Optical dark resonance in multilevel systems with a treelike configuration

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Optical dark resonance in systems with arbitrary multilevels and multiphotons is studied theoretically. When the system configuration is treelike (the term of graph theory), all the coherence terms in the system can be categorized into several groups on dark resonance. It is proved for generic system configurations with arbitrary multilevels that the resonance conditions can be obtained successively by setting the determinant of the submatrix of the Hamiltonian with respect to each group to zero. We calculate, as one of the practical examples, all the conditions of dark resonance of potassium atoms with two laser fields and one rf field mixing one excited state and eight ground states in the hyperfine structure. Although the system configuration is very complicated, the condition is easily obtained in an analytic form and verified by the result of a numerical simulation. Furthermore, our theory can reveal the fine structure of dark lines with power broadening in the observed light-induced-fluorescence spectra and numerical calculations. In addition, the dark lines with the complicated splitting pattern observed in experiments can be assigned to the relevant level configuration and transitions by the present theory. [S1050-2947(96)00808-6]

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I. INTRODUCTION

Optical dark resonance (ODR) is one of the coherent multiphoton processes and can prohibit fluorescence with a narrower resonance in the observed spectrum than the natural linewidth [1]. Its wide applicability to previous experiments such as trapping experiments and precise spectroscopies is due to its Doppler sensitive and sub-natural property. Multiphoton processes have also been investigated from the viewpoint of Raman processes from the early days [2–7]. One curious feature of the multiphoton process ODR is found as black lines in the spectrum of sodium atoms [1,8]. This type of nonabsorbing atomic coherence was discussed theoretically by Arimondo and Orriols [1] using the steady state theory for coherent two-photon process [9]. ODR in the three-level Λ configuration has been discussed in several papers [10–13] and observed in trapping and multiwave mixing experiments [8,14,15].

One of the important applications of ODR is the laser cooling below the one-photon recoil limit [16–23]. The deep cooling proved to be realized by the nonabsorbing (no pressure) states for trapped atoms at zero velocity due to ODR. Another application is the analytic prediction and intuitive interpretation of the line-shape function of laser-induced fluorescence (LIF), as discussed in this paper. Because the direct calculation of the LIF spectrum is known to give a large analytical result, as in our previous paper [24], even for a five-level system, the determination of the condition of ODR is indispensable to get a more intuitive interpretation of the spectrum. The line shape of LIF is affected drastically by ODR because ODR may appear at nearly resonant condition between fields and materials and make the strong fluorescence vanish completely. Especially in the intense fields, the effect of ODR becomes dominant, and the explanation of the whole line shape of LIF requires not only the analysis of the usual intense-field approximation but also the fine structure due to ODR.

For example, consider the double-resonance experiment of Fig. 1(a), which may often be the case of various atoms such as potassium ($I=\frac{3}{2}$). The line shape of LIF might be expected to have a single peak when the frequency of the rf field is fixed and that of the laser field is scanned. However, taking into account the detailed level structure (Zeeman sublevels) and the selection rule, the Fig. 1(a) becomes a system that includes nine levels and eight radiations, as in Fig. 1(b). (We assume appropriate polarizations for the laser and rf fields here.) Note that one rf and laser field can drive several transitions simultaneously, as seen in Figs. 1(a) and 1(b), when the relevant transitions are nearly degenerate or the resonant Rabi frequencies of the fields are large enough. The intensity of LIF from this system of Fig. 1(b) is numerically calculated as in Fig. 1(c), in which the frequency of the rf field is fixed and that of the laser is scanned (x axis). The resonant Rabi frequency of the laser fields is intentionally set much larger than the reciprocal number of the lifetime of the atom to make ODR dominant in the spectrum. Though the outline of this spectrum surely has a single peak as expected, the complicated structure of the near-resonant condition is remarkable. This is because ODR makes the fluorescence vanish even in the near-resonant condition for strong fluorescence and therefore ODR may have a large influence on line shape.

In this paper we show the analytic solution of all the conditions for ODR in multilevel and multiradiation systems under the rotating-wave approximation (RWA). The condition of the radiation frequency and strength for ODR in the multilevel system is determined for arbitrary multiphoton processes. For example, all the positions of ODR in the system of Fig. 1(b) are determined analytically by a simple matrix calculation. From the present theory, the LIF spectrum of Fig. 1(c) turns out to contain 15 dark lines (see later Fig. 7), where the numerical result [Fig. 1(c)] is too obscure to find this fine structure. In addition, the intuitive interpretation of the splitting pattern of dark lines has proved to be possible for complicated systems such as that in Fig. 1(b). Therefore the parametric dependence of the LIF spectrum in multilevel

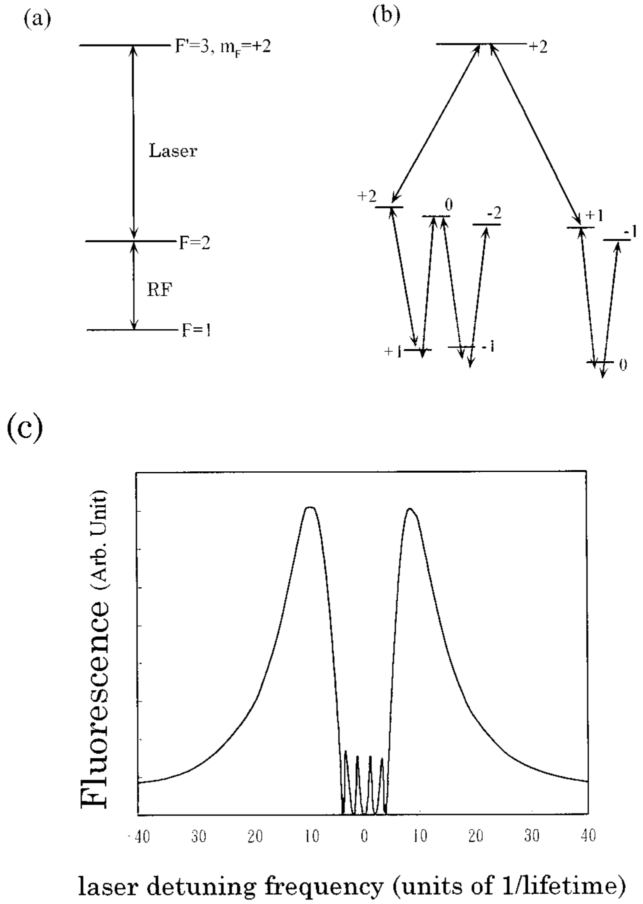

FIG. 1. (a) Example of multilevel laser rf double-resonance system, one of which is an alkali metal having $I=\frac{3}{2}$. (b) Detailed configuration of (a), considering Zeeman sublevels. (c) Typical LIF spectrum for the system (b) when one of the frequencies of the laser fields is scanned. All the parameters are the same as the example discussed in Fig. 7.

systems on field intensity and detuning frequency can be determined in detail.

In Sec. II we show that the steady-state resonance of ODR in the multilevel system could be categorized into two kinds of resonance conditions described by simple matrix determinant forms under the RWA of the optical Bloch equation. One of these determinants is proved to be the condition for ODR and the other is reduced to a simpler form with again two kinds of subdeterminants in Sec. III. Therefore, successive application of this procedure reveals all the branches of dark resonance in an arbitrarily complicated system (Sec. III B). However, not all these branches are observed in the LIF spectrum in some critical cases due to the double valuedness of the steady-state condition, which we discuss in Sec. III C. We calculate, as one of the practical examples, the condition of dark resonance of potassium atoms with two laser fields and one rf field mixing one excited state and eight ground states in the hyperfine structure (Sec. IV). We also show that our theory can resolve the dark lines with power broadening in the observed LIF spectrum into a fine structure. Finally, we conclude our theory in Sec. V.

II. THE CONDITION FOR DARK RESONANCE IN AN *N*-LEVEL SYSTEM

In this paper we treat those systems with arbitrary multilevels and multi-radiations such that all the levels are connected by the radiations without constructing any cycle. In other words, the level scheme forms a tree (a term of graph theory). Several proofs below are based on this requirement. Note that a tree need not grow in one direction. [Figure 1(b) is a kind of tree.] Binary trees are not assumed. In addition, while the decay terms for the population or the coherence of the optically excited level are always necessary for the phenomenon of ODR, those in the ground states tend only to make the resonance of ODR dull. In order to make our theory clear, we neglect the latter throughout this paper. This is a reasonable assumption when the energy spacings within the ground states are assumed to be in the rf (or microwave) region and they can be stable enough in the realistic experiments.

The well-known optical Bloch equation for the density matrix ρ is

$$i\hbar \frac{\partial}{\partial t}\rho = H\rho - \rho H + i\hbar \mathscr{D}, \tag{1}$$

where the decay term $\mathscr{D}$ is

$$\mathscr{D}=\begin{cases} -\dfrac{1}{T}\rho_{00} & \text{for the excited-state population} \\ -\dfrac{1}{T_2}\rho_{pq} & \text{for optical coherence } (p=0 \text{ or } q=0) \\ +\dfrac{f_p}{T}\rho_{00} & \text{for the ground-state populations} \end{cases}$$

and

$$\sum f_n = 1, \quad \sum \rho_{nn} = 1. \tag{2}$$

The subscript 0 means the unique optically excited level. (Considering more than two excited states does not lead to any essential change in the proof below.) T and T_2 are the decay times for the optical population and coherence, respectively. f_p is called the repopulation ratio [1] and defines the ratio of how the population in the optical excited state is redistributed to the ground state p. The Hamiltonian is defined as

$$H_{pq} = \delta_{pq}E_p + \hbar\Omega_{pq}(\exp\{i[(E_p - E_q)/\hbar + \Delta_{pq}]t + i\,\theta_{pq}\} + \text{c.c.}), \tag{3}$$

where E_p is the energy of the level p, Δ_{pq} is the detuning frequency of the radiation from the atomic transition between levels p and q, θ_{pq} is the constant phase factor of the field, and $\Omega_{pq} = \mu_{pq}E_{pq}/2\hbar$ is the Rabi frequency for the resonant radiation between p and q levels.

First, we introduce slow varying amplitude $\tilde{\rho}$ to subtract the phase factors rotating at the frequencies of the radiations and apply the RWA in the ordinary manner. $\tilde{\rho}$ is defined as

$$\rho_{pq}(t)=\tilde{\rho}_{pq}(t)\exp\left[-i\left((E_p-E_q)/\hbar+\sum_{p,\ldots,ab,\ldots,q}(\pm\Delta_{ab})\right)\times t-i\sum_{p,\ldots,ab,\ldots,q}(\pm\theta_{ab})\right], \tag{4}$$

where p and q need not to be adjacent levels. $p,\ldots,ab,\ldots,q$ means that the subscript ab runs along the radiation network from the level p to q. The uniqueness of the path from p to q is ensured since the system is like a tree and has no cycle. The double sign of Δ_{ab} or θ_{ab} is taken to be the same as that of (E_a-E_b). The diagonal term $\tilde{\rho}_{nn}$ is the same as ρ_{nn}. After substituting Eqs. (3) and (4) in the Schrödinger equation (1), setting $d\tilde{\rho}_{nn}/dt=0$, and neglecting the terms rapidly rotating at the double frequency of any radiation, one gets a set of linear equations that does not contain the time t explicitly. In addition, a careful calculation verifies that all of the phase constants θ_{ab} cancel each other.

When the system under study has more than one optically excited state and several optical transitions, the definition of dark resonance becomes ambiguous: Is dark resonance defined for the state with no fluorescence for all optical transitions or one specific transition? In any case, the system with one optically excited state and two laser fields is the basis of analysis, which we assume here the system is like. (We discuss the system with several excited levels and more than two laser fields at the end of Sec. III A.) We categorize all the ground states into two groups P and Q, where $p_i\in P$ $(i=0,1,\ldots,n_p-1)$ and $q_s\in Q$ $(s=0,1,\ldots,n_q-1)$ are linked only with the element(s) in the same group by rf transition(s). p_0 and q_0 denote those top levels in each group that have optical transitions with the excited level [Fig. 1(c)]. This discrimination is not trivial, because it is essential for ODR that the optical coherence $\tilde{\rho}_{0x},\tilde{\rho}_{x0}$ and population $\tilde{\rho}_{00}$, which would connect the two groups, vanish and disconnect the group P from Q. (For example, the cascade type of three-level system does not exhibit ODR because the ground states cannot be divided like P and Q.)

Dark resonance is observed as a sharp dip in the LIF spectrum [1]. It is well known that the depth of the dip depends on the relaxation time of the coherence and population in the ground states (not the excited states). On our assumption that all the relaxations within the ground states are negligible, the dip drops exactly to zero (i.e., no fluorescence is observed). Therefore, in order to obtain the condition required for ODR, we can replace the optical population $\tilde{\rho}_{00}$ in the set of the linear equations for the steady state simply with 0 since no population exists in the excited state on the ODR condition. $\tilde{\rho}_{0x}=\tilde{\rho}_{x0}=0$ follows the condition of $\tilde{\rho}_{00}=0$. Note that, while setting $\tilde{\rho}_{nn}=\alpha$ does not determine $\tilde{\rho}_{nx}$ and $\tilde{\rho}_{xn}$ in general, $\tilde{\rho}_{nn}=0$ does whether the system is in a pure or mixed state. After the substitutions mentioned above, we get

$$\dot{\tilde{\rho}}_{p_ip_i}=\sum_{p_x\in P}\Omega_{p_ip_x}\tilde{\rho}_{p_xp_i}-\sum_{p_x\in P}\tilde{\rho}_{p_ip_x}\Omega_{p_xp_i}=0, \tag{5a}$$

$$\dot{\tilde{\rho}}_{p_ip_j}=-(\pm\Delta_{p_ip_j})\tilde{\rho}_{p_ip_j}+\Omega_{p_ip_j}\tilde{\rho}_{p_jp_j}-\Omega_{p_jp_i}\tilde{\rho}_{p_ip_i}+\sum_{x\neq j}\Omega_{p_ip_x}\tilde{\rho}_{p_xp_j}-\sum_{x\neq i}\Omega_{p_xp_j}\tilde{\rho}_{p_ip_x}=0 \quad (\Omega_{p_ip_j}\neq 0,\ i\neq j), \tag{5b}$$

$$\dot{\tilde{\rho}}_{0p_0}=-\Omega_{0p_0}\tilde{\rho}_{p_0p_0}-\Omega_{0q_0}\tilde{\rho}_{q_0p_0}=0, \tag{5c}$$

$$\dot{\tilde{\rho}}_{0p_i}=-\Omega_{0p_0}\tilde{\rho}_{p_0p_i}-\Omega_{0q_0}\tilde{\rho}_{q_0p_i}=0 \quad (i\neq 0), \tag{5d}$$

$$\dot{\tilde{\rho}}_{p_ip_j}=-\left(\sum_{p_i,\ldots,ab,\ldots,p_j}(\pm\Delta_{ab})\right)\tilde{\rho}_{p_ip_j}+\sum_{x\neq i}\Omega_{p_ip_x}\tilde{\rho}_{p_xp_j}-\sum_{x\neq j}\Omega_{p_xp_j}\tilde{\rho}_{p_ip_x}=0 \quad (\Omega_{p_ip_j}=0,\ i\neq j), \tag{5e}$$

$$\dot{\tilde{\rho}}_{p_iq_s}=-\left(\sum_{p_i,\ldots,ab,\ldots,q_s}(\pm\Delta_{ab})\right)\tilde{\rho}_{p_iq_s}+\sum_{x\neq i}\Omega_{p_ip_x}\tilde{\rho}_{p_xq_s}-\sum_{x\neq s}\Omega_{q_xq_s}\tilde{\rho}_{p_iq_x}=0, \tag{5f}$$

where $i=0,1,\ldots,n_p-1$, $j=0,1,\ldots,n_p-1$, and $s=0,1,\ldots,n_q-1$. Note that Eqs. (5) include no decay terms. The positions at which ODR appears are independent on the lifetime T of the atoms, the decay time T_2 of optical coherence, and the repopulation ratios f_n. (The line profile is of course affected by them.)

One of our main purposes in this paper is to obtain the condition in which (5) has nontrivial solution of $\tilde{\rho}$. Note that, when the duplicated complex conjugates, the meaningless equation for $d\tilde{\rho}_{00}/dt$, and the variables $\tilde{\rho}_{0x},\tilde{\rho}_{x0},\tilde{\rho}_{00}$ are excluded, Eqs. (5) are ${}_{n_p+n_q+1}C_2+(n_p+n_q+1)$ equations with ${}_{n_p+n_q+1}C_2$ variables. First, we reduce the number of the equations by applying a relation

$$\mathrm{Im}(\tilde{\rho}_{p_ip_j})=0 \quad \text{when} \quad \Omega_{p_ip_j}\neq 0, \tag{6}$$

which is equivalent to Eq. 5(a). The proof of Eq. (6) is based on the assumption that the network of the energy levels and the radiations is treelike again (Appendix A).

Therefore we can exclude the n_p+n_g equations of (5a) from consideration, requiring that $\tilde{\rho}_{ij}=\tilde{\rho}_{ji}$ $(\Omega_{ij}\neq 0)$. At this point, one can define a square matrix H_{ODR} and a vector $\tilde{\rho}_{\mathrm{ODR}}$, which consist of the components of the density matrix, and get an equivalent expression $H_{\mathrm{ODR}}\tilde{\rho}_{\mathrm{ODR}}=0$ for them because (5b)–(5f) have the same number of variables as that of the equations (5b–5f). It is clear that $|H_{\mathrm{ODR}}|=0$ is the necessary condition to seek. We expand $|H_{\mathrm{ODR}}|$ systematically into a simpler form below.

In simplifying $|H_{\mathrm{ODR}}|$, (5b), (5c), and the columns for $d\tilde{\rho}_{p_ip_i}/dt$ and $d\tilde{\rho}_{p_ip_i}/dt$ can be expanded with nonzero factors such as $\Omega_{p_ip_j}$. The proof is also based on the assumption of the system being a tree (Appendix B).

The remainders in (5) are (5d)–(5f). Equation (5f) does not have such matrix elements like $\widetilde{\rho}_{p_i p_j}$ and (5e) does not have those like $\widetilde{\rho}_{p_i q_j}$. Therefore, using the matrix H'_{ODR} relevant for (5d)–(5f), they can be symbolically expressed in the form

$$H'_{\mathrm{ODR}}\widetilde{\rho}_{\mathrm{ODR}}=0, \tag{7}$$

where

$$H'_{\mathrm{ODR}}=\begin{pmatrix}
-\Sigma(\pm\Delta) & -\Omega_{q_0q_s} & \Omega_{p_ip_x} & \cdots & Q_{p_ip_0} & -\Omega_{q_xq_s} & \cdots & 0 & 0 & \cdots & 0 & \cdots & 0 & 0 & 0 & \cdots & 0 & \cdots & 0 \\
\vdots & \vdots & \vdots & & \vdots & \vdots & & \vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots \\
0 & 0 & 0 & \cdots & 0 & 0 & \cdots & -\Sigma(\pm\Delta) & \Omega_{p_ip_x} & \cdots & -\Omega_{p_xp_f} & \cdots & -\Omega_{p_0p_f} & 0 & 0 & \cdots & 0 & \cdots & 0 \\
\vdots & \vdots & \vdots & & \vdots & \vdots & & \vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots \\
0 & 0 & 0 & \cdots & 0 & 0 & \cdots & 0 & 0 & \cdots & 0 & \cdots & 0 & -\Sigma(\pm\Delta) & \Omega_{q_xq_i} & \cdots & -\Omega_{q_xq_j} & \cdots & -\Omega_{q_0q_j} \\
\vdots & \vdots & \vdots & & \vdots & \vdots & & \vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots \\
0 & -\Omega_{0q_0} & 0 & \cdots & 0 & 0 & \cdots & 0 & 0 & \cdots & 0 & \cdots & -\Omega_{0p_0} & 0 & 0 & \cdots & 0 & \cdots & 0 \\
\vdots & \vdots & \vdots & & \vdots & \vdots & & \vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots \\
0 & 0 & 0 & \cdots & -\Omega_{0p_0} & 0 & \cdots & 0 & 0 & \cdots & 0 & \cdots & 0 & 0 & 0 & \cdots & 0 & \cdots & -\Omega_{0q_0} \\
\vdots & \vdots & \vdots & & \vdots & \vdots & & \vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots
\end{pmatrix}$$

and

$$\widetilde{\rho}_{\mathrm{ODR}}={}^{T}(\widetilde{\rho}_{p_iq_s}\widetilde{\rho}_{p_iq_0}\widetilde{\rho}_{p_xq_s}\cdots\widetilde{\rho}_{p_0q_s}\widetilde{\rho}_{p_iq_x}\cdots \quad \widetilde{\rho}_{p_ip_j}\widetilde{\rho}_{p_xp_j}\cdots\widetilde{\rho}_{p_ip_x}\cdots\widetilde{\rho}_{p_ip_0}\widetilde{\rho}_{q_iq_j}\widetilde{\rho}_{q_sq_j}\cdots\widetilde{\rho}_{q_iq_s}\cdots\widetilde{\rho}_{q_iq_0}).$$

The determinant of H'_{ODR} is reduced in a well-known manner as

$$|H'_{\mathrm{ODR}}|=\begin{vmatrix}
-\Sigma(\pm\Delta) & -\Omega_{q_0q_s} & \Omega_{p_ip_x} & \cdots & \Omega_{p_ip_0} & -\Omega_{q_xq_s} & \cdots \\
\vdots & \vdots & \vdots & & \vdots & \vdots &
\end{vmatrix}$$

$$\times\begin{vmatrix}
-\Sigma(\pm\Delta) & \Omega_{p_ip_x} & \cdots & -\Omega_{p_xp_j} & \cdots & -\Omega_{p_0p_j} & 0 & 0 & \cdots & 0 & \cdots & 0 \\
\vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots \\
0 & 0 & \cdots & 0 & \cdots & 0 & -\Sigma(\pm\Delta) & \Omega_{q_xq_i} & \cdots & -\Omega_{q_xq_j} & \cdots & -\Omega_{q_0q_j} \\
\vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots \\
0 & 0 & \cdots & 0 & \cdots & -\Omega_{0p_0} & 0 & 0 & \cdots & 0 & \cdots & 0 \\
\vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots \\
0 & 0 & \cdots & 0 & \cdots & 0 & 0 & 0 & \cdots & 0 & \cdots & -\Omega_{0q_0} \\
\vdots & \vdots & & \vdots & & \vdots & \vdots & \vdots & & \vdots & & \vdots
\end{vmatrix}. \tag{8}$$

(Note that the first determinant of the right-hand side is that of a squared matrix.) The second one has a single component, such as $(-\Omega_{0p_0}\tilde{\rho}_{p_0p_i})$, in the row that can be expanded along the row with the nonzero factor $(-\Omega_{0p_0})$. Simultaneously, the columns for $\tilde{\rho}_{p_0p_i}$ (or $\tilde{\rho}_{q_0q_i}$), that is, the coherence between the ground state q_0 (or p_0) and all the levels belonging to the group P (or Q), can be eliminated in Eq. (8). The dark resonance in a complicated N-level system that has several branches in its level configuration is characterized by this elimination. The requirement $\rho_{00}=0$ descends to the subsystem P (or Q) to let $\tilde{\rho}_{p_0p_i}$ and $\tilde{\rho}_{q_0q_i}$ be zero, which we discuss in Sec. III B.

The first determinant on the right-hand side in Eq. (8) contains the coefficients for $d\tilde{\rho}_{pq}/dt$ and their complex conjugates for $d\tilde{\rho}_{qp}/dt$. From Eq. (5f), it can be seen that the coefficients for $\tilde{\rho}_{pq}$ are completely decoupled from those for $\tilde{\rho}_{qp}$. If we define the coefficient matrix for $\tilde{\rho}_{pq}$ as H_S, the first determinant on the right-hand side in Eq. (8) including both $\tilde{\rho}_{pq}$ and $\tilde{\rho}_{qp}$ can be reduced as $|H_S|^2$.

We conclude this section with the following proposition. The condition required for ODR is that at least one of the following sets of equations has a nontrivial solution for $\tilde{\rho}$:

$$\begin{aligned}\left(\sum_{p_i,\ldots,ab,\ldots,p_j}(\pm\Delta_{ab})\right)\tilde{\rho}_{p_ip_j}&=\sum_{x\neq i,j,0}\Omega_{p_ip_x}\tilde{\rho}_{p_xp_j}-\sum_{x\neq i,j,0}\Omega_{p_xp_j}\tilde{\rho}_{p_ip_x}\quad (p_ip_j:\quad \Omega_{p_ip_j}=0,\ i\neq 0,\ j\neq 0,\ i\neq j),\\ 0&=\sum_{x\neq i,j,0}\Omega_{p_ip_x}\tilde{\rho}_{p_xp_j}-\sum_{x\neq i,j,0}\Omega_{p_xp_j}\tilde{\rho}_{p_ip_x}\quad (p_ip_j:\quad \Omega_{p_ip_j}=0,\ i=0\ \text{or}\ j=0,\ i\neq j),\end{aligned}\tag{9}$$

or

$$\left(\sum_{p_i,\ldots,ab,\ldots,q_j}(\pm\Delta_{ab})\right)\tilde{\rho}_{p_iq_j}=\sum_{x\neq i}\Omega_{p_ip_x}\tilde{\rho}_{p_xq_j}-\sum_{x\neq j}\Omega_{q_xq_j}\tilde{\rho}_{p_iq_x},\tag{10}$$

or an equation similar to Eq. (9) for the group Q. That is, in a matrix form,

$$|H_{S_1}|=0\tag{11}$$

or

$$|H_{S_2}|=0\tag{12}$$

or

$$|H_{S_3}|=0,\tag{11$'$}$$

where the definition of H_{S_1} is, for the indices $m=(p_ip_j)$ and $n=(p_kp_l)$,

$$(H_{S_1})_{mn}=\begin{cases}\sum_{p_i,\ldots,ab,\ldots,p_j}(\pm\Delta_{ab}), & i=k,\ j=l\\ -\Omega_{p_lp_j} & i=k,\ j\neq l\\ \Omega_{p_ip_k} & i\neq k,\ j=l\\ 0 & \text{otherwise}\end{cases}\tag{13}$$

and H_{S_2}, for $m=(p_iq_j)$ and $n=(p_kq_l)$,

$$(H_{S_2})_{mn}=\begin{cases}\sum_{p_i,n\ldots,ab,\ldots,q_j}(\pm\Delta_{ab}), & i=k,\ j=l\\ -\Omega_{p_lq_j} & i=k,\ j\neq l\\ \Omega_{p_iq_k} & i\neq k,\ j=l\\ 0 & \text{otherwise.}\end{cases}\tag{14}$$

H_{S_3} can be obtained if one replaces the p's with q's in the definition of H_{S_1}. Whether Ω_{xy} is zero or nonzero depends on the actual form of the system under study. It is easily verified that H_S is Hermitian (or symmetric because we take Ω to be real), if we redefine H_S so that the coefficients $\Sigma_{ab}(\pm\Delta_{ab})$ are located on the diagonal part of the matrix.

III. TWO BRANCHES OF DARK RESONANCE

The condition for ODR obtained in the preceding section consists of two kinds of solutions derived from Eqs. (9) and (10). These two branches of ODR have different properties. For example, (9) is independent of the frequencies of lasers, but (10) is not, which means that even their applications for the experiments differ from one another. We first discuss the condition of Eq. (12) in Sec. III A, which is important especially for trapping experiments, and second those of Eqs. (11) and (11$'$) in Sec. II B. One more important problem is the convergence of the time development of the ODR signal. It is proved that the assumption of $\rho_{00}=0$ gives us the resonance condition of ODR in the steady-state treatment for almost all configurations of the system in Sec. III C.

A. Coherence across the optical bridge

Recall that the Hermitian of Eq. (14) is obtained by picking up, from the Hamiltonian for the whole system under study, the components of the coherence created between the two groups of the energy levels P and Q that share a common level by radiations. The resultant matrix proves to coincide with the Hamiltonian of a new system including only P and Q, which are uncoupled from each other, while the basis set of it is the direct product of each of the states in P and Q, $\{|P_i\rangle|Q_j\rangle\}$. The components of the diagonal part are determined by sequential sums of the detuning frequencies along the radiation network. The eigenvalues of H_{DR} are the direct sum of the perturbed energy (self-energy in the dressed-atom picture) of P and Q by the radiations within each of them. Dark resonance appears when the difference of the detuning frequencies of the two laser fields is equal to that of the eigenvalues. In other words, it is when the laser fields resonantly stimulate the transition between P and Q, which are shifted by rf transitions. It must be emphasized that the optical transitions do not shift the resonance position at all, which discriminates dark resonance from other multiphoton processes. For example, Eq. (10) and its determinant for the five-level symmetric system [24] are written as

$$\begin{vmatrix} \Delta_{\rm laser} & \Omega_{\rm rf1} & -\Omega_{\rm rf2} & 0 \\ \Omega_{\rm rf1} & \Delta_{\rm laser}-\Delta_{\rm rf2} & 0 & -\Omega_{\rm rf2} \\ -\Omega_{\rm rf2} & 0 & \Delta_{\rm laser}+\Delta_{\rm rf1} & \Omega_{\rm rf1} \\ 0 & -\Omega_{\rm rf2} & \Omega_{\rm rf1} & \Delta_{\rm laser}+\Delta_{\rm rf1}-\Delta_{\rm rf2} \end{vmatrix} = 0, \tag{15}$$

$$\Delta_{\rm laser}^4 + 2\Delta_{\rm rf}\Delta_{\rm laser}^3 + \{\Delta_{\rm rf}^2 - \Delta_{\rm rf1}\Delta_{\rm rf2} - 2\Omega_{\rm rf}^2\}\Delta_{\rm laser}^2 - \{\Delta_{\rm rf1}\Delta_{\rm rf2} + 2\Omega_{\rm rf}^2\}\Delta_{\rm rf}\Delta_{\rm laser} + (\Omega_{\rm rf1}^2 - \Omega_{\rm rf2}^2)^2 - (\Omega_{\rm rf2}^2\Delta_{\rm rf1}^2 + \Omega_{\rm rf1}^2\Delta_{\rm rf2}^2) + \Omega_{\rm rf}^2\Delta_{\rm rf1}\Delta_{\rm rf2} = 0, \tag{16}$$

where

$$\Omega_{\rm rf} = \sqrt{\Omega_{\rm rf1}^2 + \Omega_{\rm rf2}^2},$$

$$\Delta_{\rm rf} = \Delta_{\rm rf1} - \Delta_{\rm rf2}.$$

We return to the general case, Eq. (10). Noticing that the subscript ab in $\Sigma_{p_i,\ldots,ab,\ldots,q_j}(\pm\Delta_{ab})$ always runs through $p_0\rightarrow 0$, $0\rightarrow q_0$, that is, the optical bridge. Because p_i and q_j belong to the deferent banks, it can be said that $\Sigma_{p_i,\ldots,ab,\ldots,q_j}(\pm\Delta_{ab})$ always contains both $\Delta_{\rm laser1}$ and $\Delta_{\rm laser2}$ as a form of $\Delta_{\rm laser1}-\Delta_{\rm laser2}\equiv\Delta_{\rm laser}$. This property is not seen in (9), where ab runs within a bank in $\Sigma_{p_i,\ldots,ab,\ldots,p_j}(\pm\Delta_{ab})$. In other words, the Doppler-sensitive dark resonance, which is important for trapping experiments, is obtained only from (10) and the branches determined by (9) are insensitive to the velocity of an atom because the rf's are low enough and the effect of the Doppler shift on $\Delta_{\rm rf}$ for the condition of ODR is negligible.

In summary, the optical frequencies at which the dark lines appear are therefore easily obtained, for any system configuration with a tree form, without quite a lengthy derivation of $\tilde{\rho}_{00}$ [24] by picking up the equations for $\tilde{\rho}_{p_iq_j}$ (the coherence between two banks P and Q) from Eq. (5) and requiring the determinant of the coefficient matrix to be zero. In addition, one finds a useful relation between the system configuration and the number of dark lines. The number of lines is

$$n_{\rm ODR} = n_p n_q \tag{17}$$

if one neglects the accidental degeneration of dark lines.

In addition, it is clear that ODR does not appear when $n_q=0$ (single bank), which corresponds to, e.g., the three-level cascade system. Furthermore, if the system has many banks such as $(P,Q,R,\ldots)$, the number of ODR lines is $\Sigma_{A,B}(n_A n_B)$, $A,B\in\{P,Q,R,\ldots\}$, $A\neq B$. (The proof is given in the same way as that for two banks.) The condition for ODR is $|H_S|=0$ for any two combination of two banks A and B.

B. Coherence within each bank

This subsection is devoted to the explanation of the dark resonance of Eqs. (11) and (11′). This type of resonance is detected by scanning the frequencies not of lasers, but those of rf fields while the signal is observed as a dip in LIF as ODR is. [This is easily confirmed from the fact that the required condition (9) does not contain the detuning frequencies of lasers at all.] One might think that the dark resonance in the rf domain (RFDR) should be the same as the ODR discussed above. For example, in Fig. 2, a well-known type of dark resonance within the three-level Λ configuration $\{|1\rangle,|3\rangle,|4\rangle\}$ should occur when $\Delta_{13}=\Delta_{14}$, with no fluorescence observed. However, in our derivation of Eq. (9) from Eq. (1), we assume that only the population in $|0\rangle$ is exhausted and not in $|1\rangle$, like the case of Sec. III A. In addition, none of the optical pumping required for dark resonance occurs since we assume no incoherent decays within $\{|1\rangle,|3\rangle,|4\rangle\}$. (This means that the time development does not necessarily converge to the dark state.) In spite of these different situation, most cases allow RFDR to appear in the LIF spectrum when $\Delta_{13}=\Delta_{14}$, as follows.

It is clear that Eq. (9) includes only the coherence components of the density matrix within the group P. In addition, the terms $\rho_{p_0p_X}$ or $\rho_{p_Xp_0}$, which are relevant to the top level of the tree P, have been already eliminated from the equations in Sec. III A or, in another sense, they can be said to be assumed zero. These situation are just the same as Eqs. (5d)–(5f) in Sec. II, where ρ_{0X} and ρ_{X0} are forced to be zero for

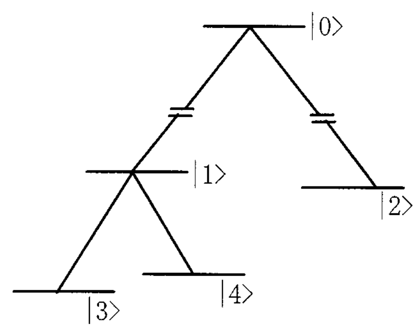

FIG. 2. Simplest level configuration in which RFDR appears as well as ODR.

ODR if we replace the index p_0 by 0. It may be surprising that the condition for RFDR, that is, $\rho_{p_0 p_X}=\rho_{p_X p_0}=0$, is not assumed but automatically introduced by the condition for ODR, $\rho_{00}=\rho_{0X}=\rho_{X0}=0$.

Now we call the subbanks hooked on the level p_0 by R and S, whose elements of the levels are r_i and s_i, respectively. Successive application of the procedure explained below Eqs. (5d)–(5f) reduces the condition (9) for RFDR to

$$\begin{aligned}\left(\sum_{r_i,\ldots,ab,\ldots,r_j}(\pm\Delta_{ab})\right)\tilde{\rho}_{r_i r_j}&=\sum_{x\neq i,j,0}\Omega_{r_i r_j}\tilde{\rho}_{r_x r_x}-\sum_{x\neq i,j,0}\Omega_{r_x r_j}\tilde{\rho}_{r_i r_x}\quad (r_i r_j:\quad \Omega_{r_i r_j}=0,\ i\neq 0,\ j\neq 0,\ i\neq j),\\ 0&=\sum_{x\neq i,j,0}\Omega_{r_i r_x}\tilde{\rho}_{r_x r_j}-\sum_{x\neq i,j,0}\Omega_{r_x r_j}\tilde{\rho}_{r_i r_x}\quad (r_i r_j:\quad \Omega_{r_i r_j}=0,\ i=0\ \text{or}\ j=0,\ i\neq j),\end{aligned}\tag{18}$$

$$\left(\sum_{r_i,\ldots,ab,\ldots,s_j}(\pm\Delta_{ab})\right)\tilde{\rho}_{r_i s_j}=\sum_{x\neq i}\Omega_{r_i r_x}\tilde{\rho}_{r_x s_j}-\sum_{x\neq j}\Omega_{s_x s_j}\tilde{\rho}_{r_i s_x},\tag{19}$$

with an equation similar to Eq. (18) for group S. Again, the condition in which Eq. (19) has a nontrivial solution for the ρ_{ij}s can be simply represented as a determinant of the coefficient matrix being zero. Equation (18) can be reduced further by the same procedure. Therefore, the present theory is self-scalable for the subbanks in the system. The condition for dark resonance can be obtained for an arbitrary multilevel system by a cascaded application of the procedure in Sec. II. Eventually, all of the RFDR conditions can be obtained by picking up all of the coherences across the bridge in the subbanks successively.

RFDR is Doppler insensitive. In fact, Eqs. (18) and (19) do not contain the detuning frequency that is determined by the velocity of atoms as well as the Rabi frequency of the optical field (but rf field) and one cannot observe RFDR in LIF when the frequency of laser is scanned. In addition, RFDR can be missing in LIF if the system is configured in a particular way, discussed in the next subsection.

C. Critical condition for RFDR

One problem important especially for ODR and RFDR is whether or not the steady-state solution of the system is uniquely determined under any initial population distribution. For example, since three-level Λ [1] and five-level symmetric configurations [24] have an (analytic) unique expression for the steady-state line profile of LIF, the observed spectrum is proved to be independent of the initial condition. The conclusion is that ODR is always observable for any case. On the other hand, RFDR can be missing when the system is configured in a particular way, as discussed in this subsection. Before proceeding, we show the simplest example of this critical condition. Consider the dark resonance of a five-level system, such as that in Fig. 2. ODR appears at those laser and rf frequencies derived from Eq. (10),

$$\begin{vmatrix}\Delta_{01}-\Delta_{02} & \Omega_{13} & \Omega_{14}\\ \Omega_{13} & \Delta_{13}+\Delta_{01}-\Delta_{02} & 0\\ \Omega_{14} & 0 & \Delta_{14}+\Delta_{01}-\Delta_{02}\end{vmatrix}=0,\tag{20}$$

taking into account the coefficients that prefix the coherences across the optical bridge, that is, ρ_{12}, ρ_{32}, and ρ_{42} in the Schrödinger equation (1). In addition, RFDR within the bank $\{|1\rangle,|3\rangle,|4\rangle\}$ is derived from Eq. (19) simply as

$$\Delta_{13}=\Delta_{14}.\tag{21}$$

Note that Eqs. (20) and (21) are independent of the repopulation ratio of fluorescence f_n. However, it can be confirmed that if $f_3=f_4=0$, the system has two steady states when $\Delta_{13}=\Delta_{14}$. The dark resonance (RFDR) $\rho_{00}=0$ is not a unique solution. Another one having $\rho_{00}\neq 0$ and the arbitrary linear combination of them can appear in the system, depending on the initial condition. In fact, the line profile of the fluorescence to be observed is Fig. 3(a) (the initial condition $\rho_{22}=1$ and all the other components of the density matrix are zero) and the dark resonance is completely missing in it. [But the solution of $\rho_{00}=0$, $\rho_{33}=\Omega_{13}^2/(\Omega_{13}^2+\Omega_{14}^2)$, $\rho_{44}=\Omega_{14}^2/(\Omega_{13}^2+\Omega_{14}^2)$, and $\tilde{\rho}_{34}=-\Omega_{13}\Omega_{14}/(\Omega_{13}^2+\Omega_{14}^2)$, with all the other components being zero, does satisfy Eq. (1) for the steady state. In addition, this solution is of course stable because Eq. (1) is a linear, simultaneous differential equation and its steady-state solution is always stable.] On the other hand, if $f_3\neq 0$ or $f_4\neq 0$, the system does exhibit dark reso-

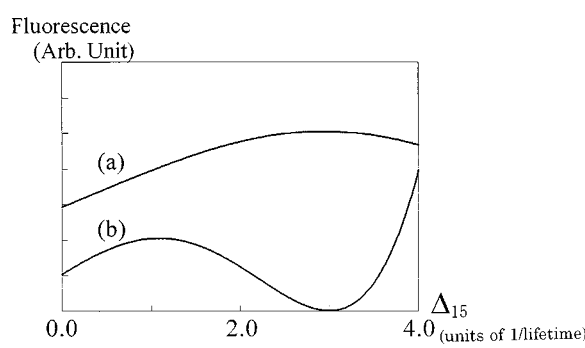

FIG. 3. Fluorescence intensity of the system in Fig. 2 when the detuning frequency of the rf field between $|1\rangle$ and $|5\rangle$ is scanned. The intensity is appropriately normalized. (a) The case where fluorescence from $|0\rangle$ to $|4\rangle$ and $|5\rangle$ in Fig. 2 is forbidden, while that to $|1\rangle$ and $|2\rangle$ is allowed, that is, $f_1=f_2=\frac{1}{2}$, $f_3=f_4=0$. RFDR at $\Delta_{15}=3$ is missing. (b) The case where fluorescence from the excited state falls to all the ground states: $f_1=f_2=f_3=f_4=\frac{1}{4}$. Fluorescence completely vanishes when $\Delta_{15}=3$. The common conditions of the calculation are $T=\frac{1}{2}$, $T_2=1$, $\Omega_{13}=10$, $\Omega_{23}=10$, $\Omega_{14}=10$, $\Omega_{15}=10$, $\Delta_{13}=1$, $\Delta_{23}=2$, and $\Delta_{14}=3$.

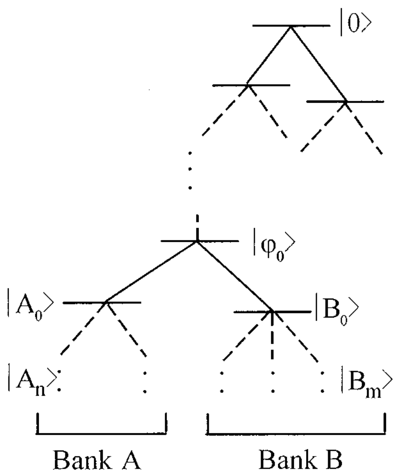

FIG. 4. Example of a level configuration in which RFDR may occur. Fluorescence from $|0\rangle$ to $|A\rangle$'s or $|B\rangle$'s is necessary for RFDR to appear completely. (Note that fluorescence to $|\varphi_0\rangle$ has nothing to do with the existence of RFDR.)

nance [Fig. 3(b)]. Below, we discuss this drastic difference and find the sufficient and practical condition to avoid missing dark lines.

The uniqueness of the steady state is closely related to the condition of RFDR. The number of the steady states for Eq. (1) coincides with the degree of the degeneracy of the solution $\lambda=0$ in the characteristic equation of the Hamiltonian H, $|H-\lambda E|=0$, where E is the identity matrix. It is well known that $|H-\lambda E|$ always has a factor $(1-\Sigma f_n)$ and if the system is a closed system [which means no incoherent drain to or from the other system(s), i.e., $(1-\Sigma f_n)=0$], it has at least one steady-state solution. Therefore a certain special condition for H is required for $|H-\lambda E|=0$ to have another solution $\lambda=0$. In other words, $|H|$ has to have a zero factor other than $(1-\Sigma f_n)$. In this paper, we focus our attention on those cases

$$|H|=\left(1-\sum f_n\right)|H_S|(\text{const}), \tag{22}$$

where H_S is defined as in Eq. (14). The dark resonance condition $|H_S|=0$ immediately leads to the double valuedness of the steady state in this case. This condition is not a special or critical one for the practical level configuration. (See the last example in Sec. IV.) In Appendix C we prove that, when fluorescence is forbidden to fall on all energy levels of the subgroups in Fig. 4. i.e., $|A\rangle$ and $|B\rangle$, the determinant $|H|$ can be factorized as Eq. (22). The ODR condition $|H_S|=0$ introduces another zero pole in $|H|$ besides $(1-\Sigma f_n)=0$. It follows that another steady state with $\rho_{00}\neq 0$ can suppress or hide the dark steady state. In contrast, if fluorescence falls on at least one of the levels in such a group, this critical condition Eq. (22) is broken and the system converges into the unique steady state of dark resonance even when $|H_S|=0$ (Fig. 3).

This phenomenon shows the role of the incoherent decay in dark resonance. If no fluorescence to the levels in the relevant tree is allowed, the mechanism of optical pumping for RFDR does not work and the population in the bright state can exist all the time. This fact has a serious effect on the validity of conventional dark resonance spectroscopy (DRS). It is well known that in DRS [25,26] the decay time in the ground states can be determined by the depth of the dip of dark resonance. In contrast, the critical level configuration mentioned above is an exception to which the conventional interpretation of the depth of the dip is applied. (However, ODR is free from this exception. The cases to which one must pay attention are only those of RFDR.)

In summary, the condition for dark resonance in any tree-like system is obtained as a zero-determinant form of the submatrix of the Hamiltonian. If all fluorescences are forbidden to fall on all the levels included in a subtree, the system has another steady state and ODR can be missing. We verify that at least one level on which fluorescence is allowed is necessary to avoid this critical condition.

IV. DISCUSSION

One of the important cases of ODR for realistic experiments is a hydrogen-like atom, e.g., alkali metal, with a nuclei spin $I\neq 0$. The ground state of it, $J=1/2$, splits into two hyperfine structures $F=I+1/2, I-1/2$. Assume that such an atom is located on a static magnetic field B_{static} along the z axis to yield Zeeman splitting Δ in first order and that two of the Zeeman components with the highest m_F in each hyperfine structure share a common excited level by the laser fields. Figure 5(a) represents the case for $I=\frac{3}{2}$. If a rf magnetic field that is linearly polarized along the x axis, with B_{rf} for its amplitude, is applied to mix the Zeeman components within the same F, the dark line splits itself into several branches. (Note that g factors for the states $F=I+1/2, I-1/2$ accidentally have the same absolute value and opposite signs because of $J=1/2$ and that one rf magnetic field can be simultaneously resonant for both.) For this case we can associate groups P and Q discussed in Secs. II and III with

$$\{|F=I+\tfrac{1}{2},\ m_F=I+\tfrac{1}{2}\rangle,\dots,|F=I+\tfrac{1}{2},\ m_F=-(I+\tfrac{1}{2})\rangle\}$$

and

$$\{|F=I-\tfrac{1}{2},\ m_F=I-\tfrac{1}{2}\rangle,\dots,|F=I-\tfrac{1}{2},\ m_F=-(I-\tfrac{1}{2})\rangle\},$$

respectively. The total number of the dark lines can be obtained immediately from Eq. (17),

$$n_P n_Q=4I(I+1). \tag{23}$$

(See Sec. III A.)

The frequencies at which these dark lines appear are analogous with those of Zeeman splitting lines of a system consisting of two angular momenta $J_1=I-1/2$ and $J_2=I+1/2$ in a static magnetic field $\mathbf{B}=(B_{\text{rf}}, 0, B_{\text{static}})$. In fact, the off-diagonal part of H_{DR} mixes all the adjacent levels within each of J_1 and J_2 and the matrix elements are the same as those for the Zeeman interaction, that is, the proper Clebsh-Gordan coefficients multiplied by the rotational components of the amplitude of the rf magnetic field $B_+=B_-=B_{\text{rf}}/2$ (of course, because the time dependence of B_{rf} has been already removed and the effect of $\mathbf{B}$ seems static). On the other hand, each component of the diagonal part is the direct sum of the detuning frequency $\Sigma\Delta$ in Eq. (5f), that is, $\{0,\Delta,2\Delta,\dots,(2I+1)\Delta\}\oplus\{0,\Delta,2\Delta,\dots,2I\Delta\}$. There-

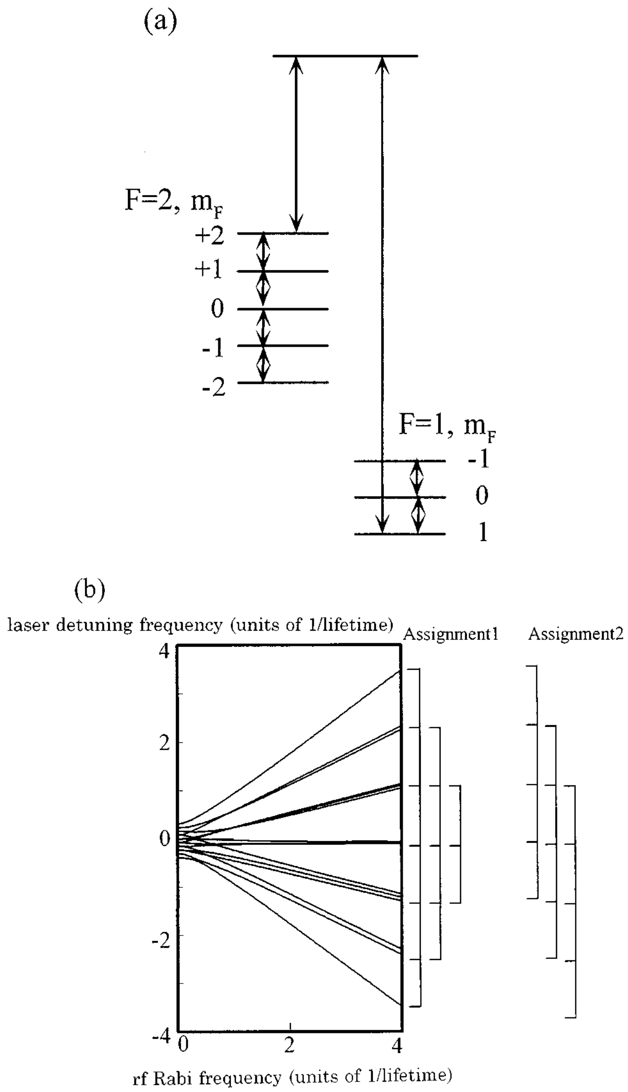

FIG. 5. Potassium atom with two laser fields and one rf magnetic field. (a) System configuration. Since the g factors for the two hyperfine structures have the same absolute value, the rf magnetic field can be resonant with all the Zeeman spacings simultaneously. (b) Calculated splitting pattern of dark lines in the LIF spectrum when the rf power is increased. The parameters are determined for the potassium atom $I=\frac{3}{2}$ in a static magnetic field of 3 G. The frequency of the rf field is 4 MHz.

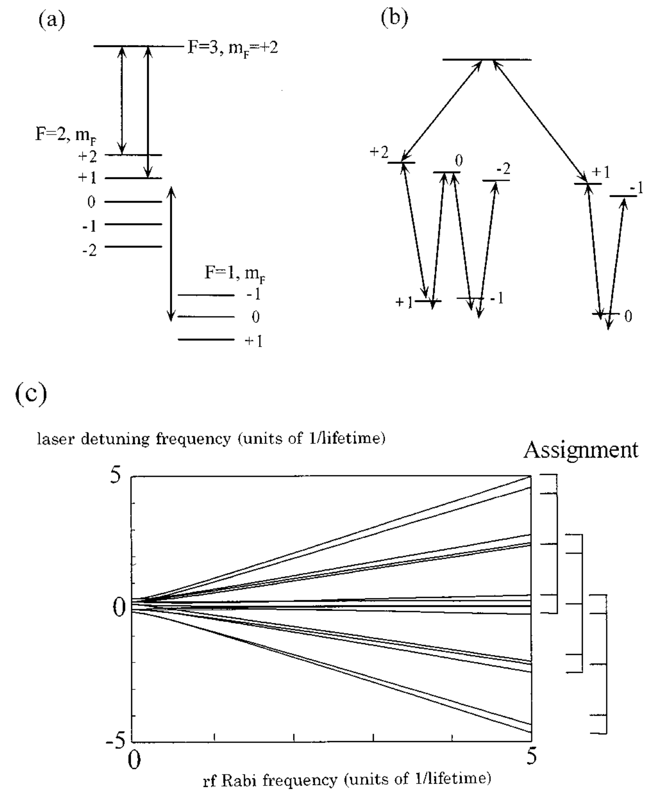

FIG. 6. Potassium atom with two laser fields and one rf magnetic field. (a) System configuration. (b) Taking into account the possible transitions, (a) can be rewritten as this complicated treelike system. One rf field can be resonant with the hyperfine splittings with different detuning frequencies for each transition. (c) Calculated splitting pattern of dark lines in the LIF spectrum when the rf power is increased. The parameters are determined for the potassium atom $I=\frac{3}{2}$ in a static magnetic field of 1.5 G. The frequency of the rf field is 461.7 MHz.

fore the components in the diagonal part correspond to the (first-order) Zeeman energy of a system including two decoupled angular momentum J_1,J_2 in a static magnetic field $\mathbf{B}=(0,0,B_{\text{static}})$ for their decoupled base set $|m_{J_1}\rangle|m_{J_2}\rangle$. For example, the dark resonance for the system $I=\frac{3}{2}$ in Fig. 5(a) is graphically shown in Fig. 5(b). (The parameters are determined for potassium atom $I=\frac{3}{2}$ in static magnetic field of 3 G and the rf magnetic field at 4 MHz.) The Rabi frequency of laser field does not affect the line position, as we already mentioned. The total number of the dark lines is 15 from Eq. (23). When B_{rf} is sufficiently large, the system is separated into several branches. This can be explained by the analogy of the system $\mathbf{J}=\mathbf{J}_1+\mathbf{J}_2$, which will split in a large field into branches corresponding to $J=J_1+J_2,J_1+J_2-1,\ldots,|J_1-J_2|$ and their Zeeman components. For the case of Fig. 5(a) where $J_1=1$ and $J_2=2$, there are three structures, categorized by the total $J=3,2,1$ and their Zeeman components m_J, whose numbers of lines are 7,5,3, respectively (assignment 1). Therefore these dark lines can be assigned by the total J and m_J. Another direct assignment is also possible: the five Zeeman components of $J_2=2$ are offset by the three values of the Zeeman energy of $J_1=1$ (assignment 2). This interpretation is based on the fact that the resonance condition of ODR is not affected by the optical field that would mingle the two angular momenta and that the two systems are isolated from each other. When $B_{\text{rf}}\approx 0$, the dark lines are separated by $\Sigma\Delta$. Because of the nonlinear Zeeman effect by B_{static}, the splitting pattern is rather complicated in the condition above. (When B_{static} is small enough for Δ to be evaluated in the linear region, the pattern resembles that in the strong B_{rf}.)

Another important case is when a rf magnetic field is applied to mix the two hyperfine structures with each other [Fig. 6(a)]. This case can be rewritten as Fig. 6(b), taking into account the possible transitions. The matrix elements of the off-diagonal part in H_{DR} for this case does not coincide with those for Zeeman splitting because the relevant Clebsch-Gordan coefficients are calculated between the two different hyperfine structures. However, since the ladderlike form of the network of the system resembles the previous

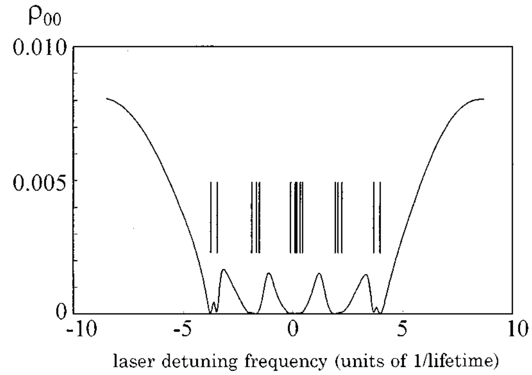

FIG. 7. LIF spectrum obtained from numerical integration of the Bloch equation for the case of Fig. 6. The resonant Rabi frequencies of the rf field and laser fields are 4 and 60 multiplied by the appropriate Clebsch-Gordan coefficients, respectively. The straight lines indicate the positions of dark lines derived from the analytic expression of the present theory [Fig. 6(c)]. (The rf power is equal to 4.)

example, the splitting pattern can be assigned similarly. [See Fig. 6(c). The parameters are determined for the potassium atom in a static magnetic field of 1.5 G and a rf magnetic field at 461.7 MHz.] The inhomogeneous frequency spacings are due to those transition moments not corresponding to the Zeeman interaction. The dark lines in the weak field appear at the detuning frequencies $\Sigma\Delta$, which are more complicated than the case in Fig. 5(a) because of the different signs of the g factors in each hyperfine structure. The splitting pattern also can be assigned to five energy levels offset by three, which corresponds to the number of ladderlike energy levels relevant to ODR.

Figure 7 shows a comparison between the results of the numerical integration of Eq. (1) and the dark line position expected from the analytic expression of our theory. Not all dark lines are resolved due to the power broadening. One can see that a fine structure of dark lines does exist in the line shape obtained by simulation. The degree of degeneracy of the dark lines qualitatively explains the inhomogeneous line widths of the (unresolved) dark lines. Then it can be said that our simple theory also gives information for the line shape of LIF without a long calculation or simulation.

Finally, we consider the case of Fig. 8, which is quite similar to Fig. 6(a). We do not show the splitting pattern of it, which is almost the same as Fig. 6(c). [In fact, the off-diagonal part of H_{DR} is the same as that of Fig. 6(a). A few elements in the diagonal part are different, which contribute only to the weak-field region.] However, this level configuration yields RFDR, which we discussed in Sec. III B. Now we define Δ as the linear Zeeman splitting frequency. (We neglect the nonlinear component of it in order to make the calculation brief.) The level configuration of $|F,m_F\rangle=\{|2,+2\rangle,|1,+1\rangle,|2,0\rangle,|1,-1\rangle,|2,-2\rangle\}$ is rewritten as the same graph as Fig. 1(b) if we set $\Delta_{\mathrm{laser1}}=-(\Delta+\delta)$, $\Delta_{\mathrm{laser2}}=\Delta-\delta$, $\Delta_{\mathrm{rf1}}=\Delta-\delta$, and $\Delta_{\mathrm{rf2}}=3\Delta-\delta$, where δ is the detuning frequency of the rf field from $0\leftrightarrow 0$ transition. Taking into account the Clebsch-Gordan coefficients $\pm\sqrt{1/5}$ of the rf transitions, the condition for RFDR can be obtained from Eq. (15) as

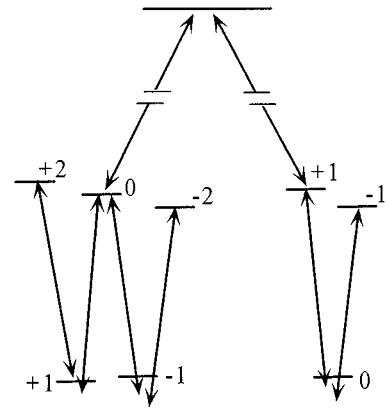

FIG. 8. Level configuration of the potassium atom with two laser fields and one rf field in which RFDR may occur. The left bank of ground states is graphically equivalent to a symmetric five-level case [24]. Besides the dark lines of ODR, those of RFDR can appear when the rf frequency is scanned.

$$\begin{vmatrix} \Delta-\delta & -\dfrac{B_{\mathrm{rf}}}{2\sqrt{5}} & -\dfrac{B_{\mathrm{rf}}}{2\sqrt{5}} & 0 \\ -\dfrac{B_{\mathrm{rf}}}{2\sqrt{5}} & -2\Delta & 0 & -\dfrac{B_{\mathrm{rf}}}{2\sqrt{5}} \\ -\dfrac{B_{\mathrm{rf}}}{2\sqrt{5}} & 0 & 4\Delta & -\dfrac{B_{\mathrm{rf}}}{2\sqrt{5}} \\ 0 & -\dfrac{B_{\mathrm{rf}}}{2\sqrt{5}} & -\dfrac{B_{\mathrm{rf}}}{2\sqrt{5}} & \Delta+\delta \end{vmatrix}=0; \tag{24}$$

therefore

$$\delta=\pm\left(\Delta^2+\frac{B_{\mathrm{rf}}^2}{40}\right)^{1/2}. \tag{25}$$

These new dark resonances would appear when the frequency of the rf field is scanned. However, recall that RFDR is allowed only when at least one of the levels in the subsystem accepts fluorescence from the excited level (Sec. III C). If the optically excited level in Fig. 8 is $|F=3, m_F=0\rangle$, fluorescence is forbidden to fall on all the levels in the subset $|F,m_F\rangle=\{|2,+2\rangle,|1,+1\rangle,|1,-1\rangle,|2,-2\rangle\}$ (the left bank of the ground state in the figure), except $|2,0\rangle$, the top level of the group. In this case, RFDR is incomplete from the discussion in Sec. III C. Figure 9(a) is the result of the numerical calculation of the line shape. (On our assumption of negligible incoherent decay in ground states, no fluorescence is observed in ODR.) Otherwise, if the excited state is one of the possible states, i.e., $|F,m_F\rangle=|3,+1\rangle,|2,0\rangle,|2,+1\rangle,|1,0\rangle,|1,+1\rangle$, fluorescence to at least one level in the left bank is allowed and complete RFDR occurs. For example, when the excited state is $|F,m_F\rangle=|3,+1\rangle$, the fluorescence through the allowed transition $|3,+1\rangle\rightarrow|2,+2\rangle$ enables RFDR to appear [Fig. 9(b)].

In summary, the frequencies of laser detuning at which dark lines appear are determined by the eigenvalues of a Hermitian H_{DR}, which can be interpreted as a Hamiltonian of a system including two isolated systems under the decoupled basis set. This interpretation enables one to predict the position of dark resonance intuitively or to assign the ob-

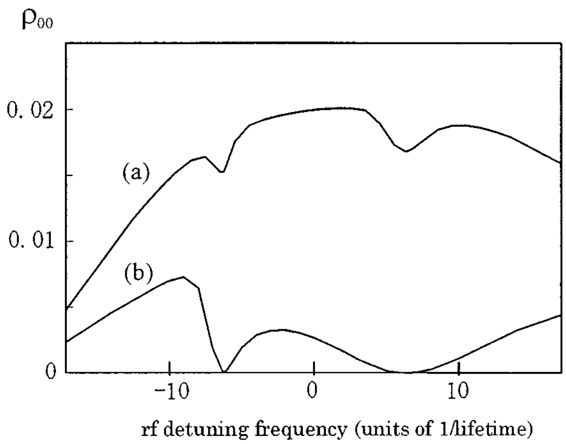

FIG. 9. LIF spectra obtained from numerical integration of Bloch equation for the case of Fig. 8. (a) When the excited state is $|F=3, m_F=0\rangle$, fluorescence to the left bank of ground states in Fig. 8 is forbidden by the selection rule. (Fluorescence to $|F=2, m_F=0\rangle$ has nothing to do with the existence of RFDR.) The dark resonance is incomplete due to the double valuedness of the steady state. (b) The case where the excited state is $|F=3, m_F=+1\rangle$. Fluorescence completely vanishes on RFDR ($\Delta_{\rm rf}=\pm 6.3$). The common conditions are $T=\frac{1}{2}$ and $T_2=1$. The resonant Rabi frequencies of the rf field and laser fields are 40 and 60 multiplied by the appropriate Clebsch-Gordan coefficients, respectively. The detuning frequencies of the laser fields are 1 for the left bank and 0 for the right bank. The Zeeman spacing frequency is 0.1.

served complicated resonance pattern of dark lines with the relevant energy levels and transitions. Because dark resonance spectroscopy is a kind of Raman spectroscopy, it possesses the same advantageous properties of them such as sub-natural resonance width. We showed that attention must be paid to some critical level configurations in which ODR can be hidden. It may be interesting that fluorescence condition affects the existence of ODR, which is not the usual case of the Raman process.

V. CONCLUSION

We obtain the analytic expression of the condition for optical dark resonance in multilevel and multiradiation system with an arbitrary treelike configuration under the rotating-wave approximation. The splitting pattern of the dark lines is found to be analogous to the energy shift of the composite system of two systems. These dark lines are expected to be useful for the analysis of complicated multilevel systems. Our theory can reveal the fine structure of dark lines with power broadening. Furthermore, it is possible that the complicated splitting pattern of the dark lines in the observed fluorescence spectrum can be assigned to the relevant level configuration and transitions. The advantage of the present theory, that is, the general formalism for the arbitrary multiradiation and multilevel system without perturbation treatment, is expected to be promising for the analytic studies of multiphoton effects [27,28].

APPENDIX A: PROOF OF EQ. (6) IN EQ. (5)

A brief and elementary proof is as follows. Assume that a level $p_A \in P$ is the tip of a tree. Because p_A has only one transition with, e.g., p_B, the relation equation (5a) for $\tilde{\rho}_{p_Ap_A}$ has only one term and implies

$$\begin{aligned}\tilde{\rho}_{p_Ap_A} &= \Omega_{p_Ap_B}\tilde{\rho}_{p_Bp_A} - \Omega_{p_Bp_A}\tilde{\rho}_{p_Ap_B}\\ &= 2i\ \mathrm{Im}(\Omega_{p_Ap_B}\tilde{\rho}_{p_Bp_A}) = 0. \end{aligned} \tag{A1}$$

This relation is satisfied for all the tips in the network and therefore they are cut down from the tree, assuming the imaginary parts of the relevant ρ's to be zero. This procedure produces new tips in the network, which can be reduced using (A1). For example,

$$\begin{aligned}&(\Omega_{p_Bp_A}\tilde{\rho}_{p_Ap_B} - \Omega_{p_Ap_B}\tilde{\rho}_{p_Bp_A}) + (\Omega_{p_Bp_C}\tilde{\rho}_{p_Cp_B} - \Omega_{p_Cp_B}\tilde{\rho}_{p_Bp_C})\\ &= 2i\ \mathrm{Im}(\Omega_{p_Bp_C}\tilde{\rho}_{p_Cp_B}) = 0. \end{aligned} \tag{A2}$$

All edges can be cut in these processes when the network is treelike.

APPENDIX B: EXPANSION OF $|H_{\rm ODR}|$ WITH THE NONZERO FACTOR

Assume that a level $p_A \in P$ is the tip of a tree and p_B is the only energy level having a transition with p_A. The equations including p_A are only Eqs. (5b) and (5c),

$$\begin{aligned}&-\Omega_{p_Bp_A}\tilde{\rho}_{p_Ap_B} + \{-(\pm\Delta_{p_Ap_B})\tilde{\rho}_{p_Ap_B} + \Omega_{p_Bp_A}\tilde{\rho}_{p_Bp_B}\\ &-\sum_{X\approx B}\Omega_{p_Xp_B}\tilde{\rho}_{p_Ap_X}\} = 0. \end{aligned} \tag{B1}$$

The fact that $\tilde{\rho}_{p_Ap_A}$ is included only in this equation enables $|H_{\rm ODR}|$ to be expanded as $(\pm\Omega_{p_Bp_A})|H'_{\rm ODR}|$, eliminating the column for $\tilde{\rho}_{p_Ap_A}$ and the row for $\tilde{\rho}_{p_Bp_A}$. (The choice of the double sign depends on the position of the key in the matrix, but it is not important to our proof.) Removing all the tips like p_A brings about the new tips like p_B. Though $\tilde{\rho}_{p_Bp_B}$ can be included in two (or more) equations in the original equations, (5b) or (5c), all of them except one have already been deleted by the previous procedure. One can always expand $|H'_{\rm ODR}|$ using the key $\tilde{\rho}_{p_Bp_B}$ and proceed until all the tips $\tilde{\rho}_{p_ip_j}$ have been removed. No tip remains after the same procedure for the group Q. (The last tip $\tilde{\rho}_{00}$ has already been eliminated by the assumption of ODR.) Eventually the rows of Eqs. (5c) and (5d) and the column for $\tilde{\rho}_{nn}$ are eliminated from the condition $|H_{\rm ODR}|=0$ with nonzero factors.

APPENDIX C: PROOF OF EQ. (25) IN THE CRITICAL CASE

When we call the two banks A and B (Fig. 4), the condition of RFDR can be written as

$$|H_S| = \begin{vmatrix} \Delta_{A_1B_1} & \Omega_{A_1B_2} & \cdots \\ \Omega_{A_1B_2} & \Delta_{A_2B_2} & \\ \vdots & & \ddots \end{vmatrix} = 0 \tag{C1}$$

from Eq. (12). We define the energy level φ_0 as the stem on which A and B are hooked. We now return to the Schrödinger equation (1), in which the optical population and coherence have not been set to zero. If we focus our attention on the subtrees A, B, and φ_0, (1) can be rewritten, using H_S in (C1),

$$\frac{d}{dt}\begin{pmatrix} \rho_{A_iB_i} \\ \rho_{A_iA_i} \text{ or } \rho_{B_iB_i} \\ \text{otherwise} \end{pmatrix} = \begin{pmatrix} H_S & 0 & M & 0 \\ 0 & L & N & if_{A_i} \text{ or } if_{B_i} \\ \cdots & \cdots & \cdots & \cdots \end{pmatrix} \times \begin{pmatrix} \rho_{A_iB_i} \\ \rho_{A_iA_j} \text{ or } \rho_{B_iB_j} \\ \rho_{\varphi_0A_0} \text{ or } \rho_{\varphi_0B_0} \\ \text{otherwise} \end{pmatrix}. \tag{C2}$$

The components $\rho_{A_iA_j}$, $\rho_{B_iB_j}$, $\rho_{\varphi_0A_0}$, and $\rho_{\varphi_0B_0}$ on the right-hand side are those coherences created directly by the applied radiation, that is, $\Omega_{A_iA_j}\neq 0$, $\Omega_{B_iB_j}\neq 0$, $\Omega_{\varphi_0A_0}\neq 0$, and $\Omega_{\varphi_0B_0}\neq 0$. On the other hand, those on the left-hand side are the coherences across the bridge and the population.

Now we assume that all if_{A_i} and if_{B_i} are zero and get

$$\frac{d}{dt}\begin{pmatrix} \rho_{A_iB_i} \\ \rho_{A_iA_i} \text{ or } \rho_{B_iB_i} \\ \text{otherwise} \end{pmatrix} = \begin{pmatrix} H_S & 0 & M & 0 \\ 0 & L & N & 0 \\ \cdots & \cdots & \cdots & \cdots \end{pmatrix} \times \begin{pmatrix} \rho_{A_iB_i} \\ \rho_{A_iA_j} \text{ or } \rho_{B_iB_j} \\ \rho_{\varphi_0A_0} \text{ or } \rho_{\varphi_0B_0} \\ \text{otherwise} \end{pmatrix}. \tag{C3}$$

What we show below is that the expansion of the determinant $|H|$ with the key of the submatrix M yields zero and that $|H|$ can be expanded as

$$|H| = \begin{vmatrix} H_S & 0 & M & 0 \\ 0 & L & N & 0 \\ \cdots & \cdots & \cdots & \cdots \end{vmatrix} = |H_S| \begin{vmatrix} L & N & 0 \\ \cdots & \cdots & \cdots \end{vmatrix}. \tag{C4}$$

Within groups A and B, to which no fluorescence is allowed to fall because of, e.g., the selection rule, Eq. (6) is satisfied, that is,

$$\mathrm{Im}(\tilde{\rho}_{A_iA_j}) = 0 \quad \text{when} \quad \Omega_{A_iA_j}\neq 0, \tag{C5}$$

$$\mathrm{Im}(\tilde{\rho}_{B_iB_j}) = 0 \quad \text{when} \quad \Omega_{B_iB_j}\neq 0. \tag{C6}$$

Note that those substitutions are valid for subsystems A and B because we assume that no decay terms are included in groups A and B. In contrast, the proof of Eq. (6) in Sec. II is based on the condition of dark resonance, which forces no fluorescence to appear. (This is equivalent to the assumption of no decay terms.) From (C5) and (C6), the coherences directly driven by the applied radiation, that is, $\rho_{A_iA_j}, \rho_{B_iB_j}, \rho_{\varphi_0A_0}$, and their complex conjugates can be replaced with their real parts.

The submatrix M has only four nonzero components. In fact, all the time differentials of the coherence across the bridge $d\tilde{\rho}_{A_iB_j}/dt$ are made only by $\rho_{A_XB_Y}$, except $\rho_{\varphi_0A_0}$ and $\rho_{\varphi_0B_0}$, that is,

$$d\tilde{\rho}_{A_0B_0}/dt = i\Omega_{\varphi_0A_0}\,\mathrm{Re}(\tilde{\rho}_{\varphi_0B_0}) - i\Omega_{\varphi_0B_0}\,\mathrm{Re}(\tilde{\rho}_{\varphi_0A_0}) + \mathscr{F}(\tilde{\rho}_{A_iB_j}), \tag{C7}$$

$$d\tilde{\rho}_{B_0A_0}/dt = i\Omega_{\varphi_0A_0}\,\mathrm{Re}(\tilde{\rho}_{\varphi_0B_0}) - i\Omega_{\varphi_0B_0}\,\mathrm{Re}(\tilde{\rho}_{\varphi_0A_0}) + \mathscr{F}(\tilde{\rho}_{A_iB_j}), \tag{C8}$$

where $\mathscr{F}$ is a function of $\tilde{\rho}_{A_iB_j}$. Therefore M is written as

$$M = \begin{pmatrix} i\Omega_{\varphi_0A_0} & -i\Omega_{\varphi_0B_0} & 0 & \cdots \\ i\Omega_{\varphi_0A_0} & -i\Omega_{\varphi_0B_0} & 0 & \cdots \\ 0 & 0 & 0 & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}. \tag{C9}$$

The submatrix N has only two nonzero components for $d\tilde{\rho}_{A_0A_0}/dt$ and $d\tilde{\rho}_{B_0B_0}/dt$ through the relations

$$d\tilde{\rho}_{A_0A_0}/dt = 2i\Omega_{\varphi_0A_0}\,\mathrm{Re}(\tilde{\rho}_{\varphi_0A_0}) + \mathscr{F}(\tilde{\rho}_{A_iA_j}), \tag{C10}$$

$$d\tilde{\rho}_{B_0B_0}/dt = 2i\Omega_{\varphi_0B_0}\,\mathrm{Re}(\tilde{\rho}_{\varphi_0B_0}) + \mathscr{F}(\tilde{\rho}_{B_iB_j}) \tag{C11}$$

and the matrix N is expressed as

$$N = \begin{pmatrix} 2i\Omega_{\varphi_0A_0} & 0 & 0 & \cdots \\ 0 & 2i\Omega_{\varphi_0B_0} & 0 & \cdots \\ 0 & 0 & 0 & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}. \tag{C12}$$

The matrices L and N are expanded in the same manner as in Sec. II with a nonzero factor. In fact, the proof in Appendix B is also valid for any steady state. After the final stage of this expansion, all of the four components of M, that is, the first and second column of (C9), are already eliminated, with the components of (C12) being adopted for the key of expansion. Therefore $|H|$ can be expanded as

$$|H| = \prod (\pm\Omega) \begin{vmatrix} H_S & 0 \\ \cdots & \cdots \end{vmatrix} = C|H_S|. \tag{C13}$$

[1] E. Arimondo and G. Orriols, Lett. Nuovo Cimento **17**, 333 (1976).
[2] Richard G. Brewer and E. L. Hahn, Phys. Rev. A **8**, 464 (1973).
[3] R. M. Whitley and C. R. Stroud, Jr., Phys. Rev. A **14**, 1498 (1976).
[4] G. S. Agarwal and Sudhanshu S. Jha, J. Phys. B **12**, 2655 (1979).
[5] J. N. Elgin, Phys. Lett. **80A**, 140 (1980).
[6] F. T. Hioe and J. H. Eberly, Phys. Rev. Lett. **47**, 838 (1981).
[7] D. A. Cardimona, Phys. Rev. A **41**, 5016 (1990).
[8] G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento **36**, 5 (1976).
[9] R. G. Brewer and E. L. Hahn, Phys. Rev. A **11**, 1641 (1975).
[10] H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (1978).
[11] J. D. Stettler, C. M. Bowden, N. M. Witriol, and J. H. Eberly, Phys. Lett. **73A**, 171 (1979).
[12] P. M. Radmore and P. L. Knight, J. Phys. B **15**, 561 (1982).
[13] M. G. Prentiss, N. P. Bigelow, M. S. Shahriar, and P. R. Hemmer, Opt. Lett. **16**, 1695 (1991).
[14] M. A. Bouchene, A. Debarre, J. C. Keller, J. L. Le Gouet, P. Tchenio, V. Finkelstein, and P. R. Berman, Europhys. Lett. **18**, 409 (1992).
[15] I. Siemers, M. Schubert, R. Blatt, W. Neuhauser, and P. E. Toschek, Europhys. Lett. **18**, 139 (1992).
[16] J. Dalibard, S. Reynaud, and C. Cohen-Tannoudji, J. Phys. B **17**, 4577 (1984).
[17] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, Phys. Rev. Lett. **61**, 826 (1988).
[18] P. D. Lett, R. N. Watts, C. I. Westbrook, W. D. Phillips, P. L. Gould, and H. J. Metcalf, Phys. Rev. Lett. **61**, 169 (1988).
[19] E. Bonderup and K. Molmer J. Opt. Soc. Am. B **6**, 2125 (1989).
[20] P. J. Ungar, D. S. Weiss, E. Riis, and Steven Chu, J. Opt. Soc. Am. B **6**, 2058 (1989).
[21] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, J. Opt. Soc. Am. B **6**, 2122 (1989).
[22] Y. Castin, H. Wallis, and J. Dalibard, J. Opt. Soc. Am. B **6**, 2046 (1989).
[23] S. Chang, B. M. Garraway, and V. G. Minogin, Opt. Commun. **77**, 19 (1990).
[24] H. Kanokogi and K. Sakurai, Phys. Rev. A **57**, 2650 (1996).
[25] S. Saikan, N. Hashimoto, T. Kushida, and K. Namba, J. Chem. Phys. **82**, 5409 (1985).
[26] Yehiam Prior, A. R. Bogdan, M. Dagenais, and N. Bloembergen, Phys. Rev. Lett. **46**, 111 (1981).
[27] Yehiam Prior and Abraham Ben-Reuven, Phys. Rev. A **33**, 2362 (1986).
[28] Benjamin Fain, A. Boeglin, and S. H. Lin, J. Chem. Phys. **88**, 7559 (1988).