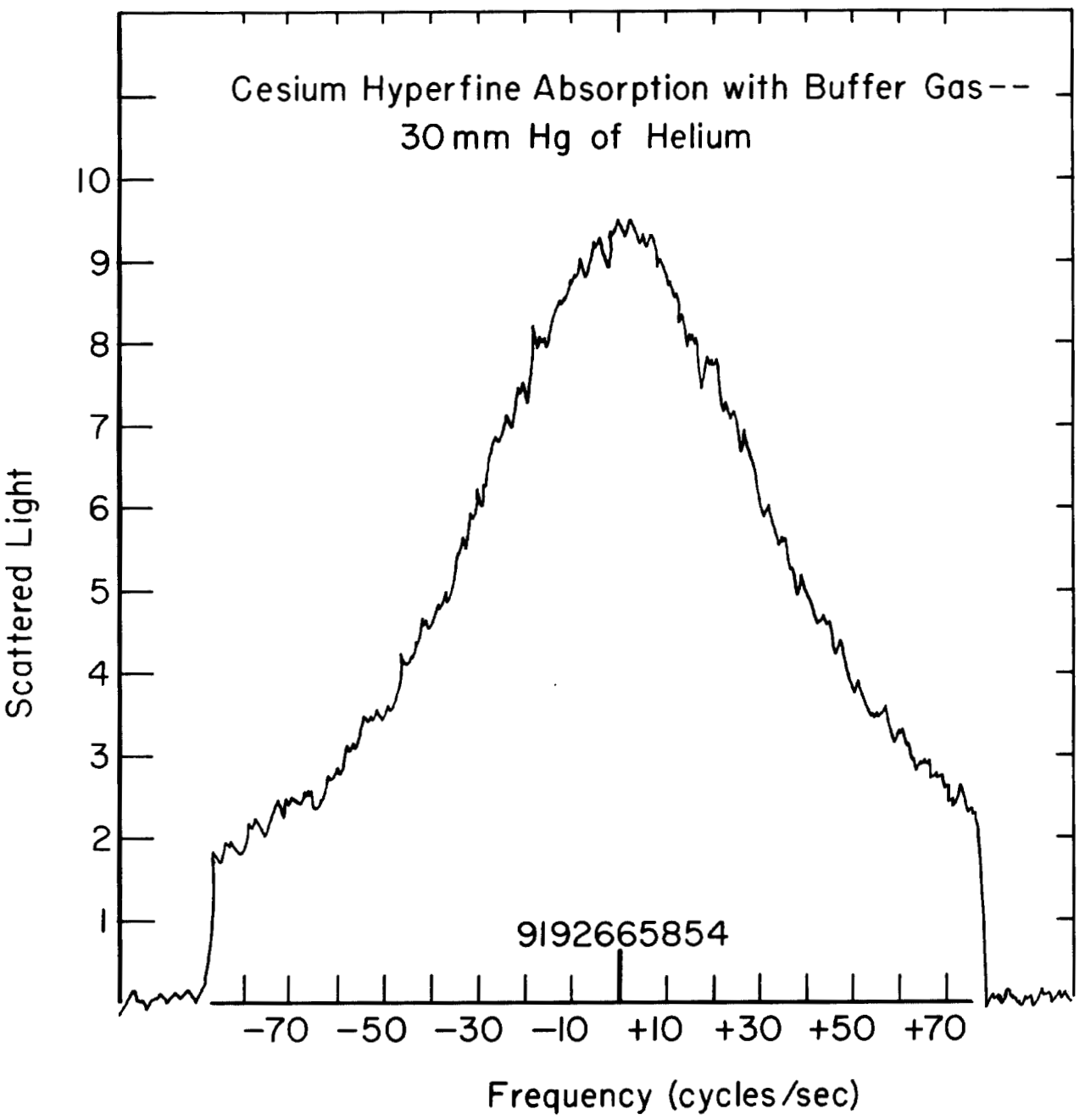

Cesium Hyperfine Absorption with Buffer Gas--
30 mm Hg of Helium
Scattered Light
10
9
8
7
6
5
4
3
2
1
9192665854
-70 -50 -30 -10 +10 +30 +50 +70
Frequency (cycles/sec)

Figure 3

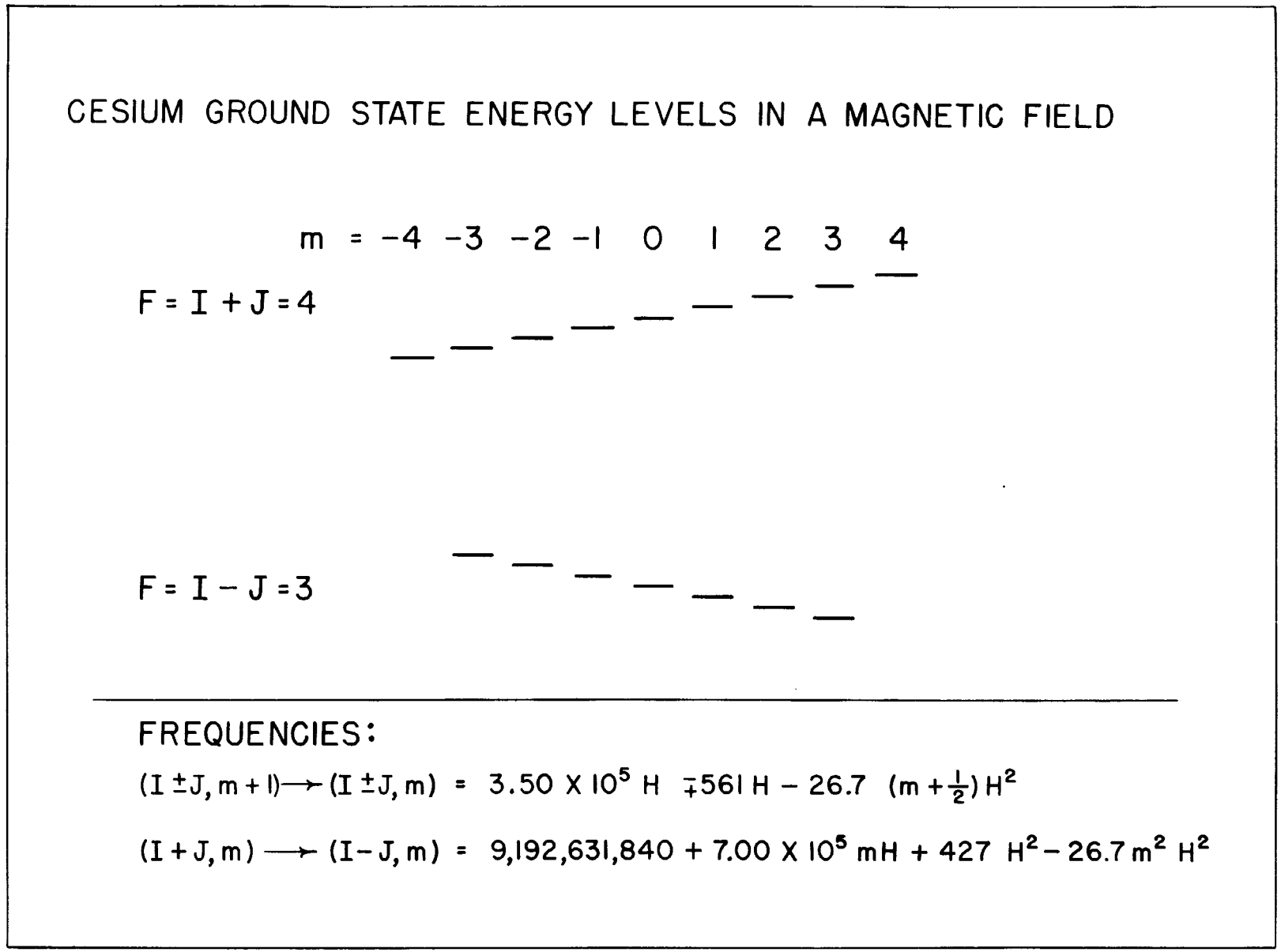

CESIUM GROUND STATE ENERGY LEVELS IN A MAGNETIC FIELD
m = −4 −3 −2 −1 0 1 2 3 4
F = I + J = 4
F = I − J = 3
FREQUENCIES:
(I ±J, m + 1) → (I ±J, m) = 3.50 X 10^5 H ∓561 H − 26.7 (m + ½) H^2
(I + J, m) → (I − J, m) = 9,192,631,840 + 7.00 X 10^5 mH + 427 H^2 − 26.7 m^2 H^2

Figure 4