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Theory of Resonance Frequency Shift
Due to the Radiation Field*

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TABLE OF CONTENTS

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ABSTRACT

A new formalism is developed to calculate radiative transition processes, and applied to calculate the shift of resonance frequency due to the radiation field itself. The zeroth approximation gives the Bohr resonance condition, but a shift proportional to the photon density is obtained in the next approximation.

The first order shift is made of two terms: electric and magnetic. Both of them can be interpreted as the second order Stark effect and Zeeman effect due to the oscillating field respectively.

A comparison with experimental data on Cs is made. A good agreement is obtained by choosing the value of parameters suitably. These values of parameters can be checked by a future experiment.

HAMILTONIAN

The Hamiltonian of our system is made of three parts, namely, that of the radiation field, that of an atom in a vacuum, and the interaction between them. The eigen value of the radiation field is well known to be $\sum_k \hbar \omega_k n_k$, where ω_k is the frequency and n_k is the quantum number of the k-th photon. The eigen value for the atom in vacuum is actually very difficult to calculate, but can be measured by spectroscopic experiments. The interaction between the radiation field and the atom is

$$H_{int} = -\sum_i \left[\left(\frac{e_i}{\mu_i}\right) \vec{P}_i \cdot \vec{A} - \left(\frac{e_i^2}{2\mu_i}\right) A^2\right] - \sum_i (e_i/\mu_i)\, \vec{s}_i\, (\vec{\nabla} \times \vec{A}) \quad (1)$$

in the non-relativistic approximation. In this formula e_i, μ_i, s_i and P_i are the change, mass spin, and momentum of the i-th particle of the atom, and A is the vector potential at the position of the i-th particle.

Thus we write

$$H = H_{rad.} + H_{a.} + H_{int.} \quad (2)$$

This expression seems simple, but actually needs a long comment of renormalization of charge and mass[1]. By $H_{a.}$ we mean the Hamiltonian of the atom with renormalized quantities. In this case some parts of the interaction Hamiltonian is already included in the other part.

The atom is in a cavity which provides the boundary conditions for the field variable $\vec{A}$. Since $H_{rad.}$ is quadratic in $\vec{A}$, one can always find normal coordinates to satisfy such boundary condit ion and express $H_{rad.}$ as the sum of components due to each normal coordinate without interaction between them. Although some of these normal vibrations have a wave length comparable to the atomic dimension, only normal vibrations with longer wave length are excited in our case. We thus consider only normal vibration with a wave length very much larger that the atomic dimension. In that case we can expand $\vec{A}$ in (1) and take the first few terms only. Thus

$$H_{int.} \cong \sum_{\kappa}(e/\mu)\,\vec{P}\cdot\vec{A}_{o\kappa} - \sum_{\kappa}(Ze^2/2\mu)\,A^2_{o\kappa}$$
$$-\sum_{\kappa} i(e/2\mu)(\vec{L} + 2\vec{S}) \cdot (\vec{A}_{o\kappa} \times \vec{K}) \qquad (3)$$

where $\vec{P}$ is the total linear momentum of electrons, $\vec{A}_o$ is the amplitude of the κ-th normal coordinate, Z is the total number of electrons in this atom, $\vec{L}$ and $\vec{S}$ are total orbital and spin angular momenta, respectively, and K is the propagation vector of the κ-th normal coordinate. The normal coordinate is not necessarily a plane wave, but inside the atomic volume it can be approximated as a place wave. The propogation vector $\vec{K}$ should be interpreted in such an approximation.

It is convenient to take a representation in which $H_{rad.}$ and $H_{a.}$ are both diagonal. Neglecting the zero-point vibration of the field the diagonal terms of our Hamiltonians in such a representation are

$$(n_{\kappa} a\,|H_{rad.}|\,n_{\kappa} a) = n_{\kappa}\hbar\omega$$
$$(n_{\kappa} a\,|H_{a.}|\,n_{\kappa} a) \quad W_a \qquad (4)$$

In this formula n_{κ} is the quantum number of the κ-th mode, and W_a is the energy of the atom in its a-th state.

Using the matrix element formula

$$(n_{\kappa}|A_{o\kappa}|n_{\kappa}+1) = (n_{\kappa}+1|A^*_{o\kappa}|n_{\kappa}) = \left[\hbar(n_{\kappa}+1)/2\omega_{\kappa}\epsilon_o V\right]^{1/2} \qquad (5)$$

we have, for non-diagonal matrix elements,

$$(n_{\kappa}\, a\,|\,H_{int.}|\,n_{\kappa}+1\; b) = -i(e/2\mu)(\vec{e} \times \vec{K})\cdot(a|\vec{L} + 2\vec{S}|b)$$
$$\times\left[\hbar\,(n_{\kappa}+1)/2\omega_{\kappa}\epsilon_o V\right]^{1/2} G'_{\kappa} \qquad (6)$$
$$(n_{\kappa} a|H_{int.}|n_{\kappa}+1\; c) = i\hbar\omega_{ac}\,(a|\vec{e}\cdot\vec{M}_e\,|\,c)$$
$$\times\left[\hbar\,(n_{\kappa}+1)/2\omega_{\kappa}\epsilon_o V\right]^{1/2} G_{\kappa} \qquad (7)$$

$$(n_\kappa\, n_\lambda + 1a \,|\, H_{int.} |\, n_\kappa + 1\; n_\lambda\; a) = -(Ze^2/2\mu)(\hbar/2\,\epsilon_0 V)\, G_\kappa\, G_\lambda \times \left[(n_\kappa + 1)(n_\lambda + 1)/\omega_\kappa \omega_\lambda\right]^{1/2} \tag{8}$$

where

$$\omega_{ac} = (W_a - W_c)\,/\,\hbar \tag{9}$$

$$\vec{M}_e = e \sum_i \vec{r}_i \tag{10}$$

G and G' are electric and magnetic field amplitude,
and $\vec{e}$ is the unit vector parallel to $\vec{A}_0$, or the polarization vector.
Atomic state c is different from a and b but a and b can be the same.
Note that A^2 term has diagonal matrix elements, as well as non-diagonal matrix elements (8).

EIGEN VALUES AND TRANSITION PROBABILITY

It is very difficult to obtain eigen values of the Hamiltonian matrix we obtained above, but approximate results can be obtained by using the perturbation method. We are particularly interested in a degenerate case where

$$\omega_\kappa + \omega_{\beta\alpha} = 2\Delta/\hbar \tag{11}$$

is very small compared to ω itself. In such a case we take

$$\xi\, |n\;\; \alpha) + \eta\;\; |n + 1\; \beta) \tag{12}$$

as the zeroth approximation for the eigen function. In this formula ξ and η are the numerical coefficients and n is the quantum number for the κ-th mode, or n is the abreviation for our previous n . Solving 2 x 2 sub metrix formed by $|n\; a)$ and $|n + 1\; b)$ states given by (4) and (6) we have

$$E_{n,\alpha,\beta,\pm} \cong (n + 1/2)\,\hbar\omega + 1/2\,(W_\alpha + W_\beta) \pm \left[\Delta^2 + |\,(n\;\alpha|H_{int.}|\; n{+}1\;\beta)|^2\right]^{1/2} \tag{13}$$

where $(n\;\alpha|H_{int.}|n + 1\;\beta)$ can be either (6) or (7). We obtain

$$\xi_{\pm} = (n\,\alpha|H_{int.}|\,n+1\,\beta)\ /\ X\,Y_{\pm}\sqrt{2} \tag{14}$$

$$\eta_{+} = |\xi_{-}| \qquad \eta_{-} = -|\xi_{+}| \tag{15}$$

where

$$X = \left[\Delta^2 + |(n\,\alpha|H_{int.}|n+1\,\beta)|^2\right]^{1/4} \tag{16}$$

$$Y_{\pm} = \left[X^2 \mp \Delta\right]^{1/2} \tag{17}$$

From (6) and (7) we see ξ_{+} and ξ_{-} are both pure imaginary.

If we set the above two approximate eigen functions as

$$\psi_{+} = \xi_{+}\,|n\,\alpha) + \eta_{+}\,|n+1\beta) \tag{18}$$

$$\psi_{-} = \xi_{-}\,|n\,\alpha) + \eta_{-}\,|n+1\beta) \tag{19}$$

we see

$$|n\,\alpha) = \xi_{+}\,\psi_{+} + \eta_{+}\psi_{-} \tag{20}$$

$$|n+1\beta) = i\eta_{+}\psi_{+} - \xi_{+}\,\psi_{-} \tag{21}$$

Since the wave function which is $|n\,\alpha)$ at $t = 0$ is

$$|\xi_{+}|\psi_{+}\exp(-iE_{+}t/\hbar) + \eta_{+}\psi_{-}\exp(-iE_{-}t/\hbar) \tag{22}$$

the probability of finding state $|n+1\beta)$ at time t starting from state $|n\,\alpha)$ at $t = 0$ is

$$S\,(n\alpha \rightarrow n+1\beta,\ t)$$

$$= \left|\int(-i\,\eta_{+}\psi_{+}^{*} - \xi_{+}^{*}\psi_{-}^{*})\left[\xi_{+}\,\psi_{+}\exp(-iE_{+}t/\hbar) + \eta_{+}\psi_{-}\exp(-iE_{-}t/\hbar)\right]dv\right|^2$$

$$= \left|-i|\xi_{+}|\eta_{+}\exp(-iE_{+}t/\hbar) - \xi_{+}^{*}\,\eta_{+}\exp(-iE_{-}t/\hbar)\right|^2$$

$$= 2|\eta_{+}|^2|\xi_{+}|^2\left[1-\cos(2X^2\,t/\hbar)\right] \tag{23}$$

$$= 2\,|(n\alpha\,|H_{int.}|\,n+1\beta)|^2\left[1-\cos(2X^2t/\hbar)\right]/2X^4$$

Since

$$\frac{1}{\pi} \lim_{t \to \infty} \frac{1 - \cos xt}{x^2 t} = \delta(x) \tag{24}$$

we see

$$\lim_{t \to \infty} S(n\alpha \longrightarrow n + 1\beta, t) =$$
$$= (2\pi/\hbar^2) \left|(n\ \alpha \left| H_{int.} \right| n + 1\beta)\right|^2 \ \delta(2X^2/\hbar)\ t \tag{25}$$

If we put (16) in we have

$$\delta(2X^2/\hbar) \cong \delta(2\Delta/\hbar) = \delta(\omega_{\kappa} + \omega_{\beta\alpha}) \tag{26}$$

The $(n\ \alpha \left| H_{int.} \right| n + 1\beta)$ part of Δ will produce the natural line width if not neglected. Since we are not interested in the natural line width now we just neglected the matrix element of $H_{int.}$ in (26).

Equation (25) gives the familiar transition probability formula and (26) gives the Bohr's expression of the resonance frequency

$$\omega = \omega_{\alpha\beta} \tag{27}$$

FREQUENCY SHIFT

The conventional results obtained in the previous section can be improved by taking higher order approximation to our eigen value over (13).

The ordinary second order perturbation method can be used for that purpose. According to (6), (7), and (8) our state

$$\xi |nn'\alpha) + \eta |n+1\ n'\beta) \tag{28}$$

interacts with the states listed in Table I. Calculating matrix elements we have

$$E_{n,\alpha,\beta,\pm} = n'\hbar\omega' + (n+1/2)\hbar\omega + 1/2\,(W_\alpha + W_\beta) \pm X^2$$
$$+ \sum_{\substack{n=n,n' \\ \omega=\omega,\omega'}} \left\{ \left|\xi_\pm\right|^2 \sum_\gamma |(\alpha | D | \gamma)|^2 (\omega \epsilon_0 V)^{-1} \left[n(3\omega + \omega_{\alpha\gamma} + \omega_{\beta\gamma})^{-1} + \right.\right.$$

$$+(n+1)(-\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}\Big]+|\xi_{\pm}|^2\ (Ze^2/2\mu)(\hbar/2\epsilon_o V\omega)(2n+1)$$

$$+|\eta_{\pm}|^2\sum_{\gamma}|(\beta|D|\gamma)|^2(\omega\epsilon_o V)^{-1}\Big[(n+1)(\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}+$$

$$+(n+2)(-2\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}\Big]+|\eta_{\pm}|^2\ (Ze^2/2\mu)(\hbar/2\epsilon_o V\omega)(2n+3)\Big\}$$

$$-\Big[|\xi_{\pm}|^2\ |(\alpha|D|\alpha)|^2+|\eta_{\pm}|^2\ |(\beta|D|\beta)|^2\Big](2\omega^2\epsilon_o V)^{-1}$$

$$-\Big[2|\xi_{\pm}|^2|\eta_{\pm}|^2-\xi_{\pm}|\xi_{\pm}|\eta_{\pm}^{*2}-\xi_{\pm}^{*}|\xi_{\pm}|\eta_{\pm}^{2}\Big](n+1)(\alpha|D|\alpha)(\beta|D|\beta)(\omega^2\epsilon_o V)^{-1}$$

$$+\Big[n+2|\eta_{\pm}|^2\Big]|(\alpha|D|\beta)|^2\ (4\omega^2\epsilon_o V)^{-1}$$

$$-|\xi_{\pm}|^2\Big\{\Big[(\xi_{\pm}\eta_{\pm}^{*}-|\xi_{\pm}|\eta_{\pm})(\beta|D|\alpha)+(\xi_{\pm}^{*}\eta_{\pm}-|\xi_{\pm}|\eta_{\pm}^{*})$$
$$(\alpha|D|\beta)\Big]n(n-1)^{1/2}$$

$$-\Big[(\xi_{\pm}\eta_{\pm}^{*}+|\xi_{\pm}|\eta_{\pm})(\beta|D|\alpha)+(\xi_{\pm}^{*}\eta_{\pm}+|\xi_{\pm}|\eta_{\pm}^{*})(\alpha|D|\beta)\quad(n+2)$$
$$(n+1)^{1/2}\Big\}\quad(Ze^2/2\mu)\hbar^{1/2}\ (2\omega)^{-5/2}\ (\epsilon_o V)^{-3/2}$$

$$-\Big\{\Big[\xi_{\pm}\ \eta_{\pm}^{*}\quad(\beta|D|\alpha)+\xi_{\pm}^{*}\ \eta_{\pm}\ (\alpha|D|\beta)\Big]\ n(n+1)^{1/2}$$

$$+(|\xi_{\pm}|^2-|\eta_{\pm}|^2)\Big[\xi_{\pm}\ \eta_{\pm}^{*}\ (\beta|D|\alpha)-\xi_{\pm}^{*}\ \eta_{\pm}(\alpha|D|\beta)\Big](n+2)(n+3)^{1/2}\Big\}$$

$$(Ze^2/2\mu)\hbar^{1/2}\ (2\omega)^{-5/2}\ (\epsilon_o V)^{-3/2}$$

$$-\Big[|\xi_{\pm}|^2\ (4n+2)+|\eta_{\pm}|^2(4n+6)+(2|\xi_{\pm}|^2|\eta_{\pm}|^2-\xi_{\pm}|\xi_{\pm}|\eta_{\pm}^{*2}-\xi_{\pm}^{*}|\xi_{\pm}|\eta_{\pm}^{2})$$
$$\Big\{(n+2)(n+1)^{1/2}\ (n+3)^{1/2}-n(n^2-1)^{1/2}\Big\}\Big]\ x$$

$$x\ (Ze^2/2\mu)^2\ (\hbar/4\omega^3\epsilon_o^2\ V^2)$$

$$+|\xi_{\pm}|^2\ (Ze^2/2\mu)^2(\hbar/2\omega\omega'\epsilon_o^2V^2)\ \Big[(n+1)\ n'\ (-\omega+2\omega'+\omega_{\beta\alpha})^{-1}$$
$$+n(n'+1)(3\omega-2\omega'+\omega_{\beta\alpha})^{-1}\Big]$$

$$+|\eta_{\pm}|^2\ (Ze^2/2\mu)^2(\hbar/2\omega\omega'\epsilon_o^2)\ \Big[(n+2)\ n'(-3\omega+2\omega'-\omega_{\beta\alpha})^{-1}$$
$$+(n+1)(n'+1)(\omega-2\omega'-\omega_{\beta\alpha})^{-1}\Big]\qquad(29)$$

where

$$D = i(e/2\mu)\left[-(\vec{e}\times\vec{K})\cdot(\vec{L}+2\vec{S})\,G_m + 2(H_{a.}\,\vec{e}\cdot\vec{M}_e - \vec{e}\cdot\vec{M}_e\,H_{a.})\,\mu^{-1}G_e\right] \tag{30}$$

According to the discussion in the previous section $E_{n,a,b,+} - E_{n,a,b,-}$ gives the resonance frequency. In calculating this quantity from (29) the following formula is useful.

$$|\xi_+|^2 - |\xi_-|^2 = \Delta/X^2 = |\eta_-|^2 - |\eta_+|^2 \tag{31}$$

We see from (29) and (31)

$$E_{n,\alpha,\beta,+} - E_{n,\alpha,\beta,-} = 2X^2 - 2\Omega(\Delta/X^2) \tag{32}$$

where

$$\begin{aligned}\Omega = \sum_{\substack{n=n,n'\\ \omega=\omega,\omega'}} \Big\{ &-\sum_\gamma |(\alpha|D|\gamma)|^2\,(2\omega\epsilon_o V)^{-1}\left[n(3\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1} + \right.\\ &\left. + (n+1)(-\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}\right]\\ &+\sum_\gamma|(\beta|D|\gamma)|^2\,(2\omega\epsilon_o V)^{-1}\left[(n+1)(\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}+\right.\\ &\left.(n+2)(-3\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}\right] + (Ze^2/2\mu)(\hbar/2\epsilon_o V\omega)\Big\}\\ &+\left[|(\alpha|D|\alpha)|^2 - |(\beta|D|\beta)|^2 + |(\alpha|D|\beta)|^2\right](4\omega^2\epsilon_o V)^{-1}\\ &\quad - |(\alpha|D|\beta)|^2\left[(n+2)(n+1)^{1/2} - n(n-1)^{1/2}\right]X^{-2}(Ze^2/8\mu)\hbar^{1/2}\\ &\quad \times(2\omega)^{-5/2}\,(\epsilon_o V)^{-3/2}\\ &-(Ze^2/2\mu)^2\,(\hbar/2\omega^3\epsilon_o^2V^2)\\ &-(Ze^2/2\mu)^2\,(\hbar/4\omega\omega'\epsilon_o^2V^2)\left[(n+1)\,n'\,(-\omega+2\omega'+\omega_{\beta\alpha})^{-1}\right.\\ &+n(n'+1)(3\omega-2\omega'+\omega_{\beta\alpha})^{-1} - (n+2)\,n'\,(-3\omega+2\omega'-\omega_{\beta\alpha})^{-1}\\ &\quad\left.-(n+1)(n'+1)(\omega-2\omega'-\omega_{\beta\alpha})^{-1}\right]\end{aligned} \tag{33}$$

Since from (32)

$$\begin{aligned}E_{n,\alpha,\beta,+} - E_{n,\alpha,\beta,-} &= 2\,(X^4 - \Omega\Delta)\,X^{-2}\\ &= 2\,(\Delta + \Omega/2)^2/\Delta\end{aligned} \tag{34}$$

if $(n\,\alpha \mid H_{int.} \mid n{+}1\,\beta)$ and Ω^2 are both negligible. The resonance condition is thus

$$\omega = \omega_{\alpha\beta} + \Omega/\hbar \qquad (35)$$

that means $\Omega/\hbar$ is the first order correction to the resonance frequency.

Since n and n' are large numbers we can neglect 1 or 2 compared to them. We see in (33) that Ω contains terms with different powers in n and n'. Neglecting small terms we can write

$$\begin{aligned}\Omega \cong\; & + n\Big\{-\sum_\gamma |(\alpha|D|\gamma)|^2\,(2\omega\,\epsilon_o V)^{-1}\big[(3\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}+(-\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}\big]\\ &\quad + \sum_\gamma |(\beta|D|\gamma)|^2\,(2\omega\,\epsilon_o V)^{-1}\big[(\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}+(-3\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}\big]\Big\}\\ & + n'\Big\{-\sum_\gamma |(\alpha|D|\gamma)|^2\,(2\omega'\,\epsilon_o V)^{-1}\big[(3\omega'+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}+(-\omega'+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}\big]\\ &\quad + \sum_\gamma |(\beta|D|\gamma)|^2\,(2\omega'\,\epsilon_o V)^{-1}\big[(\omega'+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}+(-3\omega'+\omega_{\alpha\gamma}+\omega_{\beta\gamma})^{-1}\big]\Big\}\\ & + nn'\,(Ze^2/2\mu)^2\,(\hbar/2\omega\omega'\epsilon_o^2V^2)\big[(\omega-2\omega'-\omega_{\beta\alpha})^{-1}-(3\omega-2\omega'+\omega_{\beta\alpha})^{-1}\big]\end{aligned} \qquad (35)$$

Operator D can be either electric dipole term given in (7) or magnetic dipole given in (6). Matrix elements of the electric dipole term is about 10^6 times those of the magnetic dipole term. The selection rule, however, is such that the magnetic dipole can combine almost degenerate states, but that is impossible for the electric dipole. The denominators in (35) thus can make the magnetic dipole term as important as the electric dipole term.

First examine the electric dipole term (7). Using (27) we see that the part of Ω due to the electric dipole and proportional to n is

$$\begin{aligned}\Omega_e &= n\,G_e^2\,(4\omega\,\epsilon_o V)^{-1}\Big\{-\sum_\gamma |(\alpha|\vec{e}\cdot\vec{M}_e|\gamma)|^2\,\omega_{\alpha\gamma}^2\big[(\omega+\omega_{\alpha\gamma})^{-1}+(-\omega+\omega_{\alpha\gamma})^{-1}\big]\\ &\quad + \sum_\gamma |(\beta|\vec{e}\cdot\vec{M}_e|\gamma)|^2\,\omega_{\beta\gamma}^2\big[(\omega+\omega_{\beta\gamma})^{-1}+(-\omega+\omega_{\beta\gamma})^{-1}\big]\Big\}\\ &= n\omega\,G_e^2\,(4\,\epsilon_o V)^{-1}\Big\{-\sum_\gamma |(\alpha|\vec{e}\cdot\vec{M}_e|\gamma)|^2\big[(\omega+\omega_{\alpha\gamma})^{-1}+(-\omega+\omega_{\alpha\gamma})^{-1}\big]\\ &\quad + \sum_\gamma |(\beta|\vec{e}\cdot\vec{M}_e|\gamma)|^2\big[(\omega+\omega_{\beta\gamma})^{-1}+(-\omega+\omega_{\beta\gamma})^{-1}\big]\Big\}\\ &= (\rho_e/4\epsilon_o)\big[-\alpha_{e\alpha}(\omega)+\alpha_{e\beta}(\omega)\big]\end{aligned} \qquad (36)$$

where the dispersion relation[1] is used by adding zero term

$$+ (Ze^2 G_e^2/2\mu)(\hbar/\epsilon_o V\omega)(n-n) \qquad (37)$$

to the first expression. In the last expression of (36) we defined the electric field energy density

$$\rho_e = n \hbar \omega G_e^2/V \qquad (38)$$

and the polarizability of the atom $\alpha_{e\alpha}$ and $\alpha_{e\beta}$ in the initial and the final states, respectively. Note that for a standing wave G^2 and ρ depend on the position. Since the energy density is

$$\rho_e = \epsilon_o \mathcal{E}^2/2 \qquad (39)$$

where $\mathcal{E}$ is the average electric field intensity of the radiation field at the position where the atom is, we see the dipole term (36) is

$$\Omega_e = \alpha_{e\beta}(\omega)\, \mathcal{E}^2/2 - \alpha_{e\alpha}(\omega)\mathcal{E}^2/2 \qquad (40)$$

or the resonance frequency shift is given by the difference of the average Stark shift of the initial and the final state due to the radiation field itself.

The shift due to the magnetic dipole term is

$$\begin{aligned}\Omega_m = {} & n\, G_m^2 (4\omega \epsilon_o V)^{-1}\Big\{\sum_\gamma |(\alpha|(\vec{e}\times\vec{K})\cdot\vec{M}_m|\gamma)|^2\big[(\omega+\omega_{\alpha\gamma})^{-1} + (-\omega+\omega_{\alpha\gamma})^{-1}\big] \\ & + \sum_\gamma |(\beta|(\vec{e}\times\vec{K})\cdot\vec{M}_m|\gamma)|^2\big[(\omega+\omega_{\beta\gamma})^{-1} + (-\omega+\omega_{\beta\gamma})^{-1}\big]\Big\} \\ = {} & B^2 (2|K|^2)^{-1}\Big\{-\sum_\gamma |(\alpha|(\vec{e}\times\vec{K})\cdot\vec{M}_m|\gamma)|^2\big[(\omega+\omega_{\alpha\gamma})^{-1} + (-\omega+\omega_{\alpha\gamma})^{-1}\big] \\ & + \sum_\gamma |(\beta|(\vec{e}\times\vec{K})\cdot\vec{M}_m|\gamma)|^2\big[(\omega+\omega_{\beta\gamma})^{-1} + (-\omega+\omega_{\beta\gamma})^{-1}\big]\Big\}\end{aligned} \qquad (41)$$

where

$$\vec{M}_m = (e\ \ /2\mu)(\vec{L} + 2\vec{S}) \qquad (42)$$

or the magnetic dipole moment, and B is the average magnetic flux density of the radiation field itself. Formula (41) can again be interpreted in terms of the ordinary second order Zeeman effect.

APPLICATION TO Cs 9 kMc LINE

The ground state of the Cs atom is $^2S_{1/2}$ and the nuclear spin of 7/2 split it into F=4 and 3 states. F=4 state is higher in energy than F=3 state by about 9 kMc. Beehler, Snider and Mockler[2] observed the change of the resonance frequency due to the field intensity. In his experiment atomic degeneracy is removed by the static magnetic field B_o so that our theory is applicable. The transition $F = 4 \leftrightarrow 3$ is the magnetic dipole transition.

There are two cases, namely, if the magnetic component of the microwave is parallel or perpendicular to the external static magnetic field. They are called σ and π cases, respectively. The selection rule for the magnetic dipole transition is

$$\Delta M = 0 \quad \text{for } \sigma \text{ case}$$

$$\Delta M = \pm 1 \text{ for } \pi \text{ case} \tag{42}$$

Let us consider the electric dipole shift first. Since the microwave frequency is very low the polarizability to be used in formula (40) is almost equal to that with a static field.

In Beehler, Snider, and Mockler's experiment the electric component of the microwave is always perpendicular to the static magnetic field. Haun and Zacharias[3] observed the Stark effect of $(F=3,\ M=0) \leftrightarrow (F=4,\ M=0)$ transition presumably in the same situation and obtained

$$\Delta E/h = -2.9 \times 10^{-2} \mathcal{E}^2 \text{ cps} \tag{43}$$

where $\mathcal{E}$ is the electric field in volts/m. We thus expect for $M = 0 \leftrightarrow 0$ transition

$$\Omega_e(0 \leftrightarrow 0)/h = -2.9 \times 10^{-2}\ \rho_e/\epsilon_o \text{ cps}$$

$$= -3.3 \times 10^{9}\ \rho_e \quad \text{cps} \tag{44}$$

where ρ_e is the electric energy density of the microwave field in joul/m^3.

The theory of the Stark effect (43) is proposed by the present author[4]. According to that theory we expect the energy of both F=3 and F=4 states to change as $(\Delta E_o + \Delta E_1 M^2)\mathcal{E}^2$ with different constants due to the external electric field. Thus the electric shift of the π- transitions is expected to be given by

$$\Omega_e\,(M\leftrightarrow M)/h = (-3.3 \times 10^9 + DM^2)\rho_e \quad \text{cps} \tag{45}$$

and that of the σ-transition must be

$$\Omega_e\,(F{=}3,\ M\leftrightarrow F{+}4,\ M\pm 1)/h = (-3.3 \times 10^9 + C\pm 2CM + DM^2)\rho_e \quad \text{cps} \tag{46}$$

where C and D are constants.

As for the magnetic dipole shift we see that it is negligible in the σ transitions, since in the experiment such a transition is induced with microwaves whose B component is completely parallel to the external static magnetic field.

$$\Omega_m\,(M \longleftrightarrow M) = 0 \tag{47}$$

In the π-type transition, on the other hand, we have the selection rule $\Delta M = \pm 1$, which makes the magnetic dipole term important in the frequency shift Ω. If the microwave is linearly polarized, as in the present cases, the transition (F=3 M) ↔ (F=4, M +1), for example, is perturbed by (F=3, M) ↔ (F=4. M -1) very strongly since $|-\omega+\omega_{\alpha\gamma}|$ is equal to the separation of (F=4, M +1) level and (F=4, M -1) level which is very small compared to ω. Other transitions (F=3, M) ↔ (F=3, M +1), (F=4, M +1) ↔ (F=4, M), and (F=4,M+1) ↔ (F=4,M+2) also contribute to Ω to some, but to a much smaller extent. If the static magnetic field is B_o the additional energy of F=4 and 3 states is

$$-B_o\,(e\hbar/\mu)\,M/8 \qquad \text{for } F{=}3 \tag{48}$$

$$B_o\,(e\hbar/\mu)\,M/8 \qquad \text{for } F{=}4 \tag{49}$$

First consider (F=3,M) ↔ (F=4,M+1) transition. The frequency of this transition is

$\omega_o + B_o(e\hbar/8\mu)(2M+1)$ in the zeroth approximation.

The Shift due to (F=3,M) ↔ (F=4,M-1) is, from (41)

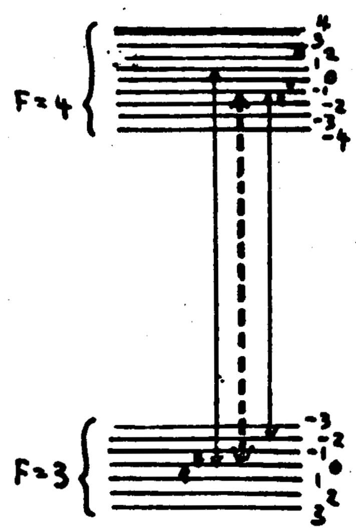


Fig. 1. Transitions Perturbing on (F=3,M_F=0) (F=4, M_F=-1) Frequency.

$$(G_m^2\ n\ \omega\ \mu_o/256V)(4-M)(5-M)(e\hbar/\mu)^2\ \left[(4\mu/B_oe) - (2\omega_o)^{-1}\right] \quad (50)$$

while that due to $(F=3,\ M+2) \leftrightarrow (F=4,\ M+1)$ is

$$-(G_m^2\ \ n\ \omega\ \mu_o/256V)(2-M)(3-M)\ \ (e\hbar/\mu)^2\ \left[(2\omega_o)^{-1} + (4\ \mu/B_oe)\right] \quad (51)$$

Neglecting $(2\omega_o)^{-1}$ terms we have

$$\Omega_m/h = G_m^2\ n\ \hbar\omega\ \mu_o(e\hbar/64\ V\ hB_o\ \mu)\ \left[(4-M)(5-M) - (2-M)(3-M)\right]$$

$$= 1100\ (\rho_m/\ B_o)(7-2M)\ \ \text{cps} \quad (52)$$

where ρ_m is the magnetic field energy density, and ρ_m and B_o are in the mks unit.

The same consideration gives the following formula for the magnetic shift of $(F=3,\ M) \longleftrightarrow (F=4,\ M-1)$ transition.

$$\Omega_m/h\ \ = -1100\ (\rho/B_o)(7+2M) \quad (53)$$

COMPARISON WITH EXPERIMENT

In Beehler, Snider, and Mockler's experiment input power was measured, but the energy densities ρ_e and ρ_m in the cavity were not. We believe, however, that the input power is proportional to ρ_e and ρ_m. Since the cavity has Q of about 5000, we estimate that input power of 1 mw corresponds to ρ of about 10^{-9} joules/m^3. This estimation. however, can be wrong by a factor of 100.

Transitions so far observed are listed in Table II. Since they did not observe any shift in $M=0 \leftrightarrow 0$ transition it means $-1.64 \times 10^9 \rho_e$ cps is negligible in the observed range of input power up to about 10 mw.

Theoretical curves are compared with experimental data with the following choice of parameters

$$D\rho_e/\text{inp.} \qquad = +1.9 \quad \text{cps/mw} \quad (54)$$

$$C\rho_e/\text{inp.} \qquad = -3.2 \quad \text{"} \quad (55)$$

$$4 \times 10^{8}\ \rho_m / \text{inp} = 4.7 \quad \text{cps/mw} \tag{56}$$

where inp. is the input power in mw. Equation (56) shows that

$$\rho_m = 1.2 \times 10^{-8}\ \text{joules/m}^3 \tag{57}$$

at 1 mw input, while from $M=0 \longleftrightarrow 0$ transition we see

$$\rho_e < 10^{-10}\ \text{joules/m}^3 \tag{58}$$

at 1 mw input. These values are reasonable. A reasonable agreement is shown in the medium power range except in one case. In the lowest power region the experimental inaccuracy is supposed to be large. In the higher input region the experiment shows large deviation from linear behavior. The reason is not known yet.

A complete disagreement is seen in $F=4$, $M=-2 \longleftrightarrow F=3$, $M=-3$ case.

NOTES

1. For example, A. I. Akhiezer and V. B. Berestetsky, "Quantum Electrodynamics, AEC-tr.-2876, p. 360

2. Beehler, Snider, and Mockler, private communication

3. R. D. Haun, Jr. and J. R. Zacharias, Phys. Rev. 107, 107 (1957).

4. M. Mizushima, to be published.

TABLE I.

STATES INTERACTING WITH

$\xi \,|\, n\, n'\, \alpha) + \eta \,|\, n+1\, n'\beta)$

STATES	ENERGY DIFFERENCE	MATRIX ELEMENTS GIVEN BY
$\|n-1\gamma)$	$(\hbar/2)(3\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})$	$\xi^*(n\alpha\|H_{int.}\| n-1\gamma)$
$\|n\gamma)$	$(\hbar/2)(\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})$	$\eta^*(n+1\beta\|H_{int.}\|n\gamma)$
$\|n+1\gamma)$	$(\hbar/2)(-\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})$	$\xi^*(n\alpha\|H_{int.}\| n+1\gamma)$
$\|n+2\gamma)$	$(\hbar/2)(-3\omega+\omega_{\alpha\gamma}+\omega_{\beta\gamma})$	$\eta^*(n+1\beta\|H_{int.}\|n+2\gamma)$
$\xi\|n-2\alpha)+\eta\|n-1\beta)$	$2\hbar\omega$	$\xi^*(n\alpha\|H_{int.}\|n-1\beta)$ $+\|\xi\|^2(n\alpha\|H_{int.}\|n-2\alpha)$ $+\|\eta\|^2(n+1\beta\|H_{int.}\|n-1\beta)$
$\eta\|n-2\alpha)-\|\xi\|\|n-1\beta)$		$-\xi^*\|\xi\|(n\alpha\|H_{int.}\|n-1\beta)$ $+\xi^*\eta(n\alpha\|H_{int.}\|n-2\alpha)$ $-\eta^*\|\xi\|(n+1\beta\|H_{int.}\|n-1\beta)$
$\xi\|n-1\alpha)+\eta\|n\beta)$	$\hbar\omega$	$\|\xi\|^2(n\alpha\|H_{int}\|n-1\alpha)$ $+\|\eta\|^2(n+1\beta\|H_{int.}\|n\beta)$
$\eta\|n-1\alpha)-\|\xi\|\|n\beta)$		$\xi^*\eta(n\alpha\|H_{int.}\|n-1\alpha)$ $-\|\xi\|\eta^*(n+1\beta\|H_{int.}\|n\beta)$
$\xi\|n+1\alpha)+\eta\|n+2\beta)$	$-\hbar\omega$	$\|\xi\|^2(n\alpha\|H_{int.}\|n+1\alpha)$ $+\|\eta\|^2(n+1\beta\|H_{int.}\|n+2\beta)$
$\eta\|n+1\alpha)-\|\xi\|\|n+2\beta)$		$\xi^*\eta(n\alpha\|H_{int.}\|n+1\alpha)$ $-\|\xi\|\eta^*(n+1\beta\|H_{int.}\|n+2\beta)$
$\xi\|n+2\alpha)+\eta\|n+3\beta)$	$-2\hbar\omega$	$\xi\eta^*(n+1\beta\|H_{int.}\|n+2\alpha)$ $+\|\xi\|^2(n\alpha\|H_{int.}\|n+2\alpha)$ $+\|\eta\|^2(n+1\beta\|H_{int.}\|n+3\beta)$

CONTINUED

TABLE I. CONTINUED

STATES	ENERGY DIFFERENCE	MATRIX ELEMENTS GIVEN BY
$\eta\|n+2\alpha) - \|\xi\|\,\|n+3\beta)$	$-2\hbar\omega$	$\|\eta\|^2(n+1\beta\|H_{int.}\|n+2\alpha)$ $+\xi^*\eta(n\alpha\|H_{int.}\|n+2\alpha)$ $-\|\xi\|\eta^*(n+1\beta\|H_{int.}\|n+3\beta)$
$\xi\|nn'\alpha) + \eta\|n+1n'\beta)$	0	$-(Ze^2/2\mu)(\hbar/2\epsilon_0 V\omega)[\|\xi\|^2(2n+1) + \|\eta\|^2(2n+3)]$
$\eta\|nn'\alpha) - \|\xi\|\,\|n+1\ n'\beta)$	0	$-(Ze^2/2\mu)(\hbar/2\epsilon_0 V\omega)$ $[\xi^*\eta(2n+1) - \|\xi\|\eta(2n+3)]$
$\|n+1\ n'-1\alpha)$	$(\hbar/2)(-\omega-2\omega'+\omega_{\beta\alpha})$	$\xi^*(nn'\alpha\|H_{int.}\|n+1\ n'-1\alpha)$
$\|n-1\ n'+1\alpha)$	$(\hbar/2)(3\omega-2\omega'+\omega_{\beta\alpha})$	$\xi^*(nn'\alpha\|H_{int.}\|n-1\ n'+1\alpha)$
$\|n+2\ n'-1\beta)$	$(\hbar/2)(-3\omega+2\omega-\omega_{\beta\alpha})$	$\eta^*(n+1\ n'\beta\|H_{int.}\|n+2\ n'-1\beta)$
$\|n\ n'+1\ \beta)$	$(\hbar/2)(\omega-2\omega'-\omega_{\beta\alpha})$	$\eta^*(n+1\ n'\beta\|H_{int.}\|n\ n'+1\ \beta)$

TABLE II

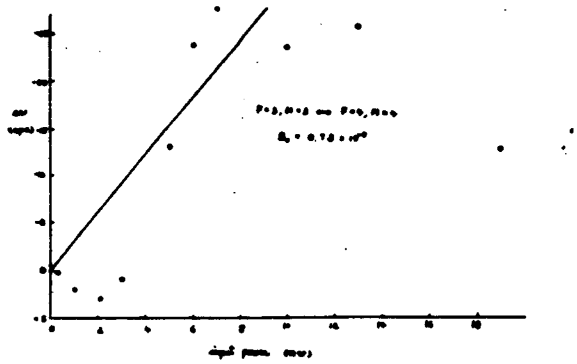
THEORETICAL SHIFTS IN cps. (ρ, and B_o are in kMs unit)

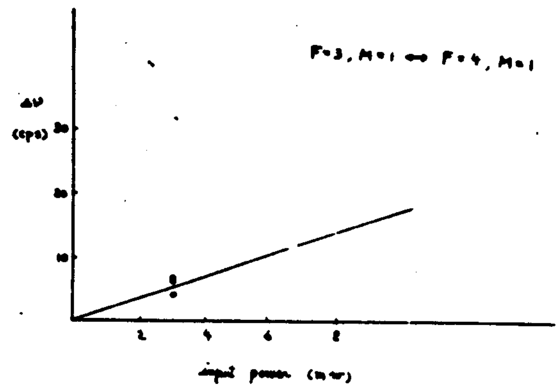
M(F=3)	M(F=4)	ELECTRIC	MAGNETIC
0	0	$-3.3 \times 10^9 \rho_e$	0
± 1	± 1	$(-3.3 \times 10^9 + D) \rho_e$	0
± 2	± 2	$(-3.3 \times 10^9 + 4D) \rho_e$	0
± 3	± 3	$(-3.3 \times 10^9 + 9D) \rho_e$	0
+3	+2	$(-3.3 \times 10^9 - 5C + 9D) \rho_e$	$\pm 1.43 \times 10^4 (\rho_m/B_o)$
+2	+1	$(-3.3 \times 10^9 - 3C + 4D) \rho_e$	$\pm 1.21 \times 10^4 (\rho_m/B_o)$
+1	0	$(-3.3 \times 10^9 - C + D) \rho_e$	$\pm 0.99 \times 10^4 (\rho_m/B_o)$
0	± 1	$(-3.3 \times 10^9 + C) \rho_e$	$\pm 0.77 \times 10^4 (\rho_m/B_o)$
± 1	± 2	$(-3.3 \times 10^9 + 3C + D) \rho_e$	$\pm 0.55 \times 10^4 (\rho_m/B_o)$
± 2	± 3	$(-3.3 \times 10^9 + 5C + 4D) \rho_e$	$\pm 0.33 \times 10^4 (\rho_m/B_o)$
± 3	± 4	$(-3.3 \times 10^9 + 7C + 9D) \rho_e$	$\pm 0.11 \times 10^4 (\rho_m/B_o)$

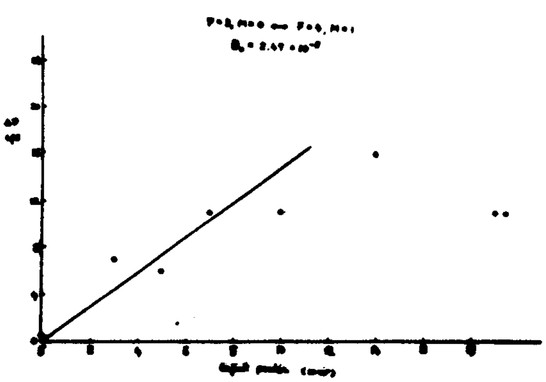
TABLE III.

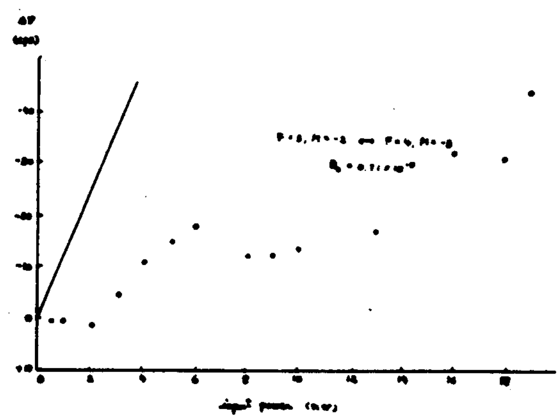
THEORETICAL RESULT WITH PARAMETERS GIVEN BY (54), (55), and (56)

M(F=3)	M(F=4)	B_o	frequency shift/ input over(cps/mw)
0	0	--	($<$1/50)
1	1	--	+1.9
0	1	2.5×10^{-5}	+ 1.5
1	0	2.5×10^{-5}	- 1.0
0	1	0.95×10^{-5}	+ 8.5
-2	-3	0.71×10^{-5}	- 14
3	4	0.73×10^{-5}	- 3.0




F=3, M=1 ↔ F=4, M=1
Δν
(cps)
input power (mw)




Δν
(cps)