Energy Shifts of the Magnetic Sublevels of $^3S_1$ Helium Caused by Optical Pumping

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It has been observed experimentally that the energy spacing between the magnetic sublevels of the $2\,^3S_1$ state of helium as detected by optical pumping varies with the intensity and character of the resonant radiation. The relative shift as a function of the intensity, polarization, and spectral distribution are observed and measured. The energy shifts are on the order of 50–60 cps and become smaller as the intensity of the light decreases. A second lamp containing He^3 incident on the absorption cell in the same manner as the pumping lamp also produces an energy shift. An explanation of the light shift in metastable helium is discussed.

INTRODUCTION

THE optical pumping process in the alkalis,[1,2] Hg,[3] and He[4,5] has permitted the observation and measurement of certain quantities in a unique environment. The hyperfine separation of the ground states of the alkalis has been measured to a high order of accuracy. Similarly, the magnetic splitting of the ground state of the alkalis and mercury, and more recently, metastable helium can be observed and the g factor measured. However, the very processes which permit these measurements to be made can also, in certain circumstances, influence the results in a very basic way.

Cohen-Tannoudji[6] describes an experiment utilizing the pumping process in Hg^{199} which demonstrates the effect of the pumping light in producing a displacement of the magnetic resonance. He shows experimentally a displacement of the magnetic resonance line which is shifted toward the low-frequency values when the energy difference between the emitting and absorbing atom is positive and the pumping light σ^+ polarized. The shift is proportional to the intensity of the resonant radiation. For this case the energy difference between centers of the emission and the absorption line was about one Doppler width and positive. His measured shifts were on the order of a cycle per second. Arditi and Carver[7] investigated the light intensity frequency shifts of the hyperfine separation of Cs^{133} and Rb^{87} using optical pumping. The measured shifts in this case were about 200 cps. Again the source of the shift is the presence of the pumping radiation and is proportional to its intensity.

The optical pumping process has been used to study the magnetic separation of the $2\,^3S_1$ state of helium. It is observed experimentally that under certain conditions the sublevels acquire an additional energy because of the presence of the resonant pumping radiation. The results of the measurements under varying conditions of the pumping light are presented and discussed.

The details of the optical pumping process in helium have been discussed elsewhere.[4,5] Those aspects of the optical polarization of helium atoms which are important to the understanding of the energy shifts of the magnetic sublevels in metastable helium are repeated here briefly. The pumping light from a helium lamp consists of two resolved components near 1μ (see Fig. 1). It can be shown[5] that the D_0 radiation plays only a small part in the pumping process, when the D_3 radiation is circularly polarized and the absorption cell pressure is sufficiently small to avoid mixing in the P states; for this reason we can safely ignore its presence. The D_3 radiation from the lamp is σ^+ polarized and is passed colinear with the external magnetic field through a helium absorption cell containing atoms in the $2\,^3S_1$ metastable state. In general, the three magnetic sublevels do not absorb radiation at equal rates. This results in unequal populations of the three sublevels. The radio-frequency resonance of the polarized atoms is detected by observing the reorienting effect of an oscillating magnetic field applied at right angles to the external magnetic field and at a frequency corresponding to the energy separation of the sublevels. A measurement of the relative shift in energy introduced between the magnetic levels because of the presence of the nonresonant radiation in the pumping beam is determined

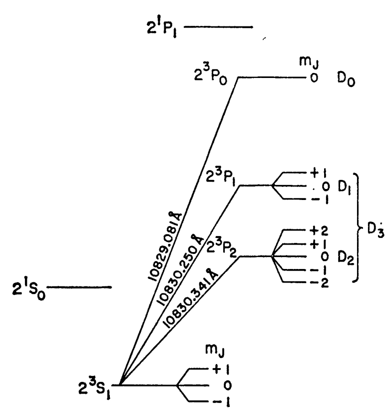

FIG. 1. Relevant energy levels of a helium atom in a weak magnetic field (not to scale).

[1] H. G. Dehmelt, Phys. Rev. **106**, 1487 (1957).
[2] W. E. Bell and A. L. Bloom, Phys. Rev. **107**, 1559 (1957).
[3] A. Kastler, Physica **17**, 191 (1951).
[4] F. D. Colegrove and P. A. Franken, Phys. Rev. **119**, 680 (1960).
[5] L. D. Schearer, *Advances in Quantum Electronics* (Columbia University Press, New York, 1961), p. 239.
[6] C. Cohen-Tannoudji, Compt. rend. **252**, 394 (1961).
[7] M. Arditi and T. R. Carver, Phys. Rev. **124**, 800 (1961).

by precisely measuring the resonance frequency in a constant magnetic field.

EXPERIMENTAL ARRANGEMENT

A block diagram of the apparatus used in this series of experiments is shown in Fig. 2. The modulation coils are used to provide a small amplitude variation of the external magnetic field. The photocell output is then modulated at the same rate when the field moves in and out of resonance. If the modulation displaces the resultant field symmetrically about the resonance line, the amplitude of the fundamental is zero. A phase sensitive detector is employed to detect this condition. The error signal derived from the phase detector is then fed back to a second set of Helmholtz coils whose axis is parallel to the magnetic field. Small coils with their axis perpendicular to the light beam provide the reorienting rf field at a frequency corresponding to the energy separation of the magnetic sublevels. This rf field is derived from a standard signal generator with excellent short-term stability. The result of the feedback to the large Helmholtz coils is to tie the magnetic field at the center of the coils directly to the frequency of the rf field. Additional details concerning the apparatus may be found in the paper by Schearer.[8]

The feedback current in the Helmholtz coils is a function of the external magnetic field and the resonance oscillator. If the magnitude of the external magnetic field and the frequency of the oscillating magnetic field remain constant, any variation in the current in the Helmholtz coils as the character of the pumping light is changed must necessarily be associated with an apparent change in the energy separation of the sublevels. The energy separation between the magnetic sublevels of the $2\,^3S_1$ state in helium is directly proportional to the external magnetic field with the exception of an anomalous part associated with the polarization, spectral distribution, and intensity of the transmitted pumping light. Thus, a change in the Helmholtz current becomes a measure of the change in the separation of the levels as the character of the pumping light is modified. In practice, the frequency change of the oscillator required to maintain the Helmholtz current at zero is the quantity measured. This avoids incorporating any errors associated with the closed loop into the observations.

The relative shifts of the levels are recorded as changes in the resonant frequency. Since $g\mu/h \approx 2.8$ (Mc/sec) G^{-1}, a change in the oscillator frequency of 2.8 cps corresponds to a change of 1 part per million in the separation of the sublevels in a field at 1.0 G. With the present apparatus it is possible to observe changes of 3 cps in the resonance frequency. The instrument itself is capable of discerning changes of less than 1 cps; however, for the purposes of this experiment, the fluctuations in the magnetic environment obscure readings smaller than 3 cps.

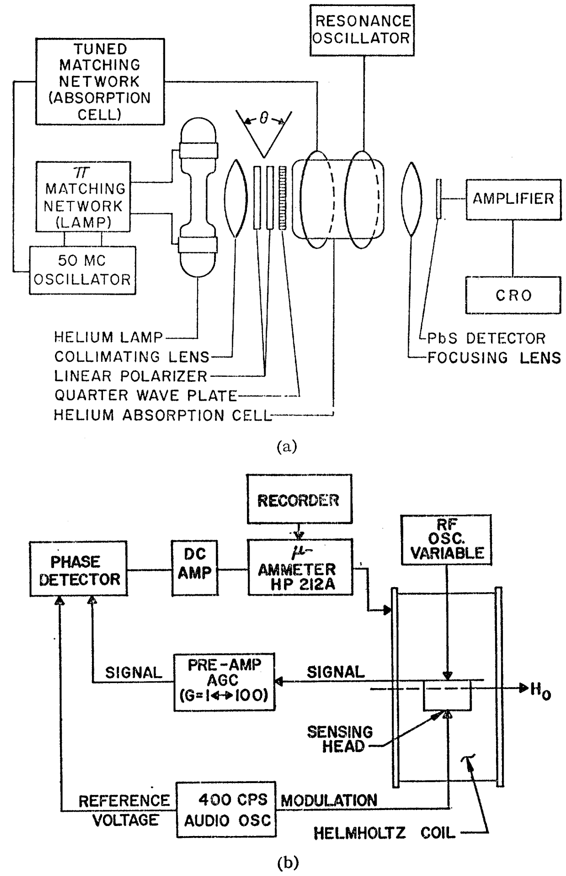

FIG. 2. (a) Schematic diagram of the optical pumping instrument. (b) Block diagram of the apparatus.

Frequency shifts of the resonance have been observed and measured under the following conditions:

(1) A change in the polarization of the pumping light;
(2) A change in the intensity of the pumping light;
(3) A change in the spectral distribution of the pumping light associated with pressure shifts of the spectral lines;
(4) A change in the density of metastable atoms in the absorption cell;
(5) Changes in the magnetic field intensity.

Experimentally the sense of the circular polarization is obtained by the rotation of the quarter-wave plate following the linear polarizer.[9] Intensity changes are made by varying the angle between two linear polarizers.

[8] L. D. Schearer, Rev. Sci. Instr. 32, 1190 (1961).

[9] M. deWit of this laboratory has pointed out that a change in the sense of the circular polarization of the light beam relative to the external magnetic field produced by a physical rotation of the quarter-wave plate is necessarily equivalent in operation to a rotation of the light beam by 180° relative to the external magnetic field. The simultaneous rotation of the quarter-wave plate and light beam is equivalent to a space inversion; parity conservation then requires that the observables be unchanged.

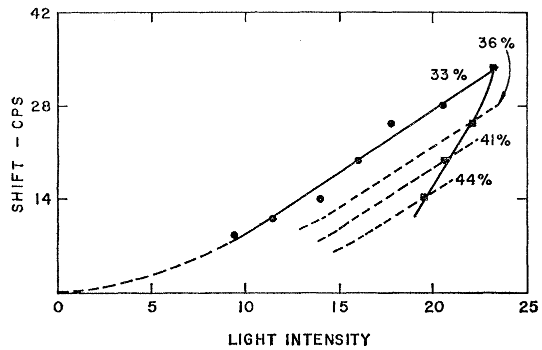

FIG. 3. Resonance displacement for $\sigma^+ \rightarrow \sigma^-$ as a function of light intensity. Light intensity is measured in arbitrary units. The lamp pressure is 2 mm Hg and the absorption cell pressure is 0.2 mm Hg.

The conditions described by 2 and 4 were performed in a gradient-free region (less than 8 cps per in.). The data from 1 and 3 were checked in the homogeneous environment.

EXPERIMENTAL RESULTS

It was found that the frequency shift of the resonance that occurred when the sense of the circular polarization of the pumping light was changed was very nearly linear with the intensity of the transmitted light. Further, it was observed that the magnitude of the shift for constant light intensity depended on the pressure of the lamp producing the pumping light as well as the density of the metastable atoms in the absorption cell. The frequency shifts of the resonance as a function of light intensity are shown in Figs. 3 through 8 for a variety of lamp and absorption cell pressures. The metastable density in the absorption cell is an additional variable, which is monitored by measuring the percentage of the resonance radiation absorbed by the cell. Figures 7–9 demonstrate the dependence of the shift on the pressure of the pumping lamp. Figure 10

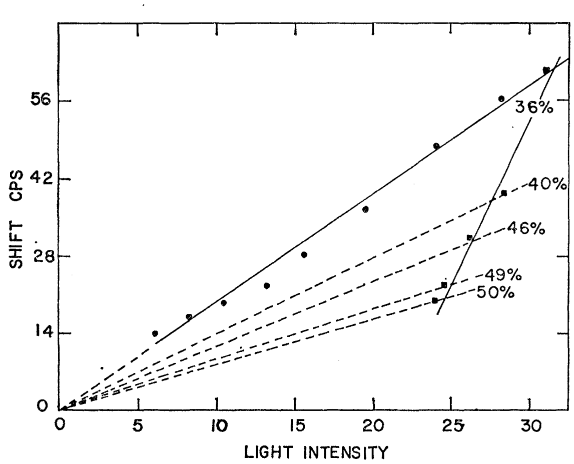

FIG. 4. Resonance displacement. Lamp pressure 5 mm Hg and cell pressure 0.2 mm Hg.

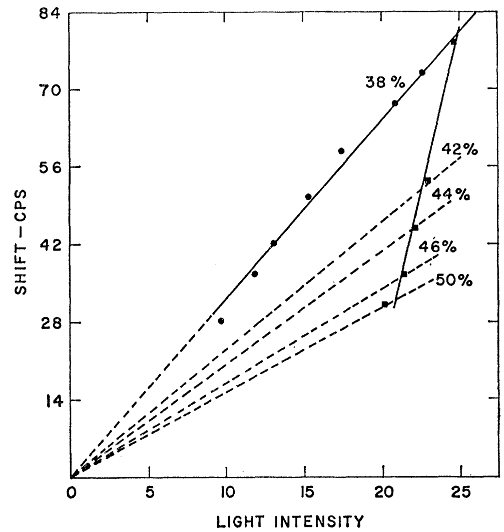

FIG. 5. Resonance displacement. Lamp pressure 7 mm Hg and cell pressure 0.2 mm Hg.

indicates the variation in signal size with percentage absorption for various conditions of lamp and cell pressure. The lamps are capillaries 0.6 cm in diameter with attached reservoirs to avoid changes in pressure during operation because of a clean-up of the helium. The absorption cell is a sphere 5 cm in diameter.

On the basis of the accumulated data the follo ing conclusions can be drawn: (a) The resonance shifts decrease nearly linearly with light intensity and appear to go to zero as the light intensity approaches zero; (b) the shift is smaller the lower the pressure of the lamp which provides the pumping light, other factors being

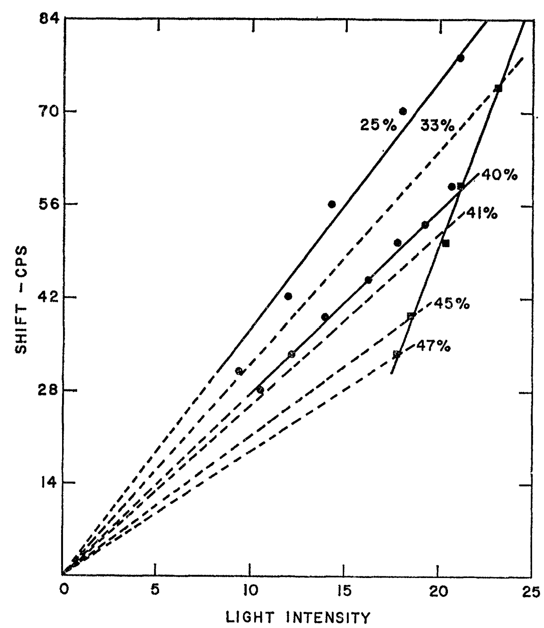

FIG. 6. Resonance displacement. Lamp pressure 7 mm Hg and cell pressure 1.4 mm Hg.

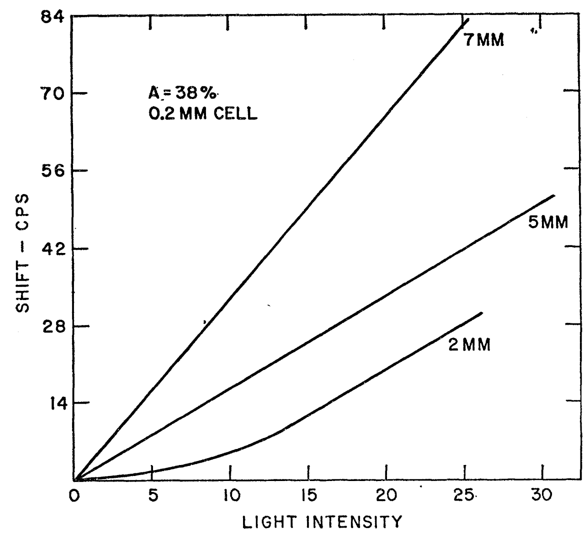

FIG. 7. Resonance displacement for $\sigma^+ \rightarrow \sigma^-$ for various lamp pressures. Cell pressure is 0.2 mm Hg. Total absorption is 38%.

equal; (c) the shift is independent of cell pressure, other things being equal, until the lamp pressure approaches the absorption cell pressure; and (d) the shift decreases as the percentage absorption increases. The relative shifts between the magnetic sublevels produced by the change in the polarization of the light was investigated at magnetic fields between 26 mG and 1.4 G. Over this range the shift was independent of field strength to within 3 cps.

DISCUSSION OF RESULTS

Heitler[10] considers the case of an atom in the state n_0 described by an energy E_0 and with an excited state n

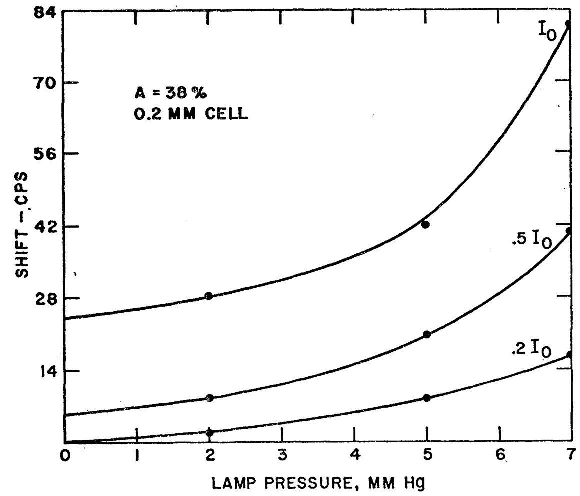

FIG. 9. Resonance shift for $\sigma^+ \rightarrow \sigma^-$ as a function of lamp pressure for different light intensities. Cell pressure is 0.2 mm Hg. 38% absorption.

and energy E_n which absorbs a photon of energy k_λ which is nearly the resonant frequency of the atom

$$k_\lambda \cong (E_n - E_0) = k_0, \tag{1}$$

with the subsequent emission of a photon k_σ. He writes the probability amplitudes for the various processes and derives an expression of the form

$$\frac{\hbar}{2}\Gamma(E) = i\sum \frac{|H_{\lambda|0}|^2}{(E-E_\lambda)+\frac{1}{2}i\hbar\gamma(E)}, \tag{2}$$

where the real part of $\Gamma(E)$ for $E=E_0$ is just the total transition probability per second from the initial state and the imaginary part is a change in the energy of the

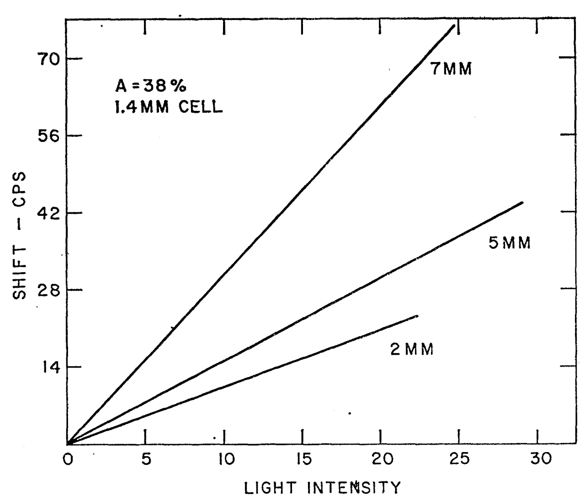

FIG. 8. Resonance displacement for various lamp pressures. Cell pressure is 1.4 mm Hg. Total absorption is 38%.

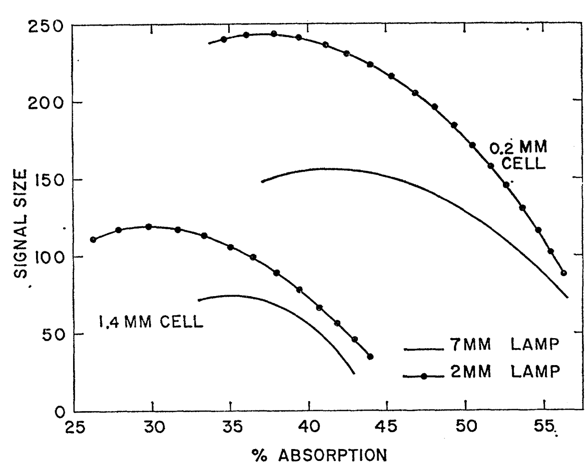

FIG. 10. Signal size dependence of % absorption. Total incident resonance radiation constant. The reduction in signal size at the higher cell pressure is associated with the increased mixing of the atoms in the excited ^3P states prior to reradiation back to the metastable state.

[10] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1954), 3rd ed., Sec. 20.

TABLE I. Relative absorption probabilities between the magnetic sublevels m' of the $2\,^3P$ states and the sublevels m of the $^2S_1$ state of helium for circularly polarized light.

m' \ m		$2\,^3P_2$ +2	+1	0	−1	−2	$2\,^3P_1$ +1	0	−1	Σ
					(a) σ^+					
	+1	6	0	0	0	0	0	0	0	6
$2\,^3S_1$	0	0	3	0	0	0	3	0	0	6
	−1	0	0	1	0	0	0	3	0	4
					(b) σ^-					
	+1	0	0	1	0	0	0	3	0	4
$2\,^3S_1$	0	0	0	0	3	0	0	0	3	6
	−1	0	0	0	0	6	0	0	0	6

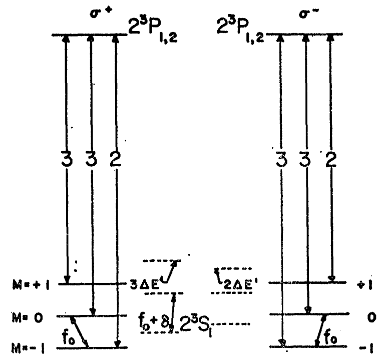

FIG. 11. Transition diagram showing displacement of magnetic sublevels in presence of radiation. For $k-k_0<0$ the levels are shifted in the opposite direction.

state n_0. $H_{\lambda|0}$ is the matrix element for absorption of k_λ, while the real and imaginary parts of γ are the total transition probabilities for emission from the excited state and the level shift of the excited state, respectively. Barrat and Cohen-Tannoudji[11] derive a similar expression

$$\frac{1}{2\tau}+i\Delta E'=\int_{-\infty}^{+\infty}\frac{u(k)|A_k|^2dk}{\frac{1}{2}\Gamma-i(k-k_0)},\qquad(3)$$

where $1/\tau$ is the width of the ground state, Γ is the natural width of the excited state, $\Delta E'$ is the energy shift of the ground state due to the absorption of photons out of the pumping beam, $k-k_0$ is the frequency difference between the emitting and absorbing atom, and $u(k)$ is the spectral distribution of the beam, and A_k depends upon the radial wave function of the atom and the intensity of the light. The sign of the energy shift then depends upon the sign of $k-k_0$ and its magnitude depends linearly on light intensity, some function of $k-k_0$, and the transition probabilities. Further, the relative displacement of the sublevels for a particular character of pumping light will be proportional to $\Delta E'$ and the difference in the transition probabilities for the levels. The situation is similar in many respects to the classical harmonic oscillator. When the oscillator is undamped it has a precise resonance frequency ν_0. When the classical oscillator is damped because of friction or the radiation of energy, the resonance not only acquires a finite width but the center of the resonance curve is shifted. If, in this analogy, the undamped resonance frequency ν_0 is identified with the magnetic sublevel energy and the damping with the induced transitions to the $2\,^3P_j$ state by the pumping light, a qualitative picture of the light shift emerges.

Table I shows the relative transition probabilities for absorption for various polarizations of the pumping light. The solid lines of Fig. 11 are the magnetic levels of the $2\,^3S_1$ state in the absence of the radiation field. The dashed lines show the energy shifts in terms of $\Delta E'$ for the cases indicated. The intensity of the light from the $2\,^3P_0-2\,^3S_1$ transition is taken to be zero although experimentally this light is about 25% of the total. The resonance signal is a weighted sum of the transitions taking place between adjacent sublevels of the $2\,^3S_1$ state. Any frequency difference between the adjacent levels will be averaged according to the degree to which it contributes to the resonance signal. Thus, if σ^+ light is used to pump the sample, the atoms tend to accumulate in the $m=+1$ level. The resonance, however, consists of transitions whose center is f_0 with an admixture of transitions centered at $f_0+\delta$. Based on these considerations, one then expects to find a variation in the frequency separation of the sublevels as the sense of polarization is changed from σ^+ to σ^-. The magnitude of $\Delta E'$ depends on the light intensity.

The dependence of the energy shift on the quantity $k-k_0$ can be demonstrated by varying the pressure of the helium gas in the pumping light. Helium spectral lines are particularly susceptible to pressure broadening and shift. Fred *et al.*[12] have observed experimentally an increase in the frequency of the spectral lines of both He^3 and He^4 with increasing lamp pressure. The shift is on the order of 0.01 cm^{-1} per mm Hg. Thus when the lamp pressure exceeds the absorption cell pressure, the center frequency of the emission line exceeds the center frequency of the absorption line, i.e., $k-k_0>0$, and the sublevels acquire an additional energy in the absorption process. This shift increases with increasing pressure as indicated in Figs. 7 and 8. Note that the energy shift of the levels disappears when the center of the emission line $u(k)$ coincides with k_0.

As a further check on the dependence of the shift on $k-k_0$ a second lamp containing He^3 is added with the light directed along the magnetic axis. About 2% of this light is absorbed in the sample cell indicating that the He^3 light induces relatively few real transitions. Depending upon the polarization of the He^3 light, the displacement of the resonance frequency is further increased or decreased. If the intensity of the He^3

[11] J. P. Barrat and C. Cohen-Tannoudji, Compt. rend. **252**, 93 (1961).

[12] M. Fred, F. S. Tomkins, J. K. Brody, and M. Hamermesh, Phys. Rev. **82**, 406 (1951).

light is sufficiently great, the sense of the displacement can be changed. The effect of the second lamp on the energy of the magnetic levels disappears if the beam is unpolarized.

Barrat and Cohen-Tannoudji[11] also suggested a model in which the optical pumping process couples a portion of the Zeeman energy of the excited state to the ground-state Zeeman levels. This effect might be expected to be important in the case of optical pumping in helium because the low absorption cell pressure permits the pumping cycle to take place without a reorientation of the atomic spins while an atom is in the P state. This effect should depend on some way on the magnetic field in that the Zeeman energy separation of the excited states is increasing with magnetic field. To test this possibility the experiment of Fig. 3 was repeated in magnetic fields from 26 mG to 1.4 G, a range of almost 50 to 1. There was no noticeable change in the shift. This lack of dependence on the field intensity also minimizes the possibility that the shifts are associated with the presence of polarized free electrons and/or helium ions.

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Hyperfine Structure of Cs^{134m}†

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The nuclear magnetic moment of Cs^{134m} has been measured independently of the electron-nucleus interaction energy ΔW by observing the separation of components of the doublet $(17/2, -13/2) \leftrightarrow (15/2, -15/2)$ and $(17/2, -15/2) \leftrightarrow (15/2, -13/2)$. The frequency difference $\delta\nu = 2g_I\mu_0 H$ is observed in the vicinity of a frequency minimum for each transition. The value obtained for μ_I is 1.0964(2) nm corrected for atomic diamagnetism, giving a hfs anomaly between Cs^{133} and Cs^{134m} of $-0.0139(2)$. The direct transition near zero field has been measured to give $\Delta\nu = 3\,684\,578\,640(175)$ cps.

INTRODUCTION

It has been shown[1] that s, and to a smaller extent p electrons, penetrate the finite volume occupied by the nucleus and, therefore, the interaction between the nucleus and the electrons will depend to a small extent upon the spatial distribution of nuclear charge and magnetization inside the nuclear volume.

The Goudsmit-Fermi-Segrè formula,[2–4]

$$\Delta W = \frac{16}{3}\pi\frac{\mu_I}{I}(2I+1)[\psi(0)]^2, \tag{1}$$

which gives a good approximate value of the interaction energy between an electron and the nucleus, considers the nucleus as a point magnetic dipole. The greatest inaccuracy in this formula is due to the uncertainty in the electron distribution. For two isotopes, however, the electron distributions would be nearly identical and one would expect that the relationship

$$\frac{\Delta\nu_1}{\Delta\nu_2}\frac{\mu_1}{\mu_1}\frac{I_1}{I_2}\frac{(2I_2+1)}{(2I_1+1)} = 1 \tag{2}$$

should hold to a high degree of precision.

If now we consider the differing distribution of nucleons in the two isotopic nuclei and the interaction with the penetrating electron, we must modify Eq. (2) slightly by the term ${}_1\Delta_2$ referred to as the hyperfine structure anomaly, defined by the relationship

$$1 + {}_1\Delta_2 = \frac{\Delta\nu_1\mu_2 I_1(2I_2+1)}{\Delta\nu_2\mu_1 I_2(2I_1+1)}. \tag{3}$$

The hyperfine structure anomaly was first considered by Bohr and Weisskopf, who found that ${}_1\Delta_2$ can be a sensitive function of the nuclear model describing the distribution of nucleon spin and magnetization associated with orbital motion. Measurements of ${}_1\Delta_2$ may therefore provide one test for nuclear models. Most anomalies heretofore reported have been less than 0.01.

The principal objective of this research was the

† Work done under the auspices of the U. S. Atomic Energy Commission.

* On leave from the University of Heidelberg, Heidelberg, Germany.

[1] A. Bohr and V. Weisskopf, Phys. Rev. **77**, 94 (1950). See for example, H. Kopfermann, *Nuclear Moments* (Academic Press Inc., New York, 1958), p. 123ff.

[2] S. Goudsmit, Phys. Rev. **43**, 636 (1933).

[3] E. Fermi and E. Segrè, Z. Physik **60**, 320 (1930).

[4] E. Fermi and E. Segrè, Z. Physik **82**, 729 (1933).