Resonance Fluorescence Revisited*

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We re-examine resonance fluorescence showing that a mathematical advantage is realized when using semiclassical dressed states. In particular the present dressed-state treatment yields exact results with a fraction of the effort required when working in the usual bare-state basis.

I. Introduction

Resonance fluorescence from a two-level atom driven by a monochromatic light field, is one of the fundamental problems in quantum optics, (see Fig. 1). It was theoretically predicted[1] and experimentally observed[2] in the early 70's. Studies in this area have been extended to include modification of the spontaneous emission in an electromagnetic resonator[3] and subnatural line narrowing in the fluorescence from a driven three-level atom[4].

In the present paper we show that the mathematics of the usual analysis can be simplified by going to a dressed-state basis[5]. In section II, working in the semiclassical dressed-state basis, we show that we can reduce the coupling between the equations of motion of the density matrix elements, which substantially reduces the calculational effort. In Appendix A, we present a detailed solution of the matrix equations of motion. Section III deals with the two-time correlation function and in section IV, we analyze the spectrum both in the weak and strong driving field limits. Furthermore, in Appendix B, we review the main features of resonance fluorescence in the strong driving field limit by applying the secular approximation. The spirit of the present paper is meant to be tutorial.

II. Description in the dressed-state basis

Consider a two-level atom (upper level $|a\rangle$ and lower level $|b\rangle$) interacting with a radiation field of frequency v, then the total Hamiltonian in the rotating wave approximation can be written as

$$\mathcal{H} = \hbar\omega_a|a\rangle\langle a| + \hbar\omega_b|b\rangle\langle b| + \mathcal{H}' \tag{1}$$

where

$$\mathcal{H}' = \frac{\hbar\Omega}{2}(|a\rangle\langle b|e^{-i\nu t} + |b\rangle\langle a|e^{i\nu t}) . \tag{2}$$

The Rabi frequency R is defined as

$$\hbar\Omega = \wp\mathcal{E} \tag{3}$$

and assumed to be real, where p is the dipole matrix element between the upper level and the lower level and $\mathcal{E}$ is the electric field amplitude of the driving radiation field. For the case of resonance (v $= \omega_a - \omega_b$) the interaction picture Hamiltonian is now given by

$$\mathcal{V} = \frac{\hbar\Omega}{2}(|a\rangle\langle b| + |b\rangle\langle a|) . \tag{4}$$

Including the relaxation between the atomic levels, we describe the given system by a master equation,

$$\dot{\rho} = -\frac{i}{\hbar}[\mathcal{V},\rho] + \mathcal{L}\rho , \tag{5}$$

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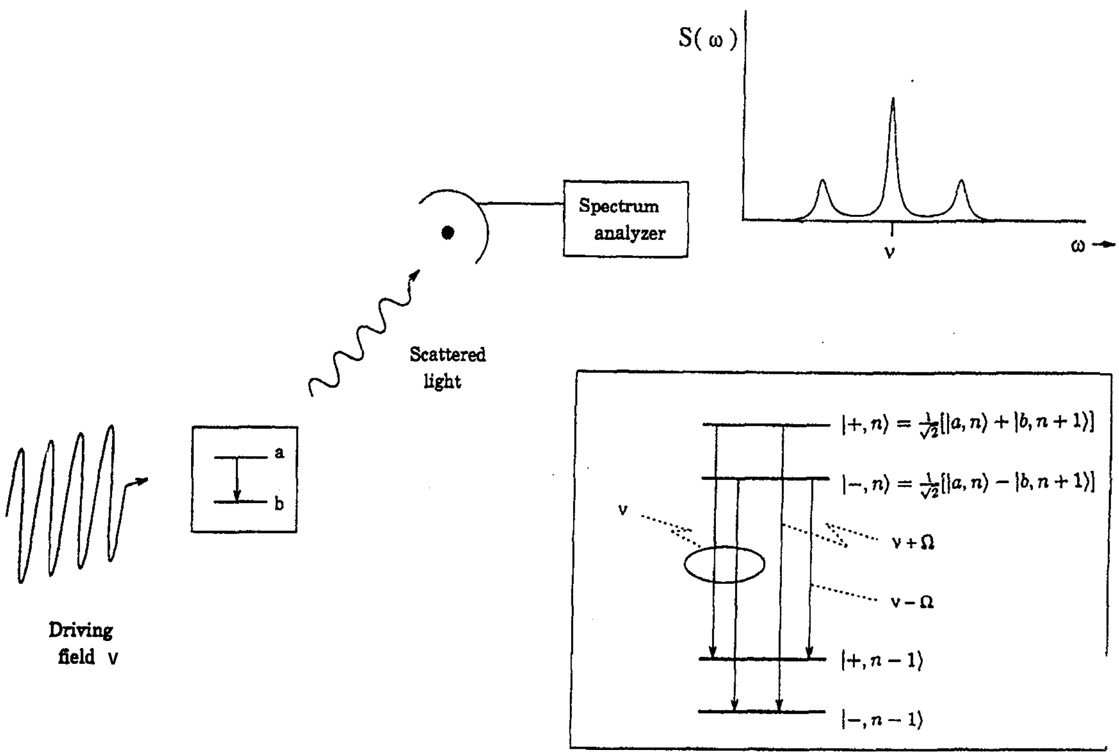

FIG. 1

Figure 1. Two-level atom driven by a continuous monochromatic laser field of frequency v. Spectrum of the spontaneous emission shows central peak at v and sidebands at $\nu\pm$ the Rabi frequency, Ω. Physical origin of spectrum can be understood via dressed states, see insert lower righthand corner.

where ρ is the density matrix of the atom and the operator L accounts for the relaxation processes. With the atomic operator a $= |b\rangle\langle a|$ the damping part is given by

$$\mathcal{L}\rho := -\frac{\gamma}{2}(\sigma^\dagger\sigma\rho - 2\sigma\rho\sigma^\dagger + \rho\sigma^\dagger\sigma) \,, \qquad (6)$$

where γ is the radiative decay rate from the upper level to the lower level.

Here we can diagonalize the interaction Hamiltonian as

$$\mathcal{V} = \frac{\hbar\Omega}{2}(|+\rangle\langle+| - |-\rangle\langle-|) \,, \qquad (7)$$

where

$$|\pm\rangle = \frac{1}{\sqrt{2}}(|a\rangle \pm |b\rangle) \qquad (8)$$

are the eigenstates of the semiclassical interaction Hamiltonian with eigenvalues $\Omega/2$ and $-\Omega/2$, respectively. With these semiclassical dressed states, the equations of motion of the density matrix elements are obtained from Eqs.(5) and (6) as follows

$$\dot{\rho}_{++} = -\frac{\gamma}{2}\rho_{++} + \frac{\gamma}{4} \,, \qquad (9a)$$

$$\dot{\rho}_{+-} = -\left(\frac{3\gamma}{4} + i\Omega\right)\rho_{+-} - \frac{\gamma}{4}\rho_{-+} - \frac{\gamma}{2} \,, \qquad (9b)$$

$$\dot{\rho}_{-+} = -\left(\frac{3\gamma}{4} - i\Omega\right)\rho_{-+} - \frac{\gamma}{4}\rho_{+-} - \frac{\gamma}{2} \,. \qquad (9c)$$

The advantage of the dressed-state basis is clearly seen in Eq.(9); the equation for ρ_{++} is decoupled from the rest and the problem involves only 2×2 matrices. We remind the reader that the usual (bare-state) treatment involves the three coupled equations,

$$\dot{\rho}_{aa} = -\gamma\rho_{aa} + i\frac{\Omega}{2}(\rho_{ab} - \rho_{ba}) \,, \qquad (10a)$$

$$\dot{\rho}_{ab} = -\frac{\gamma}{2}\rho_{ab} + i\Omega\rho_{aa} - i\frac{\Omega}{2} \,, \qquad (10b)$$

$$\rho_{ba} = -\frac{\gamma}{2}\rho_{ba} + i\Omega\rho_{aa} + i\frac{R}{2} \,, \qquad (10c)$$

The solution of Eq.(9a) is readily obtained as

$$\rho_{++}(t) = e^{-\gamma t/2}\rho_{++}(0) + \frac{1}{2}(1 - e^{-\gamma t/2}) \,. \qquad (11)$$

To solve for ρ_{+-} and ρ_{-+}, it is convenient to rewrite Eqs.(9b,c) in matrix form as

$$\dot{R}(t) = -MR(t) + B \ , \tag{12}$$

where

$$R(t) = \begin{pmatrix} \rho_{+-}(t) \\ \rho_{-+}(t) \end{pmatrix} , \quad M = \begin{pmatrix} 3\gamma/4 + i\Omega & \gamma/4 \\ \gamma/4 & 3\gamma/4 - i\Omega \end{pmatrix} , \quad B = -\frac{\gamma}{2}\begin{pmatrix} 1 \\ 1 \end{pmatrix} \tag{13}$$

As shown in Appendix A, we diagonalize the niatrix M as

$$U^{-1}MU = \begin{pmatrix} \lambda_1 & 0 \\ 0 & \lambda_2 \end{pmatrix} \equiv D ,$$

where the eigenvalues of M are given by

$$\lambda_{1,2} = \frac{3\gamma}{4} \pm i\mu \quad , \quad \mu = \sqrt{\Omega^2 - \frac{\gamma^2}{16}} , \tag{15}$$

and

$$U = \begin{pmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{pmatrix} , \text{where} \quad \tan\theta = \frac{i4}{\gamma}(\Omega - \mu) . \tag{16}$$

Eq.(12) can now be written as

$$\dot{R} = -UDU^{-1}R + B , \tag{17}$$

which allows the formal solution

$$R = Ue^{-Dt}u^{-1}R(0) + \int_0^t dt' Ue^{-D(t-t')}U^{-1}B . \tag{18}$$

It follows that

$$R(t) = Ue^{-Dt}U^{-1}R(0) + UD^{-1}(1 - e^{-Dt})U^{-1}B . \tag{19}$$

Using Eqs.(A7) and (A8), we have

$$\begin{aligned} \rho_{+-} &= e^{-3\gamma t/4}\left[\rho_{+-}(0)\left(\cos\mu t - \frac{i\Omega}{\mu}\sin\mu t\right) - \rho_{-+}(0)\frac{\gamma}{4\mu}\sin\mu t\right] \\ &+ e^{-3\gamma t/4}\frac{\gamma}{\gamma^2 + 2\Omega^2}\left[\left(\frac{\gamma}{2} - i\Omega\right)\cos\mu t - \frac{1}{2\mu}\left(2\Omega^2 + \frac{\gamma^2}{4} + \frac{3i\gamma\Omega}{2}\right)\sin\mu t\right] \\ &- \frac{\gamma}{\gamma^2 + 2\Omega^2}\left(\frac{\gamma}{2} - i\Omega\right) , \end{aligned} \tag{20}$$

and $\rho_{-+}(t) = \rho^*_{+-}(t)$.

III. Two-time correlation function

According to the Lax-Onsager regression theorem[6], the two-time correlation function can be determined from a knowledge of single-time expectation values. We are therefore interested in the single-time expectation values of the atomic operators. Since $\langle\sigma(t)\rangle = \rho_{ab}e^{-i\nu t}$ for the resonant case, we may write

$$\langle\sigma(t)\rangle e^{i\nu t} = \rho_{ab} = \frac{1}{2}(2\rho_{++} - 1 - \rho_{+-} + \rho_{-+}) , \tag{21}$$

by using the relation

$$\rho_{ij} = \langle i|\rho|j\rangle$$

$$= \sum_{\alpha,\beta}\langle i|\alpha\rangle\langle\alpha|\rho|\beta\rangle\langle\beta|j\rangle \ , \qquad (22)$$

where $i,j = a,b$ and $\alpha,\beta = +,-$. Now following Eq.(A10) in Appendix B, we have

$$\langle\rho(t)\rangle e^{i\nu t} = a_1(t)Tr(\rho(0)) + a_2(t)Tr(|b\rangle\langle a|\rho(0)) + a_3(t)Tr(|a\rangle\langle b|\rho(0)) + a_4(t)Tr(|a\rangle\langle a|\rho(0)) \ . \qquad (23)$$

Here we have used $\rho_{ij} = Tr(|j\rangle\langle i|\rho)$, $Tr(\rho) = 1$, and the $a_i(t)$ are given in Appendix A. The two-time correlation function is now formally identical to the single-time expectation value $\langle\sigma(t)\rangle$ except that $\rho(0)\sigma^\dagger(0)$ is used instead of $\rho(0)$ in Eq.(23). We find

$$\langle\sigma^\dagger(0)\sigma(0)\rangle e^{i\nu t} = a_1(t)Tr(\rho(0)\sigma^\dagger(0)) + a_2(t)Tr(|b\rangle\langle a|\rho(0)\sigma^\dagger(0)) + a_3(t)Tr(|a\rangle\langle b|\rho(0)\sigma^\dagger(0)) + a_4(t)Tr(|a\rangle\langle a|\rho(0)\sigma^\dagger(0)) \qquad (24)$$

Noting that $\sigma^\dagger = |a\rangle\langle b|$, we have

$$\langle\sigma^\dagger(0)\sigma(t)\rangle e^{i\nu t} = a_1(t)\rho_{ba}(0) + a_2(t)\rho_{aa}(0) \ . \qquad (25)$$

Since we are interested in the steady state, and note that the initial time $t = 0$ is any time after reaching steady state

$$\rho_{ba}(0) = (\rho_{ba})_{ss} = \frac{i\gamma\Omega}{\gamma^2 + 2\Omega^2} \ , \qquad (26a)$$

$$\rho_{aa}(0) = (\rho_{aa})_{ss} = \frac{\Omega^2}{\gamma^2 + 2\Omega^2} \ . \qquad (26b)$$

On substituting Eqs.(26) and (A12) into Eq.(25), we obtain the explicit expression for the two-time correlation function as

$$\langle\sigma^\dagger(0)\sigma(t)\rangle e^{i\nu t} = \frac{\Omega^2}{\gamma^2 + 2\Omega^2}\left[\frac{\gamma^2}{\gamma^2 + 2\Omega^2} + \frac{1}{2}e^{-\gamma t/2} + \frac{1}{4}e^{-3\gamma t/4}\{e^{-i\mu t}(P + iQ) + e^{i\mu t}(P - iQ)\}\right] \ , \qquad (27)$$

where the dimensionless constants P and Q are given by

$$P = \frac{2\Omega^2 - \gamma^2}{2\Omega^2 + y^2} \ , \quad Q = \frac{y}{4\mu}\frac{10\Omega^2 - \gamma^2}{2R^2 + y^2} \ . \qquad (28)$$

IV. Spectrum of the resonance fluorescence

Now the pomer spectrum of the radiation field scattered by a two-level atom driven by an incident field is obtained by taking the Fourier transform of Eq.(27), that is

$$S(\omega) \propto Re\int_0^\infty dt\ e^{i\omega t}\langle\sigma^\dagger(0)\sigma(t)\rangle \ . \qquad (29)$$

Substituting Eq.(27) into Eq.(29) and performing the integration, we obtain the expression for the spectrum

$$S(\omega) \propto \frac{\gamma^2}{\gamma^2+2\Omega^2} 2\pi\delta(\nu-\omega) + \frac{\gamma/2}{(\nu-\omega)+(\gamma/2)^2} + \frac{1}{2}\left[\frac{\alpha_+}{(\nu+\mu-\omega)^2+(3\gamma/4)^2} + \frac{\alpha_-}{(\nu-\mu-\omega)^2+(3\gamma/4)^3}\right] , \quad (30)$$

where

$$\alpha_\pm = \frac{3\gamma}{4}\ \mathrm{Pf}\ (\nu \pm \mu - \omega)Q \ . \quad (31)$$

Let us first consider the weak field limit (Q $\ll \gamma/4$). From Eq.(15) we have $\mu \cong i\gamma/4$ in this limit and

$$P \cong -1 \ , \ Q \cong i \quad \left(\Omega \ll \frac{\gamma}{4}\right) \ . \quad (32)$$

Therefore, in Eq.(27) we have $P+iQ \rightarrow -2, P-iQ \rightarrow 0$, and together with $e^{-i\mu t} \cong e^{\gamma t/4}$ we can easily see that the second and third terms in tlie bracliet cancel each other. Consequently, tlie spectrum in Eq.(30) consists of a delta function centered at tlie field frequency v.

On the other hand, in the strong field limit ($\Omega \gg \gamma/4$, i.e., $\mu \cong \Omega$), the first term in Eq.(30) vanishes and

$$P \cong 1 \ , \ Q \cong 0 \quad \left(Q \gg \frac{\gamma}{4}\right) \ , \quad (33)$$

so that we have

$$\alpha_\pm \rightarrow \frac{3\gamma}{4} \ . \quad (34)$$

Therefore we can again see that there are three peaks at v $-\,\Omega$, v, $\nu+\Omega$, and that the widths of the peaks are $3\gamma/4$, $\gamma/2$, $3\gamma/4$, respectively. The ratio of their heights is 1: 3: 1, so that the integrated intensities of tlie three peaks are in the ratio 1: 2: 1. Finally we recall, **as** is shown in Appendix B, that the strong field limit can be simply obtained upon making a secular approximation.

Appendix **A:** Reciprocal **eigenvectors**

In order to solve the Eq.(12) we seek the eigenstates and eigenvalues of M such that $M\mathbf{v}_i = \lambda_i \mathbf{v}_i$, where $i = 1, 2$; and write U = $(\mathbf{v}_1\ \mathbf{v}_2)$ so that

$$MU = (\lambda_1\mathbf{v}_1\ \lambda_2\mathbf{v}_2) \ . \quad \textbf{(A.1)}$$

Liliewise, we need the inverse of U, which we may find by conventional matrix methods, and can write as a matrix of row vectors

$$U^{-1} = \begin{pmatrix} \check{\mathbf{v}}_1 \\ \check{\mathbf{v}}_2 \end{pmatrix} \ . \quad \textbf{(A.2)}$$

It may help tlie student of modern quantum mechanics to note that $\mathbf{v}_i$ [redacted] $|i\rangle$, and $\check{\mathbf{v}}_j$ [redacted] $\langle j|$.

Note that

$$\begin{aligned} U^{-1}U &= \begin{pmatrix} \check{\mathbf{v}}_1 \\ \check{\mathbf{v}}_2 \end{pmatrix} (\mathrm{v},\ \mathbf{v}_2) \\ &= \begin{pmatrix} \check{\mathbf{v}}_1\cdot\mathbf{v}_1 & \check{\mathbf{v}}_1\cdot\mathbf{v}_2 \\ \check{\mathbf{v}}_2\cdot\mathbf{v}_1 & \check{\mathbf{v}}_2\cdot\mathbf{v}_2 \end{pmatrix} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \end{aligned} \quad \textbf{(A.3)}$$

since

$$\check{\mathbf{v}}_i \,.\, \mathrm{vj} = \delta_{ij} \ . \quad \textbf{(A.4)}$$

On the other hand, the expression for UU^{-1} again shows that

$$\begin{aligned} UU^{-1} &= (\mathbf{v},\ \mathrm{v},) \begin{pmatrix} \check{\mathbf{v}}_1 \\ \check{\mathbf{v}}_2 \end{pmatrix} \\ &= \begin{pmatrix} \breve{e}_1\cdot(\mathbf{v}_1\check{\mathbf{v}}_1+\mathbf{v}_2\check{\mathbf{v}}_2)\cdot e_1 & \breve{e}_1\cdot(\mathbf{v}_1\check{\mathbf{v}}_1+\mathbf{v}_2\check{\mathbf{v}}_2)\cdot e_2 \\ \breve{e}_2\cdot(\mathbf{v}_1\check{\mathbf{v}}_1+\mathbf{v}_2\check{\mathbf{v}}_2)\cdot e_1 & \breve{e}_2\cdot(\mathbf{v}_1\check{\mathbf{v}}_1+\mathbf{v}_2\check{\mathbf{v}}_2)\cdot e_2 \end{pmatrix} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \end{aligned} \quad \textbf{(A.5)}$$

since $\mathbf{v}_1\check{\mathbf{v}}_1 + \mathbf{v}_2\check{\mathbf{v}}_2 = 1$ by completeness and $\breve{e}_i \,.\, e_j = \delta_{ij}$. Consequently $U^{-1}MU =$ D as per Eq. (14).

In view of the fact tliat D is diagonal, we may write

$$e^{-Dt} = \begin{pmatrix} e^{-\lambda_1 t} & 0 \\ 0 & e^{-\lambda_2 t} \end{pmatrix} \tag{A.6}$$

Furthermore, we have

$$Ue^{-Dt}U^{-1} = \begin{pmatrix} e^{-\lambda_1 t}\cos^2\theta + e^{-\lambda_2 t}\sin^2\theta & (-e^{-\lambda_1 t} + e^{-\lambda_2 t}\sin^2\theta\cos\theta \\ (-e^{-\lambda_1 t} + e^{-\lambda_2 t}\sin^2\theta\cos\theta & e^{-\lambda_1 t}\sin^2\theta + e^{-\lambda_2 t}\cos^2\theta \end{pmatrix} , \tag{A.7}$$

and

$$UD^{-1}(1-e^{-Dt})U^{-1}B = -\frac{\gamma}{2}\begin{pmatrix} \frac{1-e^{-\lambda_1 t}}{\lambda_1}(\cos^2\theta - \sin\theta\cos\theta) + \frac{1-e^{-\lambda_2 t}}{\lambda_2}(\sin^2 8 - \sin\theta\cos 8) \\ \frac{1-e^{-\lambda_1 t}}{\lambda_1}(\sin^2 8 - \sin\theta\cos 8) + \frac{1-e^{-\lambda_2 t}}{\lambda_2}(\cos^2\theta - \sin\theta\cos\theta) \end{pmatrix} . \tag{A.8}$$

Noting that

$$\cos^2\theta - \sin^2 8 = \frac{\mathrm{R}}{\mu} , \ \sin\theta\cos\theta = \frac{i\gamma}{8\mu} \tag{A.9}$$

we can obtain. the solutions for $\rho_{+-}(t)$ and $\rho_{-+}(t)$ as in Eq.(20). By using Eq.(22) we can simply transform back to the bare-state representation, yielding

$$\rho_{ab}(t) = a_1(t) + a_2(t)\rho_{ab}(0) + a_3(t)\rho_{ba}(0) + a_4(t)\rho_{aa}(0) , \tag{A.10}$$

$$\rho_{aa}(t) = b_1(t) + b_2(t)\rho_{ab}(0) + b_3(t)\rho_{ba}(0) + b_4(t)\rho_{aa}(0) , \tag{A.11}$$

where the coefficients are found to be

$$\begin{aligned} a_1(t) &= \frac{-i\Omega\gamma}{\gamma^2+2\Omega^2}\left\{1 - e^{-3\gamma t/4}\left[\cos(\mu t) - \left(\frac{4\Omega^2-\gamma^2}{4\mu\gamma}\right)\sin(\mu t)\right]\right\} , \\ a_2(t) &= \frac{1}{2}e^{-\gamma t/2} + \frac{e^{-3\gamma t/4}}{8\mu}[\gamma\sin(\mu t) + 4\mu\cos(\mu t)] , \\ a_3(t) &= \frac{1}{2}e^{-\gamma t/2} - \frac{e^{-3\gamma t/4}}{8\mu}[\gamma\sin(\mu t) + 4\mu\cos(\mu t)] , \\ a_4(t) &= \frac{i\Omega}{\mu}e^{-3\gamma t/4}\sin(\mu t) , \end{aligned} \tag{A.12}$$

and

$$\begin{aligned} b_1(t) &= \frac{\Omega^2}{\gamma^2+2\Omega^2}\left[1 - \left(\cos(\mu t) + \frac{3\gamma}{4\mu}\sin(\mu t)\right)e^{-3\gamma t/4}\right] , \\ b_2(t) &= \frac{i\Omega}{2\mu}e^{-3\gamma t/4}\sin(\mu t) , \\ b_3(t) &= -\frac{i\Omega}{2\mu}e^{-3\gamma t/4}\sin(\mu t) , \\ b_4(t) &= e^{-3\gamma t/4}\left(\cos(\mu t) - \frac{\gamma}{4\mu}\sin(\mu t)\right) . \end{aligned} \tag{A.13}$$

Appendix B::Secular approximation

Here we present a simple way to obtain the main results for the resonance fluorescence spectrum in the strong field limit by a secular approximation. Let us focus on Eq.(9). In the strong field limit (such that $\Omega \gg \gamma/4$), we may ignore the last two terms in Eq.(9b) (because they will lead to rapidly oscillating terms), and write

$$\dot{\rho}_{+-} \cong -\left(\frac{3\gamma}{4} + i\Omega\right)\rho_{+-} \, . \tag{B.1}$$

Similarly for ρ_{-+}, we have

$$\dot{\rho}_{-+} \cong -\left(\frac{3\gamma}{4} - i\Omega\right)\rho_{-+} \, , \tag{B.2}$$

and get the solution

$$\rho_{+-}(t) = \rho_{+-}(0)e^{-3\gamma t/4}e^{-i\Omega t} \, ,$$

$$\rho_{-+}(t) = \rho_{-+}(0)e^{-3\gamma t/4}e^{i\Omega t} \, .$$

Substituting Eq.(B3) (together with Eq.(11) for $\rho_{++}(t)$) into Eq.(21) gives

$$(\tilde{o}(t))e^{iut} = \frac{1}{2}[(2\rho_{++}(0) - 1)e^{-\gamma t/2} - (\rho_{+-}(0)e^{-3\gamma t/4}e^{-i\Omega t} - c.c.)] \, . \tag{B.4}$$

Now using the Lax-Onsager theorem as in Eq.(24), we find

$$\begin{aligned} \langle\sigma^\dagger(0)\sigma(t)\rangle e^{i\nu t} &= \frac{1}{2}\rho_{aa}(0)e^{-\gamma t/2} \\ &- \frac{1}{4}\left[-\rho_{ba}(0) - \rho_{aa}(0)\right]e^{-3\gamma t/4}e^{-i\Omega t} + \frac{1}{4}\left[-\rho_{ba}(0) + \rho_{aa}(0)\right]e^{-3\gamma t/4}e^{i\Omega t} \, . \end{aligned} \tag{B.5}$$

As noted before, the initial time $t = 0$ can be chosen at any time after reaching steady state, and the strong field limit gives

$$(\rho_{aa})_{ss} = (\rho_{bb})_{ss} = \frac{1}{2} \, , \quad (\rho_{ab})_{ss} = (\rho_{ba})_{ss} = 0 \, . \tag{B.6}$$

Therefore, we have

$$\langle\sigma^\dagger(0)\sigma(t)\rangle e^{i\nu t} = \frac{1}{2}\left[\frac{1}{2}e^{-\gamma t/2} + \frac{1}{4}e^{-3\gamma t/4}e^{-i\Omega t} + \frac{1}{4}e^{-3\gamma t/4}e^{i\Omega t}\right] \, . \tag{B.7}$$

In the strong field limit Eq.(B7) is now identical to Eq.(27) in Section III, and readily shows the main features of the resonance fluorescence spectrum given in Eq.(30).

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