MULTIQUANTUM TRANSITIONS BETWEEN EXCITED NONEQUIDISTANT ATOMIC SUBLEVELS

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Submitted to JETP editor, August 5, 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) 52, 267–275 (January, 1967)

The possibility of multiquantum transitions between excited atomic sublevels is studied. It is shown that in the case of nonequidistant sublevels, when single quantum transitions are excluded, a stationary component and also components whose intensity is modulated with a frequency two or four times that of the radio frequency field should appear in the fluorescence radiation. The effect is of a resonant nature and involves a resonant shift that is proportional to the square of the radio-frequency field amplitude.

A great number of theoretical and experimental researches have been devoted to the multiquantum transitions between Zeeman sublevels and the ground state. A detailed exposition of the theory of these transitions and the description of the basic experiment can be found in [1-4] and the review of Bonch-Bruevich and Khodovoĭ.[5]

Multiquantum transitions can take place not only in atoms in the ground state, but also when the atoms are in one of the sublevels of an excited state. The existence of such transitions can be detected by studying the spectra of the fluorescence light emitted by atoms of rarefied gases in transitions to the ground state. The observation of such transitions between excited states by means of fluorescence light is to a certain extent simpler than the observation of transitions between sublevels of the ground state.

In the study of multiquantum transitions in atoms in the ground state, the method of optical orientation of these atoms is employed. A polarized orienting longitudinal beam of light rays of resonant frequency is used here not only to change the difference in population of the magnetic sublevels, but also to detect resonance transitions between them. The greater the degree of orientation obtained, the greater the possibilities afforded by this method. In those cases in which orientation of the atom is impossible, this method of observation is completely unsuitable.

Resonance transitions to the excited state can be studied by observing the light reradiated by the atoms, which makes it possible to avoid the necessity of bringing about orientation of the atoms. This is due primarily to the fact that the light reradiated by each atom possesses information on the resonance phenomena in the excited state. Thus it would appear to be possible to increase significantly the range of objects of investigation, going beyond the limits of the very restricted number of atoms which can be optically oriented.

For simplicity, we shall consider below only two-quantum transitions. However, the method of calculation allows us in principle to consider transitions of higher order, too.

1. METHOD OF CALCULATION

We shall consider an atom the total electron spin moment **S** of which is equal to 0 in the ground state and 1 in the excited state. For the adjacent excited state, the total orbital momentum **L** will be taken equal to 1. In this case, the total angular momentum of the atom $\mathbf{J} = \mathbf{L} + \mathbf{S}$ takes on the values 0, 1, and 2 in the excited state and 0 in the ground state.

We consider optical transitions between these states of an atom which is placed in a constant magnetic field $\mathbf{H}_0$ and a radio-frequency field $\mathbf{H}_1(t)$. An example of such a transition is the transition $6^1S_0 \leftrightarrow 6^3P_{1,2,3}$ in atoms of the four isotopes of mercury. With account of the interaction between the momenta **L** and **S**, the Hamiltonian for the problem of interest to us can be written in the form

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1(t) + \mathcal{H}_{opt}, \tag{1}$$

where

$$\mathcal{H}_0 = \mathcal{H}_0' + a[1/2(L_+S_- + L_-S_+) + L_zS_z] + \omega_L L_z + \omega_s S_z \tag{2}$$

describes the interaction of the momenta with one another and with the constant magnetic field $\mathbf{H}_0$,

directed along the Oz axis. $\mathcal{H}_0'$ includes the non magnetic interactions in the atom and the radiation damping of the excited state. $\mathcal{H}_1(t)$ describes the interaction of the angular momenta with the radio frequency magnetic field $\mathbf{H}_1(t)$, which rotates in a plane perpendicular to $\mathbf{H}_0$ with frequency ω_0:

$$\mathcal{H}_1(t) = {}^1/_2(\Omega_L L_- + \Omega_S S_-)e^{i\omega_0 t} + {}^1/_2(\Omega_L L_+ + \Omega_S S_+) \times e^{-i\omega_0 t}. \quad (3)$$

In writing (2) and (3), we have used the notation

$$\begin{aligned} L_\pm &= L_x \pm iL_y, & S_\pm &= S_x + iS_y, \\ \omega_S &= -\gamma_S H_0, & \Omega_S &= -\gamma_S H_1, \\ \omega_L &= -\gamma_L H_0, & \Omega_L &= -\gamma_L H_1, \end{aligned} \quad (4)$$

where γ_S and γ_L are the gyromagnetic ratios of the spin and orbital angular momenta.

Optical excitation is described by the last term in Eq. (1):

$$\mathcal{H}_{opt}(t) = E(t)\mathbf{e}^0\mathbf{D}, \quad (5)$$

where $\mathbf{D}$ is the operator of the electric dipole moment, and $\mathbf{e}^0$ is the unit vector of polarization of the light wave. The system of units is used in which $\hbar = c = 1$.

The state $|t\rangle$ can be represented in the form of a superposition of time-independent states

$$|t\rangle = a_0(t)\,|m_{J'}\rangle + \sum_{m_J} a_{m_J}(t)\,|m_J\rangle, \quad (6)$$

where $m_{J'} = 0$ is the magnetic quantum number of the ground state and m_J that of the excited state. Its change with time obeys the equation

$$i\frac{d}{dt}|t\rangle = \mathcal{H}|t\rangle. \quad (7)$$

For the present, we shall not impose any limitations on the value of the intensity of the radio frequency $\mathbf{H}_1$. The interaction of the atom with the light field of the radiation we shall take into account with accuracy up to first order of perturbation theory. In this case, it is convenient to express the Hamiltonian (1) in the form of a sum of two terms

$$\mathcal{H} = \mathcal{H}' + \mathcal{H}_{opt}, \quad (8)$$

where $\mathcal{H}' = \mathcal{H}_0 + \mathcal{H}_1(t)$. If we denote by $|\,\rangle$ the state of the system which is obtained from the initial state $|t'\rangle$ only under the action of the operator $\mathcal{H}'$, then the equation of motion of this state will be

$$i\frac{d}{dt}|\,\rangle = \mathcal{H}'|\,\rangle. \quad (9)$$

The evolution of the system from the instant t' to the instant t under the action of $\mathcal{H}'$ can also be described by means of the operator $U(t, t')$

$$|\,\rangle = U(t, t')\,|t'\rangle. \quad (10)$$

Assuming that the atom is initially in a single sublevel of the ground state with $m_{J'} = 0$, we obtain a first-approximation solution of Eq. (6) in the form [6]

$$|t\rangle = U(t,0)\,|0\rangle + \frac{1}{i}\int_0^t dt'\,U(t,t')\,\mathcal{H}_{opt}(t')\,U(t',0)\,|0\rangle. \quad (11)$$

To find the operator $U(t, t')$, we transform to a new set of coordinates which rotate with frequency ω_0 around the direction of the field $\mathbf{H}_0$, by means of the unitary operator $T = \exp(i\omega J_z t)$

$$|\,\rangle' = T|\,\rangle. \quad (12)$$

Then Eq. (9) transforms into

$$i\frac{d}{dt}|\,\rangle' = \mathcal{H}_1'|\,\rangle', \quad (13)$$

where

$$\begin{aligned} \mathcal{H}_1' &= \mathcal{H}_0 + \Omega_L L_x + \Omega_S S_x - \omega_0 J_z = \mathcal{H}_0'' + \mathcal{H}_0', \\ \mathcal{H}_0'' &= \Omega_L L_x + \Omega_S S_x - \omega_0 J_z + (\omega_L L_z + \omega_S S_z) + a\mathbf{SL}. \end{aligned} \quad (14)$$

The solution of Eq. (13), in which $\mathcal{H}_1'$ no longer depends on the time, immediately leads to an expression for $U(t, t')$:

$$U(t, t') = e^{-i\omega_0 J_z t}e^{-i\mathcal{H}_1'(t-t')}e^{i\omega_0 J_z t}. \quad (15)$$

In order to obtain the final form of the solution of Eq. (7), it is still necessary to compute the coefficients $a_0(t)$ and $a_{mJ}(t)$, which enter into the relation (6). Taking (11) and (6) into account, we get for the coefficients a_{mJ}:

$$a_{m_J}(t) = \frac{1}{i}\int_0^t dt' \sum_n \langle m|U(t,t')|n\rangle\langle n|\mathcal{H}_{opt}(t')|0\rangle. \quad (16)$$

To obtain (16), we have assumed that at the initial instant of time $t = 0$ the atom is in the ground state described by the state vector $|0\rangle$.

2. CALCULATION OF THE MATRIX ELEMENTS $\langle m|U(t, t')|n\rangle$

In strong magnetic fields, where the coupling between the angular momenta $\mathbf{L}$ and $\mathbf{S}$ is broken, the calculation of the matrix elements $\langle m|U(t, t')|n\rangle$ presents no difficulty [6] By means of a transformation to a set of coordinates whose z-component is identical with the direction of the effective magnetic field in the rotating set of coordinates, the Hamiltonian H_1' is converted to diagonal form. In the case of arbitrary magnetic fields, it is not possible to reduce the Hamiltonian $\mathbf{H}_1'$ to diagonal form by a simple transformation in another representation; consequently, the calculation of the matrix elements of the operator $U(t, t')$ is made considerably more difficult. Inasmuch as the operator $J_z = L_z + S_z$ commutes with the time-independent Hamiltonian, operators of the form

$\exp(-i\omega_0 J_z t)$, which enter into Eq. (15), are diagonal.

The operator $\mathcal{H}_1'$, which is defined in the rotating set of coordinates, does not depend on the time. Therefore, we shall use for the determination of the operator $\exp[-i\mathcal{H}_1'(t-t')]$ in Eq. (15) a relation which is valid for any time-independent operator A[7,8],

$$e^{-iAt} = S\{e^{-iS^{-1}AS t}\}S^{-1} = R(t), \qquad (17)$$

where S is time-independent unitary matrix, and $S^{-1}AS$ is diagonal. Then the matrix $\exp\{iS^{-1}AS t\}$ is also diagonal and its matrix elements are easily found if the matrix S is known. In the case of a single angular momentum, this matrix is identical with the unit matrix transformation to a representation in which the Hamiltonian defined in the rotating system is diagonal. Then the operator $U(t, t')$ can be written down in the form

$$\begin{aligned} U(t,t') &= e^{-iJ_z\omega_0 t} S e^{-iS^{-1}\mathcal{H}_0'' S(t-t')} S^{-1} e^{iJ_z\omega_0 t} \\ &= e^{-iJ_z\omega_0 t} R(t-t') e^{iJ_z\omega_0 t} e^{i(k_0+i\Gamma/2)(t-t')}. \end{aligned} \qquad (18)$$

Thus the problem reduces to finding the matrix S and consequently to a solution of the corresponding secular equations which are necessary for diagonalization of the energy operator. Omitting the tedious intermediate calculations, we write down the following expression for the matrix of the operator $\mathcal{H}_1'$ in the representation (J, m_J):

$$|\mathcal{H}_0''| = \begin{vmatrix} E_1-2\omega_0 & A & B & 0 & 0 & 0 & 0 & 0 & 0 \\ A & E_2-\omega_0 & 0 & C_a & C_b & C_c & 0 & 0 & 0 \\ B & 0 & E_3-\omega_0 & D_a & D_b & D_c & 0 & 0 & 0 \\ 0 & C_a & D_a & E_4 & 0 & 0 & C_a' & D_a' & 0 \\ 0 & C_b & D_b & 0 & E_5 & 0 & C_b' & D_b' & 0 \\ 0 & C_c & D_c & 0 & 0 & E_6 & C_c' & D_c' & 0 \\ 0 & 0 & 0 & C_a' & C_b' & C_c' & E_7+\omega_0 & 0 & A' \\ 0 & 0 & 0 & D_a' & D_b' & D_c' & 0 & E_8+\omega_0 & B' \\ 0 & 0 & 0 & 0 & 0 & 0 & A' & B' & E_9+2\omega_0 \end{vmatrix}, \qquad (19)$$

where

$$A = \frac{\Omega_1}{\sqrt{2}}\cos\frac{\eta}{2} + \frac{\Omega_2}{\sqrt{2}}\sin\frac{\eta}{2}, \quad B = -\frac{\Omega_1}{\sqrt{2}}\sin\frac{\eta}{2} + \frac{\Omega_2}{\sqrt{2}}\cos\frac{\eta}{2},$$

$$C_a = \frac{1}{\sqrt{2}}(\Omega_1 a_4 + \Omega_2 a_6)\cos\frac{\eta}{2} + \frac{1}{\sqrt{2}}(\Omega_2 a_5 + \Omega_1 a_6)\sin\frac{\eta}{2},$$

$$D_a = -\frac{1}{\sqrt{2}}(\Omega_1 a_4 + \Omega_2 a_6)\sin\frac{\eta}{2} + \frac{1}{\sqrt{2}}(\Omega_2 a_5 + \Omega_1 a_6)\cos\frac{\eta}{2}. \qquad (20)$$

The matrix elements C_b and C_c are obtained from C_a by the substitutions $a_i \to b_i$ and $a_i \to c_i$, respectively. Similar results yield D_b and D_c from D_a. The primed quantities are obtained from the corresponding unprimed by means of the substitution $\Omega_1 \rightleftarrows \Omega_2$:

$$a_4 = \frac{a}{\omega_S - \omega_L - E_4 - a}a_6, \quad a_5 = \frac{a}{\omega_S - \omega_L + E_4 + a}a_6,$$

$$a_6 = \frac{(E_4+a)^2 - (\omega_S-\omega_L)^2}{\{[(\omega_S-\omega_L)^2 - (E_4+a)^2]^2 + 2a^2[(\omega_S-\omega_L)^2 + (E_4+a)^2]\}^{1/2}} \qquad (21)$$

The coefficients b_i are obtained from a_i by the substitution $E_4 \to E_5$ and c_i from a_i by the substitution $E_4 \to E_6$,

$$\cos\eta = \frac{\omega_S - \omega_L}{[(\omega_S-\omega_L)^2 + 4a^2]^{1/2}}. \qquad (22)$$

The rows and columns (19) are determined by choice of the quantum numbers J and m_J. The quantities $E_i(J, m_J)$ lying in the diagonal elements of the matrix (19) are the energy of the magnetic sublevels of the excited atom in the absence of a radio frequency field for arbitrary values of H_0. For J = 2 and m_J = 2, 1, 0, −1, −2, these energies are respectively equal to

$$\begin{aligned} E_1 &= a + \omega_L + \omega_S, \\ E_2 &= {}^1/_2(\omega_S + \omega_L) + {}^1/_2[(\omega_S-\omega_L)^2 + 4a^2]^{1/2}, \\ E_5 &= -{}^2/_3 a - 2r\cos(\varphi/3), \\ E_7 &= -{}^1/_2(\omega_S + \omega_L) + {}^1/_2[(\omega_S-\omega_L)^2 + 4a^2]^{1/2}, \\ E_9 &= a - \omega_L - \omega_S. \end{aligned} \qquad (23)$$

The quantum numbers J = 1 and m_J = 1, 0, −1 correspond to the energies

$$E_3 = {}^1/_2(\omega_S+\omega_L) - {}^1/_2[(\omega_S-\omega_L)^2+4a^2]^{1/2},$$
$$E_6 = -{}^2/_3 a + 2r\cos(\pi/3+\varphi/3),$$
$$E_8 = -{}^1/_2(\omega_S+\omega_L) - {}^1/_2[(\omega_S-\omega_L)^2+4a^2]^{1/2}. \quad (24)$$

Finally, the quantum number J = 0 corresponds to the single sublevel with energy

$$E_4 = -{}^2/_3 a + 2r\cos(\pi/3-\varphi/3). \quad (25)$$

Here we have the notation

$$r = {}^1/_3\{7a^2+3(\omega_S-\omega_L)^2\}^{1/2},$$
$$\cos\varphi = a\frac{|10a^2-9(\omega_S-\omega_L)^2|}{[7a^2+3(\omega_S-\omega_L)^2]^{3/2}} \quad (26)$$

3. TWO-QUANTUM TRANSITIONS BETWEEN ZEEMAN SUBLEVELS OF AN EXCITED STATE

We shall consider two-quantum transitions between the magnetic sublevel belonging to the same value J = 1. We shall separate from the matrix (19) a submatrix which contains the states 3, 6 and 8. We then have:

$$|\mathcal{H}_0''| = \begin{vmatrix} \delta+\Delta\omega+E_6 & D_c & 0 \\ D_c & E_6 & D_c' \\ 0 & D_c' & \delta-\Delta\omega+E_6 \end{vmatrix}, \quad (27)$$

where

$$D_c = -\frac{1}{\sqrt{2}}(\Omega_1 c_4+\Omega_2 c_6)\sin\frac{\eta}{2}+\frac{1}{\sqrt{2}}(\Omega_2 c_5+\Omega_1 c_6)\cos\frac{\eta}{2},$$
$$D_c' = -\frac{1}{\sqrt{2}}(\Omega_2 c_4+\Omega_1 c_6)\sin\frac{\eta}{2}+\frac{1}{\sqrt{2}}(\Omega_1 c_5+\Omega_2 c_6)\cos\frac{\eta}{2},$$
$$\delta = (E_3-E_6) - {}^1/_2(E_3-E_8),\quad \Delta\omega = {}^1/_2(E_3-E_8)-\omega_0, \quad (28)$$

δ is the frequency distribution of the single and double quantum resonances, Δω is the deviation of the radio frequency field from $(E_3 - E_8)/2$. The single resonance will take place for $\Delta\omega = \pm\delta$, the double one corresponds to the condition Δω = 0.

Inasmuch as it is rather complicated to carry out the analysis for arbitrary fields, we shall limit ourselves to the case of weak and intermediate values of the constant magnetic field. Then the expressions E_3, E_6, and E_8, and also D_C and D_C', reduce to the form

$$E_3 = -a + a\gamma\xi - \frac{a}{2}\xi^2,\quad D_c = -\frac{\Omega_+}{2\sqrt{2}}\left[1-\frac{\Omega_-}{2\Omega_+}\left(5\xi+\frac{\xi^2}{4}\right)\right],$$
$$E_6 = -a+2a\xi^2,\qquad D_c' = -\frac{\Omega_+}{2\sqrt{2}}\left[1+\frac{\Omega_-}{2\Omega_+}\left(5\xi+\frac{\xi^2}{4}\right)\right],$$
$$E_8 = -a-a\gamma\xi-\frac{a}{2}\xi^2, \quad (29)$$

where

$$\xi = \frac{|\gamma_S-\gamma_L|}{2a}H_0,\quad \gamma = \frac{\gamma_S+\gamma_L}{\gamma_S-\gamma_L},\quad \Omega_\pm = |\gamma_S\pm\gamma_L|H_1.$$

If we write down the eigenvalues of $\mathcal{H}_0''$ in the form $E_6+\sigma_i$, then the secular equation for a determination of these eigenvalues reduces to a cubic equation in σ_i. Near the double resonance, the solution of this equation is greatly simplified, inasmuch as in this case $\Delta\omega/\delta \sim (\Omega_+/\delta)^2 \ll 1$. Finally, the approximate values of σ_i that have been obtained will be valid only under the limitations put on the value of the radio frequency field by the inequality given above. We then obtain

$$\sigma_3 = \delta\left\{1+\frac{1}{8}z^2(p+qy)-\frac{1}{32}p^2z^4+M^{1/2}\right\},$$
$$\sigma_6 = -\frac{\delta}{4}z^2\left(p+qy-\frac{1}{4}p^2z^2\right),$$
$$\sigma_8 = \delta\left\{1+\frac{1}{8}z^2(p+qy)-\frac{1}{32}p^2z^4-M^{1/2}\right\}, \quad (30)$$

where the following notation has been used:

$$z = \frac{\Omega_+}{\delta},\quad y = \frac{\Delta\omega}{\delta},\quad p = 1+\frac{25}{4\gamma^2}\xi^2,\quad q = \frac{1}{\gamma}\left(5\xi+\frac{\xi^2}{4}\right),$$
$$M = y^2-\frac{q}{4}z^2y+\frac{p^2}{64}z^4.$$

If the eigenfunctions belonging to the eigenvalues $E_6+\sigma_i$, are written

$$\Psi_i = \begin{pmatrix}\alpha_i\\ \beta_i\\ \gamma_i\end{pmatrix}, \quad (31)$$

where i = 3, 6, 8, then the solution of the equation gives an expression for α_i, β_i, γ_i:

$$\alpha_i = \left[1+\frac{(\delta+\Delta\omega-\sigma_i)^2}{D_c^2}+\frac{D_c'}{D_c}\left(\frac{\delta+\Delta\omega-\sigma_i}{\delta-\Delta\omega-\sigma_i}\right)^2\right]^{-1/2},$$
$$\beta_i = -\frac{\delta+\Delta\omega-\sigma_i}{D_c}\alpha_i,\quad \gamma_i = \frac{D_c'}{D_c}\frac{\delta+\Delta\omega-\sigma_i}{\delta-\Delta\omega-\sigma_i}\alpha_i. \quad (32)$$

Close to the double resonance, the second term in the expression for α_i can be neglected in comparison with the rest of the expression. In this case, the coefficients take on a very simple form:

$$\alpha_3 = \cos\frac{\Phi}{2},\quad \alpha_6 = 0,\quad \alpha_8 = \sin\frac{\Phi}{2},$$
$$\beta_3 = 0,\quad \beta_6 = 1,\quad \beta_8 = 0,$$
$$\gamma_3 = \sin\frac{\Phi}{2},\quad \gamma_6 = 0,\quad \gamma_8 = \cos\frac{\Phi}{2}, \quad (33)$$

where

$$\cos^2\Phi = \frac{(\Delta\omega-\omega^*)^2}{(\Delta\omega-\omega^*)^2+\Omega^2},$$
$$\Omega^2 = \frac{\Omega_+^4}{64\delta^2}\left(1-\frac{25}{2\gamma^2}\xi^2\right),\quad \omega^* = \frac{\Omega_+^2}{8\gamma\delta}\left(5\xi+\frac{\xi^2}{4}\right). \quad (34)$$

Thus the coefficients α_i, β_i and γ_i are known; then the matrix S is obtained for the group of levels of interest to us:

$$S=\begin{pmatrix}\alpha_3 & \alpha_6 & \alpha_8\\ \beta_3 & \beta_6 & \beta_8\\ \gamma_3 & \gamma_6 & \gamma_8\end{pmatrix}. \tag{35}$$

Substituting (35) in (18), we obtain the matrix elements $\langle m|U(t,t')|n\rangle$, which are necessary for finding the coefficients $a_{m_J}(t)$, and consequently, the state vectors $|t\rangle$.

4. INTENSITY OF THE FLUORESCENCE LIGHT

The component of the field of the radiation field in the direction of the unit vector **e**, is, as is well known,[9] equal to

$$\mathbf{eE}=\frac{2k^2}{r}\langle 0|\mathbf{eD}|t\rangle, \tag{36}$$

where $|t\rangle$ is the state of the atom at the time t:

$$|t\rangle=a_0|0\rangle+\frac{1}{i}\int_0^t dt'\sum_{m,n}\exp\{-i[\omega_0(mt-nt')-(k_0+i\Gamma/2)(t-t')]\}\langle m|R(t-t')|n\rangle\langle n|\mathbf{e}^0\mathbf{D}|0\rangle|m\rangle. \tag{37}$$

Then the intensity of the fluorescence light, which is determined by the expression $(\mathbf{E}^*\cdot\mathbf{E})/8\pi$, is, if we take into account (36) and (37), equal to

$$I=\frac{k^4}{2\pi r^2}\int_0^t dt'\int_0^t dt''E(t')E(t'')\sum_{m,m',n,n'}G_{mm'}R_{mn}(t-t')\times F_{nn'}R^*_{m'n}(t-t'')\times\exp\{-i[\omega_0(mt-nt'-m't+n't'')-(k_0+i\Gamma/2)\times(t-t')]\}, \tag{38}$$

where $G_{mm'}=\langle 0|\mathbf{e}\cdot\mathbf{D}|m\rangle\langle m'|\mathbf{e}\cdot\mathbf{D}|0\rangle$ is the radiation matrix, and $F_{nn'}=\langle n|\mathbf{e}^0\cdot\mathbf{D}|0\times 0|\mathbf{e}\cdot\mathbf{D}|n'\rangle$ is the excitation matrix. The matrix elements of these matrices can be found in the paper of Dodd and Series.[10]

We shall assume that the exciting light is propagated along the Ox axis and polarized along the Oy axis. In this case the atom can be excited at a Zeeman sublevel characterized by the magnetic quantum number $m_J=\pm 1$. The expression for the intensity of the reradiated light, linearly polarized in the direction **e** and propagating at angles θ and φ at some instant of time t, will consist of three groups of components. These comprise terms which are independent of the time:

$$I_1=I_0\left\{\tfrac{1}{4}(1+\cos^2\theta)\left(1+\frac{\Gamma^2\sin^2\Phi}{\Gamma^2+4\delta^2M}\right)+\Gamma\sin^2\theta\cdot\times\left[\frac{\Gamma\cos 2\varphi+2(\omega_0+\delta M^{1/2})\sin 2\varphi}{\Gamma^2+4(\omega_0+\delta M^{1/2})^2}\times\cos^4\frac{\Phi}{2}+\frac{\Gamma\cos 2\varphi+2(\omega_0-\delta M^{1/2})\sin 2\varphi}{\Gamma^2+4(\omega_0-\delta M^{1/2})^2}\times\sin^4\frac{\Phi}{2}+\frac{\Gamma\cos 2\varphi+2\omega_0\sin 2\varphi}{2(\Gamma^2+4\omega_0^2)}\sin^2\Phi\right]\right\}, \tag{39}$$

terms which describe the modulation of the intensity with frequency $2\omega_0$:

$$I_2=I_0\sin\Phi\left\{\Gamma(1+\cos^2\theta)\left[\frac{\Gamma\cos 2\omega_0t+2\omega_0\sin 2\omega_0t}{\Gamma^2+4\omega_0^2}+\frac{\Gamma\cos 2\omega_0t+2(\omega_0+\delta M^{1/2})\sin 2\omega_0t}{\Gamma^2+4(\omega_0+\delta M^{1/2})^2}\cos^2\frac{\Phi}{2}+\frac{\Gamma\cos 2\omega_0t+2(\omega_0-\delta M^{1/2})\sin 2\omega_0t}{\Gamma^2+4(\omega_0-\delta M^{1/2})^2}\sin^2\frac{\Phi}{2}\right]+\left[\cos(2\omega_0t-2\varphi)+\Gamma\frac{\Gamma\cos 2(\omega_0t-\varphi)-2\delta M^{1/2}\sin 2(\omega_0t-\varphi)\cos\Phi}{\Gamma^2+4\delta^2M}\right]\times\sin^2\theta\right\}, \tag{40}$$

and terms which describe the modulation of the intensity with frequency $4\omega_0$:

$$I_3=I_0\frac{\Gamma\sin^2\theta\sin^2\Phi}{4}\times\left\{2\frac{\Gamma\cos 2(2\omega_0t-\varphi)+2\omega_0\sin^2 2(2\omega_0t-\varphi)}{\Gamma^2+4\omega_0^2}+\frac{\Gamma\cos 2(2\omega_0t-\varphi)+2(\omega_0-\delta M^{1/2})\sin 2(2\omega_0t-\varphi)}{\Gamma^2+4(\omega_0-\delta M^{1/2})^2}+\frac{\Gamma\cos 2(2\omega_0t-\varphi)+2(\omega_0+\delta M^{1/2})\sin 2(2\omega_0t-\varphi)}{\Gamma^2+4(\omega_0+\delta M^{1/2})^2}\right\}. \tag{41}$$

In obtaining these expressions, it has been taken into account that the correlation of the incident electric field at the various times can be represented by the expression

$$\langle E(t')E(t'')\rangle=8\pi\int_{-\infty}^{+\infty}dk\rho(k)\exp\{-ik(t'-t'')\}.$$

As to the width of the frequency spectrum of the incident radiation, it is much greater than the natural line width and therefore

$$\rho(k)\approx\rho(k_0).$$

The quantities $\sin\Phi$ and $\cos\Phi$ enter into the right hand side of Eqs. (39), (40) and (41) as factors which, in accord with (34), have a resonance denominator $(\Delta\omega-\omega^*)^2+\Omega^2$, while ω^* is determined by Eq. (34) and is the amount by which the peak of the resonance curve is shifted from its position at the point of double resonance $\Delta\omega=0$. This shift depends not only on the constant field $\mathbf{H}_0$, but on the square of the amplitude of the radio

frequency field H_1. Evidently, this shift can be determined from the value of the measured resonance frequency of the double transition.

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Translated by R. T. Beyer
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