Interaction between a spin-1/2 atom and a strong rf field

T. Yabuzaki, S. Nakayama, Y. Murakami, and T. Ogawa
Ionosphere Research Laboratory, Kyoto University, Kyoto, Japan
(Received 21 May 1974)

Energies of the coupled system of a spin-1/2 atom in a static magnetic field and an oscillating rf field are analyzed with the resolvent formalism and numerically calculated by quantizing the rf field, for the general case that the strong rf field is oriented in an arbitrary direction to the static magnetic field. It is shown that, if the component of the static field along the direction of the rf field has particular values, level crossings take place and multiphoton transitions by even or odd numbers of rf photons become forbidden, even when the polarization of the rf field satisfies the parity condition for the multiphoton transitions. Particular attention is given to the direction and amount of the shifts of the level crossings by the rf field, together with the shifts of the anticrossings or of the multiphoton transitions. Theoretical results are verified by the optical-pumping experiments with cesium vapor.

I. INTRODUCTION

Interaction between a spin-$\frac{1}{2}$ atom in a static magnetic field and a strongly oscillating radio-frequency (rf) field has been analyzed by many authors for various cases. To study such interaction experimentally, optical pumping is a very useful tool, since it gives us information on both real transitions and coherence between Zeeman substates of the atom.

Consider a spin-$\frac{1}{2}$ atom with a gyromagnetic ratio γ in a static magnetic field $H_0=\omega_0/\gamma$ and an oscillating rf field $H_1 \cos\omega t=(\omega_1/\gamma)\cos\omega t$. In most of the work done so far, two particular cases in which the direction of the rf field is perpendicular or parallel to the static field H_0 have been studied extensively.

When the rf field is oriented perpendicularly to the static field H_0, an ordinary (single-photon) transition and multiphoton transitions take place at $\omega_0=(2n+1)\omega$, where n is zero or an integer. The first observation of multiphoton transitions was made by Margerie and Brossel[1] for longitudinal optical pumping (with respect to the static field H_0) and explained theoretically by Winter.[2] Good physical descriptions have been given with a semiclassical time-dependent Hamiltonian by Shirley,[3] and with a time-independent Hamiltonian in which the rf field is quantized by Cohen-Tannoudji and Haroche.[4-6] Furthermore, Stenholm[7] obtained semiclassically a complete solution by making use of the continued-fraction method. The real transitions, such as ordinary and multiphoton transitions, appear at the anticrossings of the energy diagram of the coupled atom–rf-field system, or of the atom "dressed" by rf photons.[6] In the case of transverse optical pumping, the coherence between the crossing energy levels at $\omega_0=2n\omega$ can be observed as a resonance, known as a "Haroche resonance."[6] It has been shown by Cohen-Tannoudji and Haroche that the Haroche resonances shift toward $\omega_0=0$ as the amplitude H_1 of the rf field is increased, but they are not broadened to the lowest order in the rf-field amplitudes. However, the broadening of the Haroche resonances as a higher-order effect has recently been calculated by Tsukada and Ogawa[8] and by Stenholm and Aminoff[9] by making use of the continued-fraction method. Another important effect of the rf field oriented perpendicularly to the field H_0 is that the atomic g factor is effectively modified when the condition $\omega>\omega_0$ is satisfied,[4-6] and this effect has been observed experimentally.[10-13]

On the other hand, when the rf field is oriented along the direction of the static field H_0, a parametric resonance occurs when $\omega_0=n\omega$, at which the level crossing takes place in the energy diagram of the coupled system. For this case, the energies and eigenfunctions have been obtained completely by semiclassical[14] and quantum-mechanical[15] treatments. When a weak static field $H_\perp=\omega_\perp/\gamma$ is applied perpendicular to the rf field in addition to the static field $H_\parallel=\omega_\parallel/\gamma$ along the direction of the rf field, and the condition $\omega_\parallel=n\omega$ is satisfied, it has been shown by Pegg and Series,[16] and Yabuzaki *et al.*[17] that the medium irradiated by the strong rf field becomes anisotropic, and the component of the g factor perpendicular to the rf field is modified, which can be expressed simply by Bessel functions of the first kind.

As described above, there has been no theoretical treatment of the general case in which the direction of the strongly oscillating rf field is arbitrary with respect to that of the static field H_0. Recently a new type of resonance has been observed when both components $H_\parallel$ and $H_\perp$ are not weak.[18] Since the behavior of this resonance is quite similar to that of the Haroche resonance, it is called a "Haroche-like" resonance. But the physical importance involved in this resonance has not yet been reported.

In this paper, we report on the energies of the coupled atom-rf-field system in the presence of both components $H_{\parallel}$ and $H_{\perp}$ for a wide range of the rf-field amplitudes. In this case, it is believed that there is no level crossing for an atom with the angular momentum $J=\frac{1}{2}$, hence the multiphoton transition at $\omega_0=n\omega$ is allowed for any integer n.[19] Using the resolvent formalism in which the rf field is quantized, we first show that a level crossing exists at $\omega_0=(n+2n')\omega$ (where n and n' are integers), if the condition $\omega_{\parallel}=n\omega$ is satisfied. This kind of level crossing might be called a "crossing of the second kind,"[19] because at the level crossing each elementary process of the multiphoton transition associated with $n+2n'$ photons is allowed although the sum of the elementary processes becomes forbidden. We show that the Haroche-like resonance observed by Tsukada *et al.*[18] is due to the interference between the crossing energy levels. Next, numerical calculation of energies of the coupled system is carried out by diagonalizing the 40×40 matrix of the total Hamiltonian in order to find higher-order effects. This corresponds to tenth-order perturbation theory with respect to the rf-field amplitude. We give particular attention to the number of shifts of the level crossings by the rf field, together with their direction in $(\omega_{\parallel}, \omega_{\perp})$ space.

The existence of the level crossing of the second kind is verified by an optical-pumping experiment with cesium vapor. The theoretical shifts of the level crossing and anticrossing are compared with the results of the present experiment and with the experiment by Tsukada *et al.*[18]

II. PERTURBATION THEORY WITH THE RESOLVENT FORMALISM

Here we show the existence of the level crossing of the second kind in the energy diagram of the coupled atom-rf-field system, even when the static field H_0 has both components $H_{\parallel}$ and $H_{\perp}$ parallel and perpendicular to the direction of the rf field. The analysis is made with the resolvent formalism developed by Cohen-Tannoudji and Haroche,[4-6] in which the rf field is quantized and perturbation theory is used.

An atom with spin-$\frac{1}{2}$ is subjected to the simultaneous actions of the static field H_0 and an oscillating rf field $H_1\cos\omega t$, as shown in Fig. 1. The direction of the rf field makes an angle θ from the direction of H_0; hence its polarization $\vec{e}$ can be written as

$$\vec{e}=\vec{e}_x\cos\theta+\vec{e}_z\sin\theta, \tag{1}$$

where $\vec{e}_x$ and $\vec{e}_z$ are unit vectors representing the directions of x and z axes. In this system the total Hamiltonian can be written as

$$\mathcal{H}=\mathcal{H}_0+V, \tag{2}$$

with

$$\mathcal{H}_0=\omega_0 J_z+\omega a^\dagger a, \tag{3}$$

$$V=\lambda\vec{J}(a\vec{e}+a^\dagger\vec{e}^*), \tag{4}$$

where $\vec{J}$ is the total angular momentum of an atom, J_z its z component, $a^\dagger$ and a the creation and annihilation operators of the rf photon ω, and λ the coupling coefficient. For a large number of rf photons, the coupling coefficient λ is given by

$$\lambda=\omega_1/2\bar{N}^{1/2}, \tag{5}$$

where $\bar{N}$ is the average number of rf photons. In Eq. (3), $\omega_0 J_z$ represents the Hamiltonian of an atom in a static field H_0, having eigenstates $|+\rangle$ and $|-\rangle$ and eigenvalues $\pm\frac{1}{2}\omega_0$, and $\omega a^\dagger a$ represents the Hamiltonian of the rf field only, having eigenstates $|n\rangle$ where n is the number of photons. Thus we have the eigenstates and eigenvalues of $\mathcal{H}_0$ as follows:

$$\mathcal{H}_0|\pm,n\rangle=(n\omega\pm\tfrac{1}{2}\omega_0)|\pm,n\rangle, \tag{6}$$

where $|\pm,n\rangle$ is the product of eigenstates $|n\rangle$ and $|\pm\rangle$. Consequently, in the absence of the rf field, the eigenstates $|+,n\rangle$ and $|-,n'\rangle$ degenerate at $\omega_0=(n-n')\omega$; i.e., the level crossing exists at $\omega_0=p\omega$, where p is zero or an integer.

The term V in Eq. (2) represents the interaction between the atom and the rf photons. It can be written, in the present case, as

$$V=V_\pi+V_{\sigma_+}+V_{\sigma_-}, \tag{7}$$

with

$$V_\pi=\lambda\cos\theta J_z(a+a^\dagger), \tag{8}$$

$$V_{\sigma_+}=\tfrac{1}{2}\lambda\sin\theta(J_+a+J_-a^\dagger), \tag{9}$$

$$V_{\sigma_-}=\tfrac{1}{2}\lambda\sin\theta(J_+a^\dagger+J_-a), \tag{10}$$

where

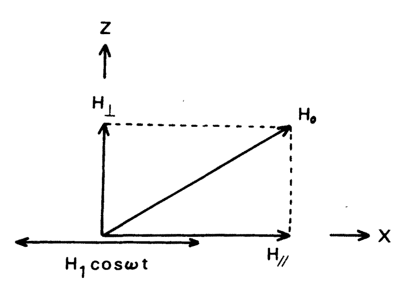

FIG. 1. Directions of magnetic fields.

$$J_\pm = J_x \pm i J_y . \tag{11}$$

In Eqs. (8)–(10), V_π, $V_{\sigma+}$, and $V_{\sigma-}$ represent the interactions of the atom with the rf photons in π, σ_+, and σ_- modes, respectively. Using Eq. (5), we can easily calculate the matrix elements of V_π, $V_{\sigma+}$, and $V_{\sigma-}$ between the eigenstates $|\pm, n\rangle$ and $|\pm, n-1\rangle$ as

$$\langle \pm, n | V_\pi | \pm, n-1\rangle = \langle \pm, n-1 | V_\pi | \pm, n\rangle = \pm\tfrac{1}{4}\omega_1 \cos\theta , \tag{12}$$

$$\langle +, n-1 | V_{\sigma+} | -, n\rangle = \langle -, n | V_{\sigma+} | +, n-1\rangle = \tfrac{1}{4}\omega_1 \sin\theta , \tag{13}$$

$$\langle +, n | V_{\sigma-} | -, n-1\rangle = \langle -, n-1 | V_{\sigma-} | +, n\rangle = \tfrac{1}{4}\omega_1 \sin\theta ; \tag{14}$$

the other matrix elements are zero.

It might be important to recall the work which has already been done for two particular cases. Specifically, we are interested in whether or not the crossing at $\omega_0 = p\omega$ in the absence of the rf field becomes an anticrossing. When $\theta = \frac{1}{2}\pi$, i.e., the rf field is oriented perpendicularly to the field H_0, it is known that the crossing at $\omega_0 = p\omega$ for p an odd integer becomes an anticrossing by virtue of the real transitions: the ordinary transition for $p=1$ and multiphoton transition for $p>1$. The energy separation between the anticrossing energy levels is closely connected to the probability for the real transition between them. The crossing at $\omega_0 = p\omega$ for p an even integer, does not become an anticrossing because the parity condition for the real transition is not satisfied at this crossing. The Haroche resonance which can be observed in the transverse-optical-pumping experiment is a coherent phenomenon between the crossing energy levels.[4-6] However, when $\theta = 0$, the crossing at $\omega_0 = p\omega$ does not become an anticrossing for any integer p, and the coherence phenomenon between the crossing energy levels can be observed as a parametric resonance in the transverse-optical-pumping experiment. In this case, if the condition $\omega_\parallel = p\omega$ is satisfied, it is important to notice that the component of the atomic g factor perpendicular to the direction of the rf field is effectively modified to the form[16,17]

$$g_\perp = g_0 J_p(\omega_1/\omega) , \tag{15}$$

where g_0 is the g factor in the absence of the rf field and $J_p(\omega_1/\omega)$ is the pth order Bessel function of the first kind.

Generally, the level crossing can be considered a special case of the level anticrossing, at which the energy separations between associated energy levels are zero, so that in any analysis it is sufficient to consider the anticrossing only. In order to discover the shifts in position of the level anticrossing and the energy separation between anticrossing levels, it is convenient to calculate the matrix elements of the level-shift operator $\overline{R}$ in the resolvent formalism developed by Cohen-Tannoudji and Haroche.[4-6] Consider two particular eigenstates $|a\rangle$ and $|b\rangle$ of the unperturbed Hamiltonian $\mathcal{H}_0$, with corresponding eigenvalues E_a and E_b. The matrix $\overline{R}$ is given by

$$\overline{R} = V + PV \frac{Q}{E_c - \mathcal{H}_0} VP + PV \frac{Q}{E_c - \mathcal{H}_0} V \frac{Q}{E_c - \mathcal{H}_0} PV + \ldots , \tag{16}$$

where

$$E_c = \tfrac{1}{2}(E_a + E_b) , \tag{17}$$

and P and Q are the projection operators defined by

$$P = |a\rangle\langle a| + |b\rangle\langle b| , \quad Q = 1 - P . \tag{18}$$

The shift of the anticrossing point by the rf field is associated with the diagonal elements of $\overline{R}$, and its magnitude $\Delta\omega$ is given by

$$\Delta\omega = \overline{R}_{aa} - \overline{R}_{bb} . \tag{19}$$

The energy separation between anticrossing levels is given by the off-diagonal element, i.e., $2|\overline{R}_{ab}|$ (see Fig. 2). In terms of these matrix elements, the transition probability P_{ab} between the states $|a\rangle$ and $|b\rangle$ is given by

$$P_{ab} = \frac{2|\overline{R}_{ab}|^2}{\Gamma^2 + 4|\overline{R}_{ab}|^2 + (E_b - E_a - \overline{R}_{aa} + \overline{R}_{bb})^2} , \tag{20}$$

where Γ is the natural width of the states $|a\rangle$ and $|b\rangle$. Thus, if $\overline{R}_{ab}$ has a nonzero value at $E_b - E_a = \Delta\omega$, we have a level anticrossing; hence the multiphoton transition between the states $|a\rangle$ and $|b\rangle$ is allowed.

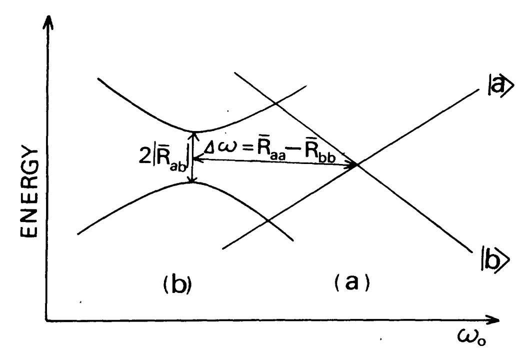

FIG. 2. Energy diagram showing the relation of elements of the matrix $\overline{R}$ with the shift and energy separation of the level anticrossing; the energy levels are modified by the rf field from (a) in the case $\omega_1 = 0$ to (b).

We have calculated $\bar{R}_{ab}$ and $\Delta\omega$ to the lowest order of the rf-field amplitude ω_1 at or near $\omega_0 = p$, where the states $|-,n\rangle$ and $|+,n-p\rangle$ are degenerate in the absence of the rf field.

a. The case $\omega_0 = \omega$. In this case the states $|-,n\rangle$ and $|+,n-1\rangle$ are coupled only by the rf photon in the σ_+ mode, as shown in the Feynman diagram of Fig. 3(a). With Eqs. (12)-(14) and (16), $\bar{R}_{ab}$ and $\Delta\omega$ to the lowest order of ω_1 can be calculated as

$$\bar{R}_{ab} = \tfrac{1}{4}\omega_1 \sin\theta , \tag{21}$$

$$\Delta\omega = (\omega_1^2/16\omega)\sin^2\theta . \tag{22}$$

From Eq. (21) we see that the level crossing occurs at $\theta = 0$, which gives rise to the parametric resonance. When $\theta = \frac{1}{2}\pi$, Eq. (22) gives the Bloch-Siegert effect by the counter-rotating rf field, which might be encountered in the ordinary magnetic resonance experiment.

b. The case $\omega_0 = 2\omega$. In this case, the transition between the states $|-,n\rangle$ and $|+,n-2\rangle$ has two elementary processes as shown in Fig. 3(b). The off-diagonal elements of $\bar{R}$ for these two elementary processes, i.e., $\bar{R}_{ab}^{(1)}$ and $\bar{R}_{ab}^{(2)}$, have the same value, and the sum becomes

$$\bar{R}_{ab} = -(\omega_1^2/8\omega)\sin\theta\cos\theta , \tag{23}$$

and the shift $\Delta\omega$ becomes

$$\Delta\omega = (\omega_1^2/6\omega)\sin^2\theta . \tag{24}$$

The level crossing at $\theta = 0$ and $\theta = \frac{1}{2}\pi$ correspond to the parametric resonance and Haroche resonance, respectively. This level crossing should be called a "crossing of the first kind",[19] since all of the elementary processes with a two-photon transition become forbidden.

c. The case $\omega_0 = 3\omega$. In this case, the transition between states $|-,n\rangle$ and $|+,n-3\rangle$ has four elementary processes as shown in Fig. 3(c). The matrix elements $\bar{R}_{ab}$ for these elementary processes are

$$\bar{R}_{ab}^{(1)} = (1/2\omega^2)(\tfrac{1}{4}\omega_1)^3 \sin\theta\cos^2\theta$$

$$= \tfrac{1}{2}\bar{R}_{ab}^{(2)} = \bar{R}_{ab}^{(3)} , \tag{25}$$

$$\bar{R}_{ab}^{(4)} = -(1/4\omega^2)(\tfrac{1}{4}\omega_1)^3 \sin^3\theta . \tag{26}$$

The sum of $\bar{R}_{ab}$ for all elementary processes becomes

$$\bar{R}_{ab} = \sum_{i=1}^{4} \bar{R}_{ab}^{(i)} = (1/4\omega^2)(\tfrac{1}{4}\omega_1)^3 \sin\theta\,(9\cos^2\theta - 1) . \tag{27}$$

At $\theta = 0$, each elementary process is forbidden as seen in Eqs. (26) and (17), and we have again the level crossing of the first kind. At $\cos\theta = \frac{1}{3}$ or $\omega_\parallel = \omega$, we have another level crossing; i.e., the three-photon transition between $|-,n\rangle$ and $|+,n-3\rangle$ becomes forbidden. In this case, it is important to notice that all four elementary processes are allowed; hence such a level crossing should be called a "crossing of the second kind." It is believed that crossings of the second kind do not exist for spin-$\frac{1}{2}$ systems.[19] The crossing at $\cos\theta = \frac{1}{3}$ corresponds to the Haroche-like resonance observed by Tsukada *et al.*,[18] and its shift $\Delta\omega$ can be calculated as

$$\Delta\omega = (3\omega_1^2/32\omega)\sin^2\theta . \tag{28}$$

d. The case $\omega_0 = 4\omega$. In this case the transition between $|-,n\rangle$ and $|+,n-4\rangle$ has eight elementary processes, and the sum of $\bar{R}_{ab}$ for these processes and the shift $\Delta\omega$ becomes

$$\bar{R}_{ab} = -(\omega_1^4/576\omega^2)\sin\theta\cos\theta(4\cos^2\theta - 1) , \tag{29}$$

$$\Delta\omega = (\omega_1^2/15\omega)\sin^2\theta . \tag{30}$$

At $\sin\theta = \frac{1}{2}$ or $\omega_\parallel = 2\omega$, we have a level crossing of the second kind, at which all eight elementary processes of the four-photon transition are allowed.

From the above analysis, we can expect generalized expressions for $\bar{R}_{ab}$ and $\Delta\omega$ to the lowest order in ω_1, for the case where the n-photon process is important.

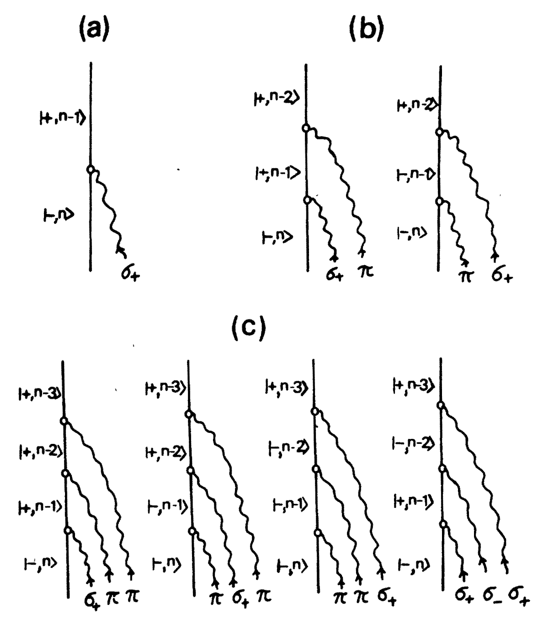

FIG. 3. Feynman diagrams for (a) the single-photon transition, (b) two elementary processes of the two-photon transition, and (c) four elementary processes of the three-photon transition.

For $\omega_0 = 2k\omega$ $(k \geq 2)$, we expect

$$\overline{R}_{ab} = -C_k(\omega_1^{2k}/\omega^{2k-1})\cos\theta\sin\theta(4k^2\cos^2\theta - 2^2) \times(4k^2\cos^2\theta - 4^2)\cdots[4k^2\cos^2\theta - (2k-2)^2], \tag{31}$$

$$\Delta\omega = \frac{k\omega_1^2}{2(4k^2-1)\omega}\sin^2\theta, \tag{32}$$

where C_k is a constant depending on the integral number k. Equation (32) holds for $k=1$. The conditions under which $\overline{R}_{ab}$ becomes zero, i.e., the level crossing occurs, are

$$\cos\theta = 0, \quad \sin\theta = 0 \quad \text{(crossing of the first kind)}, \tag{33}$$

$$\cos\theta = 1/k,\ 2/k, \ldots, (k-1)/k \quad \text{(crossing of the second kind)}, \tag{34}$$

or

$$\omega_{\parallel}/\omega = 2, 4, \ldots, 2k-2\,. \tag{35}$$

For $\omega_0 = (2k+1)\omega$ $(k \geq 1)$, we expect

$$\overline{R}_{ab} = C_k'(\omega_1^{2k+1}/\omega^{2k})\sin\theta[(2k+1)^2\cos^2\theta - 1] \times\{(2k+1)^2\cos^2\theta - 3\}\cdots[(2k+1)^2\cos^2\theta - (2k-1)^2], \tag{36}$$

$$\Delta\omega = \frac{(2k+1)\omega_1^2}{16k(k+1)\omega}\sin^2\theta, \tag{37}$$

where C_k' is a constant. The conditions under which $\overline{R}_{ab}$ becomes zero are

$$\cos\theta = 0 \quad \text{(crossing of the first kind)}, \tag{38}$$

$$\cos\theta = \frac{1}{2k+1}, \frac{3}{2k+1}, \ldots, \frac{2k-1}{2k+1} \quad \text{(crossing of the second kind)}, \tag{39}$$

or

$$\omega_{\parallel}/\omega = 1, 3, \ldots, 2k-1\,. \tag{40}$$

Equations (36) and (37) do not hold for the case $\omega_0 = \omega$ and Eq. (32) does not hold for the case $\omega_0 = 2\omega$. Instead, we have to use Eqs. (21)–(23). The reason why these general expressions do not hold for the cases $\omega_0 = \omega$ and $\omega_0 = 2\omega$ might be because all modes of rf photons do not contribute simultaneously to $\overline{R}_{ab}$ or $\Delta\omega$ in these cases.

It is important to note that the condition necessary for a level crossing of the second kind to occur is that $\omega_{\parallel}/\omega$ has an integral value, as seen in Eqs. (35) and (40). Furthermore, for the n-photon process, $\overline{R}_{ab}$ is proportional to ω_1^n as seen in Eqs. (31) and (36), but the shift $\Delta\omega$ of the anticrossing or crossing by the rf field is proportional to ω_1^2 from Eqs. (32) and (37), to the lowest order of ω_1.

III. NUMERICAL CALCULATION OF ENERGIES OF THE COUPLED SYSTEM

In the preceding section we have shown with perturbation theory that the level crossing of the second kind exists in the coupled atom–rf-field system. In order to understand the behavior of the crossing of the second kind in a strong rf field, for which the lowest-order perturbation theory is no longer valid, we have numerically calculated the energies of the coupled system in such a strong rf field. Particular attention is given to the amount of the shifts of the crossing points by the rf field and their directions in $(\omega_{\parallel}, \omega_{\perp})$ space.

In terms of the eigenstates $|\pm, n\rangle$ of $\mathcal{H}_0$, Eq. (2) can be written in matrix form as

$$\mathcal{H} - n\omega I = \begin{array}{c} \\ \cdot \\ |+,n-1\rangle \\ |-,n-1\rangle \\ |+,n\rangle \\ |-,n\rangle \\ |+,n+1\rangle \\ |-,n+1\rangle \\ \cdot \end{array} \begin{array}{c} \begin{array}{cccccccc} \cdot & |+,n-1\rangle & |-,n-1\rangle & |+,n\rangle & |-,n\rangle & |+,n+1\rangle & |-,n+1\rangle & \cdot \end{array} \\ \left[\begin{array}{cccccccc} \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \frac{1}{2}\omega_0 - \omega & 0 & B & A & 0 & 0 & \cdot \\ \cdot & 0 & -\frac{1}{2}\omega_0 - \omega & A & -B & 0 & 0 & \cdot \\ \cdot & B & A & \frac{1}{2}\omega_0 & 0 & B & A & \cdot \\ \cdot & A & -B & 0 & -\frac{1}{2}\omega_0 & A & -B & \cdot \\ \cdot & 0 & 0 & B & A & \frac{1}{2}\omega_0 + \omega & 0 & \cdot \\ \cdot & 0 & 0 & A & -B & 0 & -\frac{1}{2}\omega_0 + \omega & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \end{array}\right] \end{array}, \tag{41}$$

where

$$A = \tfrac{1}{4}\omega_1 \sin\theta = \omega_1\omega_\perp/4\omega_0\,,$$
$$B = \tfrac{1}{4}\omega_1 \cos\theta = \omega_1\omega_\parallel/4\omega_0\,, \tag{42}$$

and I is the unit matrix. The matrix in Eq. (41) corresponds to the Floquet Hamiltonian studied in detail by Shirley[3] if we set $\theta=\frac{1}{2}\pi$, i.e., $B=0$. From the argument in the preceding section, the variables $\omega_\parallel$ and $\omega_\perp$ are expected to be more convenient for expressing the position of the level crossings rather than the variables ω_0 and θ.

Machine calculation of energies of the coupled system was carried out by diagonalizing the matrix in Eq. (41) numerically with the Householder method. As an approximate expression of the infinite matrix in Eq. (41), we used the 40×40 matrix in the present calculation, and obtained two eigenvalues centered among 40 eigenvalues. Thus, the results correspond to the tenth-order approximation with respect to ω_1 in the perturbation theory. We estimated that the error in the present calculation was less than 0.01%, using the fact that the difference between the nth and $(n-2)$th eigenvalues is ω for any value of ω_1.

Figure 4 shows the numerically calculated energies in the case where $\omega_\parallel=0$, as a function of $\omega_\perp$ or ω_0, while ω_1 is varied from zero to 8ω as a parameter. There are of course, other energy levels which behave in the same manner as those in Fig. 4 but are centered at the vertical values $\pm1, \pm2, \ldots$. As seen in Fig. 4, the level crossings at $\omega_0 = 2n-1$ become anticrossings when ω_1 is increased. The crossings at $\omega_0=0$ and $\omega_0=2n\omega$ are not, however, removed: They are only shifted toward $\omega_\perp=0$. In the more precise calculation, we confirmed that the value of ω_1/ω which the crossing point at $\omega_0=2n\omega$ reached at $\omega_\perp=0$ coincided with the nth root of $J_0(\omega_1/\omega)=0$, where $J_0(\omega_1/\omega)$ is the zeroth-order Bessel function.

Similar results for cases $\omega_\parallel=\omega$ and $\omega_\parallel=2\omega$ are respectively shown in Figs. 5 and 6. From these results, we see that the level crossing of the second kind takes place if the condition $\omega_\parallel=n\omega$ is satisfied, even for a large value of ω_1 for which the lowest-order perturbation theory presented in the preceding section is no longer valid. All of the crossings of the second kind are shifted toward $\omega_\perp=0$ with a constant value of $\omega_\parallel$, as ω_1 is increased. We have found that the first crossing at $\omega_\perp=2\sqrt{2}$ or $\omega_0=3\omega$ in Fig. 5 enters $\omega_\perp=0$ at the value of ω_1/ω given by the first root of $J_1(\omega_1/\omega)$, and generally the nth crossing enters at the value of ω_1/ω given by the nth root. Similarly, for $\omega_\parallel=2\omega$, the values of ω_1/ω at which the crossings enter $\omega_\perp=0$ are given by the roots of $J_2(\omega_1/\omega)=0$. These facts are closely related to previous works by Pegg and Series,[16] and Yabuzaki *et al.*,[17] who have shown that the component of the atomic g factor perpendicular to the direction of the rf field is modified by the rf field, as shown in Eq. (15), if the condition $\omega_\parallel=n\omega$ is satisfied. In the present calculation, we were able to confirm that the gradient of the energy level with respect to $\omega_\perp$ at $(\omega_\parallel=n\omega, \omega_\perp=0)$ coincided with $\frac{1}{2}J_n(\omega_1/\omega)$ within an error of about 10^{-3}.

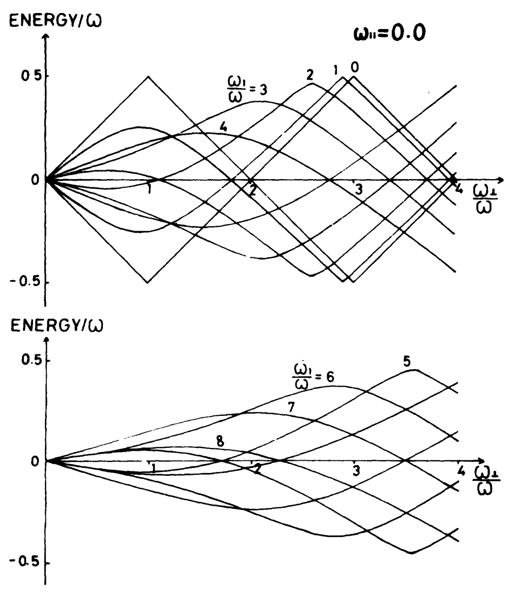

FIG. 4. Numerically calculated energies of the coupled atom–rf-field system as a function of $\omega_\perp$, in the case $\omega_\parallel=0$. ω_1/ω is treated as a parameter.

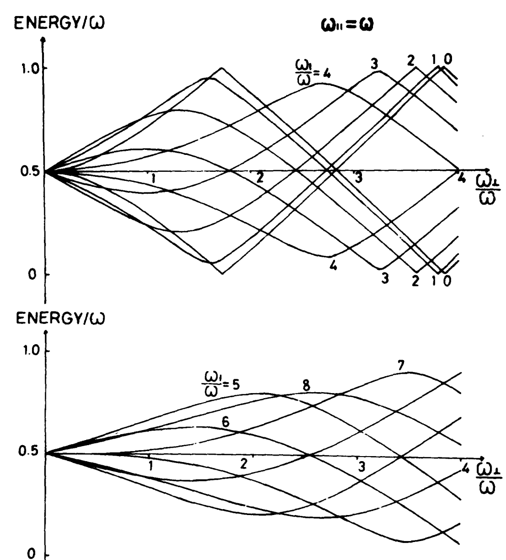

FIG. 5. Same as for Fig. 4, but with $\omega_\parallel=\omega$.

As an example of the case of $\omega_{\parallel} \neq n\omega$, we show in Fig. 7 the energies of the coupled system in the case of $\omega_{\parallel} = 0.9\omega$. This figure apparently indicates that, if the condition $\omega_{\parallel} = n\omega$ is not satisfied, all level crossings for $\omega_1 = 0$ become anticrossings; hence all multiphoton transitions are allowed.

It should be emphasized that, as seen in Figs. 5 and 6, the crossing of the second kind in the case of $\omega_{\parallel} = n\omega$ shifts along the line of $\omega_{\parallel} = n\omega$; i.e., the value of $\omega_{\parallel}$ at the crossing is not varied by the amplitude of the rf field. The amount of the shift of each level crossing calculated here is to be described later in order to compare it with the experimental results.

Figure 8 represents the energy separation, as a function of ω_1/ω, between anticrossing levels in the case of $\omega_{\parallel} = n\omega$. This energy separation is equivalent to the coupling energy between the atom and the rf field and is concerned with the probability for the multiphoton transition by the relation of Eq. (20). As seen in Fig. 8, the energy separation at the anticrossing point which is mainly related to the n-photon processes is approximately proportional to ω_1^n for a low value of ω_1, as one might expect from Eqs. (31) and (36). Furthermore, we see in Fig. 10 a saturation effect for a large value of ω_1.

IV. EXPERIMENTAL RESULTS AND COMPARISON WITH THEORY

In order to understand the existence of the level crossing of the second kind when the condition $\omega_{\parallel} = n\omega$ is satisfied, and to study quantitatively the shifts of the level crossing and anticrossing due to the rf field, an optical-pumping experiment was carried out with cesium vapor.

The directions of the static and rf fields are the same as those in Fig. 1. The optical system in the present experiment was a standard one, which was comprised of the following apparatus: a light source producing an intense collimated beam of resonance radiation of cesium, a circular polarizer, an absorption cell containing cesium vapor to be optically pumped, and a solar cell to monitor the transmission of the light beam. The absorption cell was placed in two sets of Helmholtz coils to produce the magnetic fields $H_{\parallel}$ and $H_{\perp}$, and the rf field was applied by an rf coil wound around the absorption cell. The circularly polarized resonance radiation was applied to the absorption cell in the direction of the z axis, i.e., perpendicular to the fields $H_{\parallel}$ and H_1 (see Fig. 1). The light beam transmitted through the absorption cell was detected by a solar cell, and the dc component of the output was amplified and displayed on an X-Y recorder, while $H_{\perp}$ was swept from zero. In this experimental arrangement, the multiphoton transition appears as an intensity change of the transmitted light.

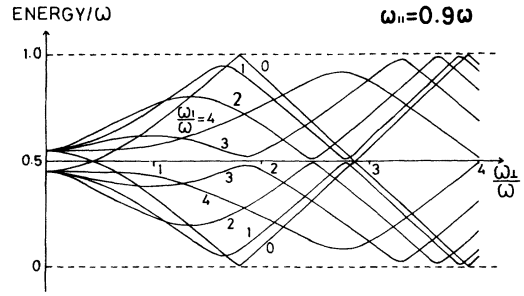

FIG. 7. Same as for Fig. 4, but with $\omega_{\parallel} = 0.9\omega$.

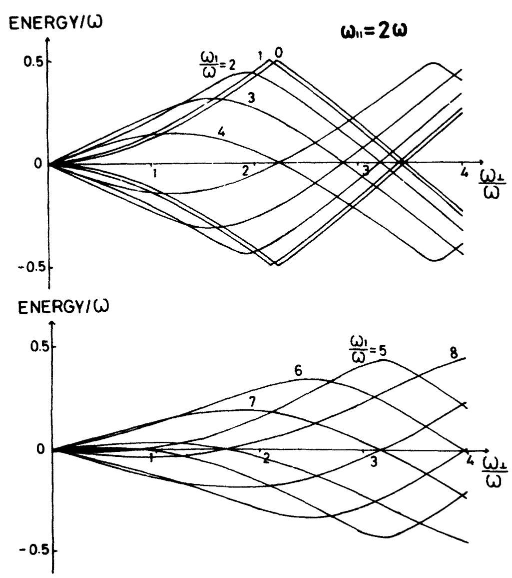

FIG. 6. Same as for Fig. 4, but with $\omega_{\parallel} = 2\omega$.

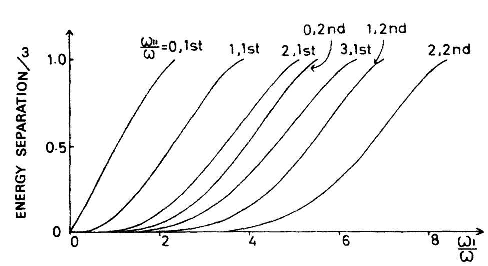

FIG. 8. Energy separation between anticrossing levels as a function of ω_1/ω. The symbol $\omega_{\parallel}/\omega = p$, "1st" and "2nd" represent the qth level anticrossing for the case $\omega_{\parallel} = p\omega$.

Figure 9 shows a typical example of the recorder traces, in which $\omega_\parallel/\omega$ is varied as a parameter and ω_1 is kept constant. In Fig. 9, V_1, which is proportional to ω_1, represents the voltage applied to the rf coil. In these recorder traces, we see that the four-photon transition becomes forbidden if $\omega_\parallel/\omega=0$ or $\omega_\parallel/\omega=2$, and the three-photon transition becomes forbidden if $\omega_\parallel/\omega=1$. These results might be sufficient to show that level crossing of the second kind takes place if the condition $\omega_\parallel=n\omega$ is satisfied, just as predicted by our theory.

Figure 10 shows the recorder traces when $\omega_\perp$ is varied and $\omega_\parallel$ is set to ω, while V_1 (or ω_1) is varied as a parameter. The resonances in these recorder traces are due to two- and four-photon transitions, and the resonance due to the three-photon transition cannot be seen for any value of ω_1. We see in Fig. 10 that the resonance lines shift toward $\omega_\perp=0$ as ω_1 is increased. But we could not determine the values of ω_1 at which the anticrossing points enter $\omega_\perp=0$ because of the broadening of the resonance lines. In the above experimental arrangement, the level crossing cannot actually be observed; hence its shifts by the rf field cannot be obtained. However, if the circularly polarized resonance radiation is applied along the direction of $H_\parallel$ instead of along that of $H_\perp$, we can observe a resonance which is an interference phenomenon between crossing energy levels. Such a resonance has actually been observed by Tsukada *et al.*[18]

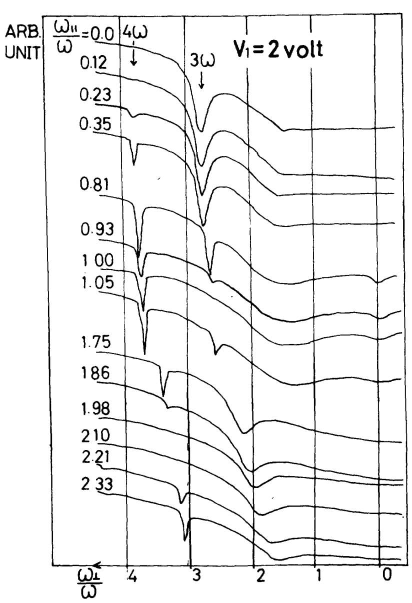

FIG. 9. Typical recorder traces showing the multiphoton transitions. $\omega_\parallel/\omega$ is treated as a parameter; V_1 represents the voltage applied to the rf coil and is proportional to ω_1.

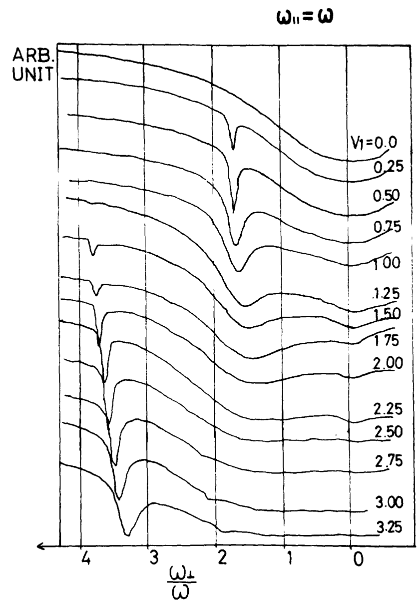

FIG. 10. Recorder traces showing multiphoton transitions for the case $\omega_\parallel=\omega$. The rf-field amplitude ω_1 (V_1 in volts) is varied as a parameter. The resonances from left to right are due to the four-photon and two-photon transitions.

The shifts of the anticrossing obtained in the present experiment and of the crossing obtained by Tsukada *et al.* are shown in Figs. 11 and 12, for the cases $\omega_\parallel=\omega$ and $\omega_\parallel=2\omega$. In these figures, (A,n) and (C,n') represent the shifts of the nth anticrossing point and the n'th crossing point, respectively, and $j_{p,q}$ represents the qth root of $J_p(\omega_1/\omega)=0$. The solid lines are the theoretical results obtained by numerical calculation in the preceding section. We see good agreement between

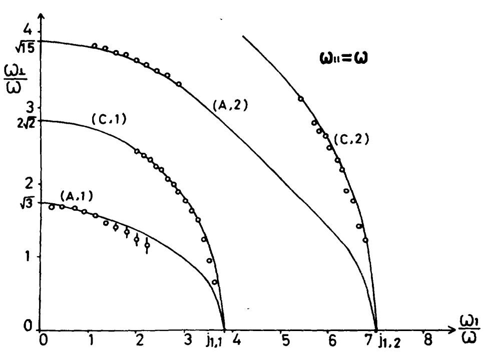

FIG. 11. Shifts of the nth anticrossing point (A,n) and the n'th crossing point (C,n') in the case of $\omega_\parallel=\omega$. Encircled points are experimental results and solid lines are theoretical ones.

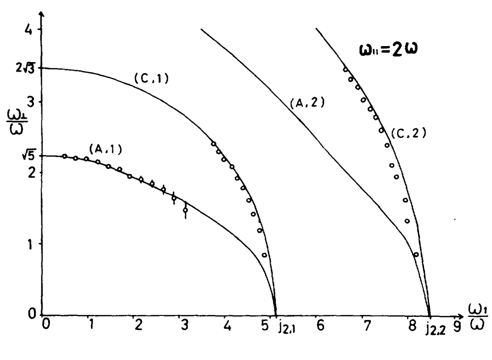

FIG. 12. Same as for Fig. 11, but with $\omega_{\parallel}=2\omega$.

experimental results and the theory. The shifts of crossing and anticrossing points when $\omega_{\parallel}=0$ are not shown here, but in this case too we obtained good agreement between the experimental results and the theory.

V. CONCLUSION

We have analyzed and numerically calculated the energies of the coupled atom–rf-field system for the case in which the oscillating rf field is oriented in an arbitrary direction with respect to the direction of the static magnetic field and the rf field amplitude is large. We have found that there exists a level crossing of the second kind when the condition $\omega_{\parallel}=n\omega$ is satisfied, where n is an integer. At the level crossing each elementary process of the multiphoton transition is allowed, but the sum of the elementary processes becomes forbidden. Furthermore, it has been found that, as the amplitude of the rf field is increased, the crossing of the second kind shifts toward ($\omega_{\parallel}=n\omega$, $\omega_{\perp}=0$) and it does not shift toward $\omega_0=0$ or ($\omega_{\parallel}=0$, $\omega_{\perp}=0$). The value of ω_1 which the crossing point reaches at the position ($\omega_{\parallel}=n\omega$, $\omega_{\perp}=0$) is determined by the root of $J_n(\omega_1/\omega)=0$. Consequently, it can be considered that the modification of atomic g factor derived by Pegg and Series[16] and Yabuzaki *et al.*[17] is due to the shifts of the level crossing of the second kind by the rf field.

The existence of the crossing of the second kind has been verified by an optical-pumping experiment with cesium vapor. The shifts of the level crossing and anticrossing by the rf field have been compared with the results of the present experiment and of the experiment by Tsukada *et al.*,[18] and good agreement has been found.

As we have treated a quite general case, the results obtained here might be applicable to the explanation of various phenomena associated with the interaction between a spin-$\frac{1}{2}$ particle and an oscillating rf field.

ACKNOWLEDGMENTS

The authors thank Dr. N. Tsukada for extensive discussions, and M. Yamanura for assistance with numerical calculations.

[1]J. Margerie and J. Brossel, C. R. Acad. Sci. (Paris) 241, 373 (1955).
[2]J. Winter, Ann. Phys. (Paris) 4, 745 (1959).
[3]J. H. Shirley, Phys. Rev. 138, B979 (1965).
[4]C. Cohen-Tannoudji and S. Haroche, C. R. Acad. Sci. (Paris) 262, 268 (1966).
[5]C. Cohen-Tannoudji and S. Haroche, J. Phys. (Paris) 30, 125 (1968); 30, 125 (1968).
[6]C. Cohen-Tannoudji, *Cargès Lectures in Physics*, edited by M. Lévy (Gordon & Breach, New York, 1968), Vol. 2, p. 347.
[7]S. Stenholm, J. Phys. B 5, 878 (1972).
[8]N. Tsukada and T. Ogawa, J. Phys. B 6, 1643 (1973).
[9]S. Stenholm and C-G. Aminoff, J. Phys. B 6, 2390 (1973).
[10]S. Haroche, C. Cohen-Tannoudji, C. Audoin, and J. P. Scherman, Phys. Rev. Lett. 24, 861 (1970).
[11]S. Haroche and C. Cohen-Tannoudji, Phys. Rev. Lett. 24, 974 (1970).
[12]G. D. Chapman, J. Phys. B 3, L36 (1970).
[13]T. Yabuzaki, N. Tsukada, and T. Ogawa, Japan. J. Appl. Phys. 11, 1071 (1972).
[14]E. B. Aleksandrov, O. V. Konstantinov, V. I. Perel', and V. A. Khodovoi, Zh. Eksp. Teor. Fiz. 45, 503 (1963) [Sov. Phys.—JETP 18, 346 (1964)].
[15]N. Polonski and C. Cohen-Tannoudji, J. Phys. (Paris) 26, 409 (1965).
[16]D. T. Pegg and G. W. Series, J. Phys. B 3, L33 (1970).
[17]T. Yabuzaki, N. Tsukada, and T. Ogawa, J. Phys. Soc. Japan 32, 1069 (1972).
[18]N. Tsukada, T. Koyama and T. Ogawa, Phys. Lett. 44A, 501 (1973).
[19]S. Haroche, Ann. Phys. (Paris) 6, 189 (1971).