Effect of four-photon interactions on coherent population trapping in Λ-systems

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The resonance fluorescence spectrum of a Λ-system excited by two resonant light fields is calculated using a Markov analysis. Analytical formulas are derived in the strong-field limit within and beyond the rotating wave approximation. It is shown that the resonance fluorescence of the system does not vanish during coherent population trapping. Its spectrum consists of two multiplets which are similar to a triplet in the resonance fluorescence spectrum of a two-level atom and lie at the electronic transition frequencies, together with two triplets located at the frequencies of four-photon processes involving the optical excitation fields. The latter are fundamental in character and impose limits on the lower bound of the dephasing rate for the Raman resonance owing to the effect of radiative decay of the dipole transitions on the dynamics of the ground state. The effect of four-photon dephasing on the absorption spectrum of a Λ-system is analyzed and found to lead to a substantial reduction in the depth of a dip in the absorption spectrum which vanishes as the laser field strength is increased. *© 1998 American Institute of Physics.* [S1063-7761(98)01001-4]

1. INTRODUCTION

The interaction of electromagnetic fields with atoms is one of the most fundamental problems in quantum optics. It is known that a much wider range of effects occur in multilevel atoms than in two-level atoms owing to field-induced coherence between the atomic states and quantum interference. The three-level systems realized in Λ-, Ξ-, and V-configurations play an important role in research on these effects, as they are of intermediate complexity between two-level and multilevel atoms. A whole series of new effects have been observed in them; coherent population trapping is one of the most intriguing and has been studied intensely, both experimentally and theoretically. (See the reviews by Agap'ev *et al.*[1] and Arimondo[2] and the references cited there.) Coherent populating trapping shows up most clearly in a three-level system with two close long-lived levels and a third level which lies far from them (Λ- or V-systems) that have been excited by two cw laser fields, so that the distant level is optically coupled to the two others. Tuning the driver fields to resonance with its dipole transitions leads to trapping of the populations of the system in a coherent superposition of the two close levels. In Raman absorption spectra this effect shows up as a very narrow dip against the background of an absorption line and in resonance fluorescence spectra it is observed as the absence of emission, which has led to its being referred to as a ''dark'' (or ''coherent population trapping'') resonance.

In this article we study the question of how four-photon interactions affect the coherent population trapping effect in a Λ-system excited by two cw laser fields, in particular, how ''dark'' the coherent population trapping resonance in the resonance fluorescence spectrum is and how deep the dip in the Raman absorption spectrum is. A rough estimate of the intensity of resonance fluorescence in a Λ-system during coherent population trapping has been made in the rotating wave approximation[3] which yields zero fluorescence intensity for two-level atoms. The same result can be seen in Fig. 5c of Narducci *et al.*,[4] which shows a calculated fluorescence spectrum for a Λ-system. This indicates that during coherent population trapping a Λ-system does not radiate and the dark line is entirely absent in its resonance fluorescence spectrum.

Our calculations, presented in this paper in the asymptotic limit of a strong field, show, however, that the resonance fluorescence of a system does not vanish during coherent population trapping. Its spectrum consists of two multiplets, similar to the triplet in the resonance spectrum of two-level atoms and located at the electronic transition frequencies, together with two triplets located at the frequencies of four-photon processes involving the pump light fields. The latter are fundamental in character and impose limits on the lower bound of the dephasing rate of the Raman resonance owing to the contribution to the dynamics of the ground state from radiative decay of dipole transitions. The effect of the four-photon dephasing mechanism on the absorption spectrum of a Λ-system is analyzed and found to lead to a substantial reduction in the depth of a dip in the absorption spectrum that vanishes as the laser field intensity is increased.

This article is organized as follows: Section 2 is devoted to a description of the complete Liouvillian of an atom interacting with a laser radiation field. The specific features of the excitation of two-level atoms and a Λ-system are analyzed. In Sec. 3 the resonance fluorescence spectrum of a Λ-system

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is calculated in the rotating wave approximation, as well as outside the range of validity of this approximation. The effect of four-photon interactions involving the driver light fields on coherent population trapping and their role in the formation of the absorption resonance and in the dispersion of the Λ-system are analyzed in Secs. 4 and 5, respectively. Most of the voluminous mathematical calculations are carried out in the Appendix. In the Conclusion we discuss an experiment for detecting the calculated structure of the resonance fluorescence spectrum of a Λ-system.

2. LIOUVILLIAN OF AN ATOM BEYOND THE RANGE OF VALIDITY OF THE ROTATING WAVE APPROXIMATION; DYNAMICAL TRANSFORMATIONS USED TO CALCULATE THE FLUORESCENCE SPECTRUM

The complete Liouvillian of an atom, which describes changes in the atomic variables according to the equation $d\hat{A}/dt=\mathscr{L}(t)\hat{A}$ in Markov theory, has the form

$$\mathscr{L}(t)=\mathscr{L}_0+\mathscr{L}_\delta+\mathscr{L}_r+\mathscr{L}_L(t). \tag{1}$$

Here $\mathscr{L}_0$ is the unperturbed Liouvillian $(i/\hbar)[\hat{\mathscr{H}}_0,\odot]$, including the free precession of the atom at the laser frequencies according to Eq. (A4) of Appendix A. (The symbol $\odot$ denotes a place for substituting a transformed operator.) $\mathscr{L}_r$ and $\mathscr{L}_\delta$ determine the dynamics of the atom owing, respectively, to relaxation and nonzero detuning of the frequencies of the driver laser fields from the resonance transition frequencies in the atom (resonance excitation is described by $\mathscr{L}_0$), while $\mathscr{L}_L(t)$ describes the laser excitation.

In terms of the interaction representation the transformation $S(0,t)$ corresponding to the Liouvillian (1) takes the form

$$S(0,t)=S_{RWA}(0,t)\widetilde{S}_0(0,t)e^{\mathscr{L}_0 t}, \tag{2}$$

where the superoperators

$$S_{RWA}(0,t)=\exp(\mathscr{L}_{RWA}t), \quad \mathscr{L}_{RWA}=\mathscr{L}_\delta+\mathscr{L}_r+\mathscr{L}_p \tag{3}$$

determine the system dynamics in the rotating wave approximation (RWA),

$$\widetilde{S}_0(0,t)=\mathrm{T}\exp\left[\int_0^t \delta\mathscr{L}_p(\tau)d\tau\right] \tag{4}$$

is the evolution superoperator for the dynamics of the system owing to the nonresonant excitation component, and

$$\delta\mathscr{L}_p(t)=e^{\mathscr{L}_0 t}\mathscr{L}_L(t)e^{-\mathscr{L}_0 t}-\mathscr{L}_p \tag{5}$$

is the deviation from the average value $\mathscr{L}_p$ of the Liouvillian for the laser excitation.[5] The symbol T used in Eq. (4) denotes the time ordering of the superoperator taken in quantum mechanics.

The deviation $\delta\mathscr{L}_p(\tau)$ in Eq. (5) oscillates at the frequencies of the laser drive fields and their combinations. Thus, we can treat it as a universally small perturbation relative to the resonant excitation contribution for the amplitude of driver waves smaller than the amplitude of the intra-atomic field. In a first-order approximation with respect to this parameter, the evolution superoperator has the form

$$S(0,t)=S(0,t)_{RWA}\left[1+\int_0^t \delta\mathscr{L}_p(\tau)d\tau\right]e^{\mathscr{L}_0 t}. \tag{6}$$

Note that this approximation is valid if the value of the integral is less than of order unity.

We now consider the specifics features of the excitation of a two-level atom and a Λ-system.

2.1. Excitation of a two-level atom

For a two-level atom excited by a laser field $E_L\cos(\omega_L t)$, Eq. (5) takes the form

$$\delta\mathscr{L}_p(\tau)=i\,\frac{\Omega_0}{2}\,[\hat{\sigma}^+\exp(-2i\omega_L t) + \hat{\sigma}^-\exp(2i\omega_L t),\odot], \tag{7}$$

where Ω_0 is the Rabi frequency and $\hat{\sigma}^\pm$ are the standard Pauli matrices. Applying Eq. (7) to the complex polarization amplitude $\hat{\sigma}^+$ and using Eq. (6), we find that $S(t)\hat{\sigma}^+$ determines the structure of a triplet (because of the presence of the term $S(t)_{RWA}$) in the resonance fluorescence spectrum of a two-level atom at a frequency of $3\omega_L$,[5] which is analogous to the known triplet at the laser excitation frequency.[6,7] Integrating with respect to τ in Eq. (6), we can easily show that the ratio of the corresponding amplitudes of the spectral components at the frequencies of the third harmonic and the laser light is proportional to the small quantity $\Omega_0/2\omega_L$.

2.2. Excitation of a Λ-system

Let us consider a Λ-system consisting of three electronic levels with transition frequencies among them of $\omega_{12}\ll\omega_{13}$, ω_{23} (Fig. 1). Two coherent fields $E\cos(\omega_L t)$ and $E'\cos(\omega_L' t)$ act, respectively, on the transitions $1\leftrightarrow 3$ and $2\leftrightarrow 3$. These fields interact with the complete dipole moment of the system determined by the operator $d_{13}\hat{\sigma}_{13}+d_{23}\hat{\sigma}_{23}$, where $\hat{\sigma}_{13,23}$ are the Pauli matrices $\hat{\sigma}_1$ for the corresponding atomic transitions. As a result, the induced dipole moment of the system oscillates at frequencies $\pm\omega_L$ and $\pm\omega_L'$.

As opposed to the case of two-level atoms, where both driver fields interact with one and the same atomic transition, during excitation of a Λ-system each field interacts with two transitions. Thus, the Liouvillian corresponding to biharmonic laser excitation with a frequency detuning $\Delta=\omega_L'-\omega_L$ takes (according to Eq. (B3)) the form

$$\delta\mathscr{L}_p(\tau)=\frac{i\hbar g_\Lambda}{2}\,[(\hat{\tau}e^{i\Delta t}+\hat{\tau}^+e^{-i\Delta t}),\odot], \tag{8}$$

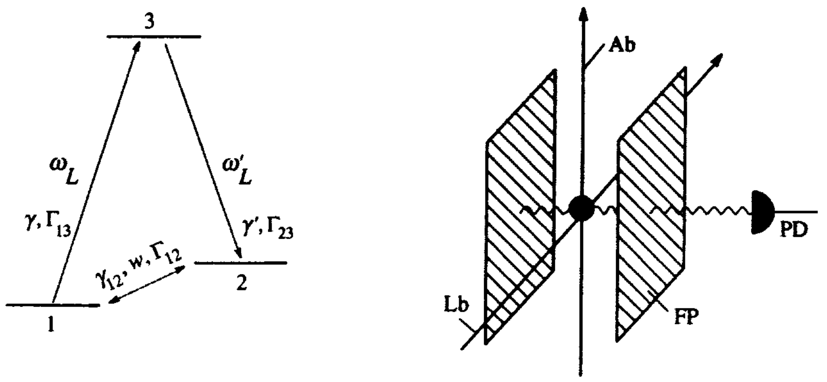

FIG. 1. A Λ-system (a) and a typical arrangement for the experimental measurement of resonance fluorescence induced by two monochromatic laser fields with frequencies ω_L and ω'_L (b). γ, γ', and γ_{12} are the relaxation rates of the populations in the upper levels; Γ_{13}, Γ_{23}, and Γ_{12} are the dephasing rates; and, w is the rate of pumping to level 2. The fluorescence spectrum of the atoms is analyzed using a Fabry-Perot interferometer (FP) and a photodiode (PD) in a direction perpendicular to the directions of the laser (Lb) and atom (Ab) beams.

where $g_\Lambda=\sqrt{g^2+g'^2}$, $g=d_{13}E'$, $g'=d_{23}E$, and the operator $\hat\tau$ is defined as

$$\hat\tau=g_\Lambda^{-1}(g\hat\sigma_{13}^{+}+g'\hat\sigma_{23}^{-}). \qquad (9)$$

In deriving Eq. (8) we have neglected terms containing sums with higher frequencies.

Equation (8) obviously determines additional spectral components at frequencies $\omega_L\pm\Delta$ and $\omega'_L\pm\Delta$, of which only the components at $\omega_L-\Delta$ and $\omega'_L+\Delta$ are new. The correspond to four-photon processes and should show up for a symmetric Λ-system as a mirror reflection of the virtual levels of the subsystem of lower levels (Fig. 2) owing to modulation of the $1\leftrightarrow3$ and $2\leftrightarrow3$ transitions by the intrinsic oscillatory frequency $\Delta\approx\omega_{12}$ of the lower level subsystem. It is known that four-photon frequency mixing leads to generation of a coherent signal at the Stokes and anti-Stokes frequencies.[8] Our later calculations show, however, that these nonlinear resonances are also accompanied by sidebands because of incoherent scattering processes.

The above analysis shows that the important difference between exciting a two-level atom (see Eq. (7)) and a Λ-system (Eq. (8)) is that excitation in the case of the Λ-system is mainly determined by the biharmonic frequency detuning Δ. In experiments this detuning is usually much smaller than the frequencies of the laser systems that are exciting the system. This means that the intensity of the additional components in the fluorescence spectrum (fine structure), which is determined by the exponential factors in Eqs. (7) and (8), is substantially higher for a Λ-system than for a two-level atom.

3. CALCULATING THE FLUORESCENCE SPECTRUM OF A Λ-SYSTEM

The spectral density of the emission from an excited atom (resonance fluorescence spectrum) is determined by the normally ordered two-time correlation function of the light emitted by the atom.[9,10] Assuming that the atomic fluctuations are Markovian, i.e., they are independent of one another at times t and $t+\tau$, we can write down the correlation function for the atomic fluorescence in the form

$$\mathcal{K}(\tau)=\langle\hat\rho_0S(0,t)|\hat\sigma^-(t)[S(t,t+\tau)\hat\sigma^+(t+\tau)]\rangle, \qquad (10)$$

where $\hat\sigma^\pm(t)$ are the Heisenberg positive (negative) frequency operators. These operators have a time dependence only in the form of high-frequency oscillations at optical frequencies. The superoperators $S(0,t)$ and $S(t,t+\tau)$ describe the relaxation and interaction of the atoms with the exciting laser fields during the time intervals $(0,t)$ and $(t,t+\tau)$, and $\hat\rho_0S(0,t)$ is the density matrix $\hat\rho(t)$ at time t. It follows from Eq. (3) that the superoperators $S(0,t)$ and $S(t,t+\tau)$ are simple exponentials of the form $\exp[\mathscr{L}_{RWA}t]$, according to the rotating wave approximation.

The term $\hat\sigma^-(t)[S(t,t+\tau)\hat\sigma^+(t+\tau)]$ in Eq. (10) is simply the product of the two operators $\hat\sigma^-(t)$ and $\hat\sigma^+(t+\tau)$ averaged over the fluctuations in the time interval $(t,t+\tau)$. This averaging is carried out with the aid of the transformation $S(t,t+\tau)$, which determines the conditional atomic quantum mechanical probability distribution function at time $t+\tau$ relative to time t. The emission spectrum of the atom can then be calculated as the Fourier transform of the correlation function (10).

In the stationary case the density matrix in the vector representation is simply the zero vector $\langle0|$ of the matrix of the evolution superoperator $\mathscr{L}_{RWA}$. Then we can obtain the stationary correlation function from Eq. (10) by averaging it over the temporal oscillations. This averaging leads to the replacement of the bilinear combination of the complete operators $\hat\sigma^\pm(t)$ by two combinations:

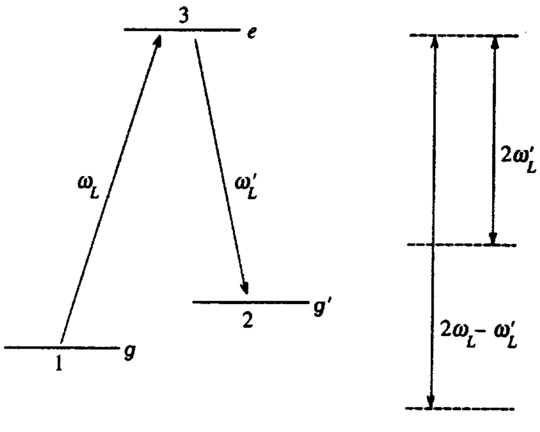

FIG. 2. A Λ-resonance and additional resonances which determine the fine structure of the resonance fluorescence spectrum.

$$\hat{\sigma}^-(t)\times\hat{\sigma}^+(t+\tau)\rightarrow\hat{\sigma}^-_{13}\times\hat{\sigma}^+_{13}(\tau)\oplus\hat{\sigma}^-_{23}\times\hat{\sigma}^+_{23}(\tau).$$

Similarly, we have

$$\mathscr{K}(\tau)\rightarrow\mathscr{K}_1(\tau)+\mathscr{K}_2(\tau).$$

When we calculate the resonance fluorescence spectrum of the atom in the rotating wave approximation the correlation functions $\mathscr{K}_{1,2}(\tau)$ correspond to the frequencies $\omega_{13}\approx\omega_L$ and $\omega_{23}\approx\omega_L'$ and when we calculate the fine structure of the spectrum beyond the range of validity of this approximation they correspond, respectively, to the frequencies $\omega_L-\Delta=2\omega_L-\omega_L'$ and $\omega_L'+\Delta=2\omega_L'-\omega_L$. (See Sec. 2.2.)

On describing the Λ-system with the aid of the Liouvillian in the rotating wave approximation and expanding it in terms of the eigen-projectors, we obtain the following relatively simple expression:

$$\mathscr{K}(\tau)=\sum_{k=0}^{8}\{\langle 0|\hat{\sigma}^-_{13}\cdot|k\rangle\rangle\langle k|\hat{\sigma}^+_{13}\rangle\exp[(\lambda_k-i\omega_L)\tau]+\langle 0|\hat{\sigma}^-_{23}\cdot|k\rangle\rangle\langle k|\hat{\sigma}^+_{23}\rangle\exp[(\lambda_k-i\omega_L')\tau]\},\quad(11)$$

where the symbol "$\cdot$" means that the operators are multiplied in accordance with the multiplication rules for operators and the result is presented in the form of a ket-vector; λ_k, $|k\rangle$, and $\langle k|$ are the eigenvalues of the matrix and the eigenvectors of the Liouvillian $\mathscr{L}_{RWA}$.

Using Eq. (11) together with Appendix B we can obtain the following expression for the correlation function that describes the structure of the resonance fluorescence spectrum of the atom outside the range of validity of the rotating wave approximation:

$$\mathscr{K}(\tau)=\frac{g_\Lambda^2}{4\omega_{12}^2}\sum_{k=0}^{8}\langle 0|\hat{\sigma}^-_{12}\cdot|k\rangle\langle k|\hat{\sigma}^+_{12}\rangle\{\exp[-i(\omega_L-\Delta)\tau]+\exp[-i(\omega_L'+\Delta)\tau]\}\exp(\lambda_k\tau),\quad(12)$$

where the $\hat{\sigma}^{\pm}_{12}$ are the complex conjugate amplitude of the subsystem of lower levels which modulate the dipole moment of the transition. This modulation gives rise to new spectral components in the fluorescence spectrum.

Recall that Eq. (12) describes only the basic structure of the fluorescence spectrum, which is determined by the parameter g_Λ/ω_{12}, which, in turn, we assume to be small. Here we have neglected the higher order contribution which makes a nonzero contribution to the coherent component of the response in the rotating wave approximation, which component equals zero when this correction is neglected in a strong field in a first-order approximation with respect to the parameter Γ/g_Λ for detunings δ, $\delta_R\approx 0$ (by analogy with the two-level atom[7]).

3.1. Fluorescence spectrum in the rotating field approximation

The fluorescence spectrum determined by Eq. (11) is the sum of Lorentz spectrum lines whose total spectral powers are proportional to the coefficient in front of the corresponding exponent in Eq. (11), while their width and frequency shift are determined by the real and imaginary parts of the eigenvalues λ_k. In general, the fluorescence spectrum can be calculated numerically. In an asymptotically strong field, however, as we shall show below (and has been demonstrated previously[4] for a special case) an analytic solution can also be obtained.

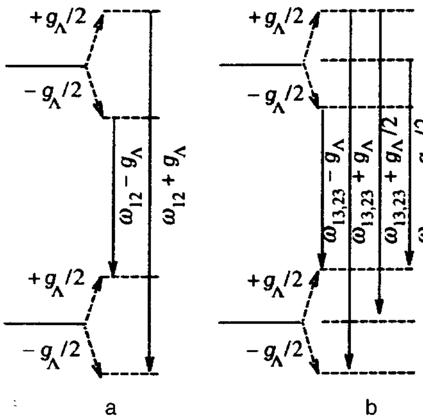

FIG. 3. The formation of Rabi nutations in a two-level atom (a) and in a Λ-system (b). Only a minimum set of transitions between the quasienergy states, corresponding to the set of all possible lines in the fluorescence spectrum, is shown,

For simplicity let us consider a Λ-system excited by two high-power laser fields of equal intensity. In this case we can average the relaxation of the system over the Rabi nutations, while the Hamiltonian corresponding to the laser-induced precession operator $\mathscr{L}_p=(i/\hbar)[\hat{\mathscr{H}}_p,\odot]$ takes the form

$$\hat{\mathscr{H}}_p=\hbar\,\frac{g}{2}\,\hat{\sigma}=\hbar\,\frac{g_\Lambda}{2\sqrt{2}}\begin{pmatrix}0&1&1\\1&0&0\\1&0&0\end{pmatrix}.$$

This Hamiltonian corresponds to quasi-energy states with energies that are shifted with respect to the eigenvalues of the Hamiltonian $\hat{\mathscr{H}}_p$, which equal $\{0,\pm g_\Lambda/2\}$.[11] (For a two-level atom the eigenvalues of the Hamiltonian are equal to $\{\pm g_\Lambda/2\}$). The temporal dynamics of these mixed quasi-energy states cause oscillations in the expected values of the physical variables at two different frequencies g_Λ and $g_\Lambda/2$. The physical significance of these nutations in terms of the quasienergy levels is illustrated in Fig. 3.

The Rabi nutations between the quasi-energy levels are described by a Liouvillian which, in the operator basis $\{\hat{n}_3,\hat{n}_1,\hat{n}_2,\hat{\sigma}^c_{12},\hat{\sigma}^s_{12},\hat{\sigma}^c_{13},\hat{\sigma}^s_{13},\hat{\sigma}^c_{23},\hat{\sigma}^s_{23}\}$ (the indices c and s denote the cosine and sine components, respectively), takes the form

$$\mathscr{L}_p=\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \frac{g_\Lambda}{2} & 0 & \frac{g_\Lambda}{2} \\ 0 & 0 & 0 & 0 & 0 & 0 & -\frac{g_\Lambda}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\frac{g_\Lambda}{2} \\ 0 & 0 & 0 & 0 & 0 & 0 & -\frac{g_\Lambda}{2\sqrt{2}} & 0 & -\frac{g_\Lambda}{2\sqrt{2}} \\ 0 & 0 & 0 & 0 & 0 & \frac{g_\Lambda}{2\sqrt{2}} & 0 & -\frac{g_\Lambda}{2\sqrt{2}} & 0 \\ 0 & 0 & 0 & 0 & -\frac{g_\Lambda}{2\sqrt{2}} & 0 & 0 & 0 & 0 \\ -\frac{g_\Lambda}{2} & \frac{g_\Lambda}{2} & 0 & \frac{g_\Lambda}{2\sqrt{2}} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{g_\Lambda}{2\sqrt{2}} & 0 & 0 & 0 & 0 \\ -\frac{g_\Lambda}{2} & 0 & \frac{g_\Lambda}{2} & \frac{g_\Lambda}{2\sqrt{2}} & 0 & 0 & 0 & 0 & 0 \end{pmatrix}. \tag{13}$$

Its eigenvalues λ_k ($k=0, \ldots, 8$) are equal to 0, 0, 0, $-ig_\Lambda/2$, $-ig_\Lambda/2$, $ig_\Lambda/2$, $ig_\Lambda/2$, $-ig_\Lambda$, ig_Λ, while the corresponding set of eigenvectors is defined as

$$\{\psi_k\}=\left\{\begin{array}{ccccccccc} 0 & 0 & 0 & 0 & 0 & \sqrt{1/2} & 0 & \sqrt{1/2} & 0 \\ 0 & -1/2 & -1/2 & \sqrt{1/2} & 0 & 0 & 0 & 0 & 0 \\ \sqrt{1/2} & 2^{-3/2} & 2^{-3/2} & 1/2 & 0 & 0 & 0 & 0 & 0 \\ 0 & i/2 & -i/2 & 0 & 0 & 0 & -1/2 & 0 & 1/2 \\ 0 & 0 & 0 & 0 & \sqrt{1/2} & -i/2 & 0 & i/2 & 0 \\ 0 & 0 & 0 & 0 & \sqrt{1/2} & i/2 & 0 & -i/2 & 0 \\ 0 & -i/2 & i/2 & 0 & 0 & 0 & -1/2 & 0 & 1/2 \\ 1/2 & -1/4 & -1/4 & -2^{-3/2} & 0 & 0 & -i/2 & 0 & -i/2 \\ 1/2 & -1/4 & -1/4 & -2^{-3/2} & 0 & 0 & i/2 & 0 & i/2 \end{array}\right\}.$$

Let us now discuss the physical significance of the dynamical variables corresponding to the eigenvectors ψ_k.

The eigenvector ψ_0 describes the stationary excitation of a system by two laser fields of equal intensity acting on the $1\leftrightarrow 3$ and $2\leftrightarrow 3$ transitions, respectively. The eigenvectors ψ_1 and ψ_2 describe a two-dimensional stationary excitation space, a combination of the polarization of the ground state and the populations of all three levels. The eigenvectors ψ_3 and ψ_6 describe excitation which involves a combination of the populations of the subsystems of the lower levels and an independent combination of the polarizations $1\leftrightarrow 3\oplus 2\leftrightarrow 3$; they oscillate at half the Rabi frequency, $g_\Lambda/2$. The eigenvectors ψ_4 and ψ_5 describe excitations which are a combination of three polarizations ($1\leftrightarrow 2$ and an independent combination $1\leftrightarrow 3\oplus 2\leftrightarrow 3$) which also oscillate at a frequency of $g_\Lambda/2$. The last two eigenvectors, ψ_7 and ψ_8, describe the excitation of the bound state together with the populations of all three levels and the polarization of the ground state, (all) oscillating at a frequency of g_Λ. Therefore, the Rabi nutations of the eigenexcitations of the system for combinations of the unbound levels take place at a frequency of $g_\Lambda/2$, while the bound states oscillate at a frequency of g_Λ. (See Eq. (A5)).

Using Eq. (13) for the nutation operator $\mathscr{L}_P$, we can average the Liouvillian $\mathscr{L}_\delta+\mathscr{L}_r$ in Eq. (1) over the nutations and write it in the form of a sum of 3×3 matrices, two 2×2, and two 1×1. (The last two are diagonal elements.) As a result, we can obtain a simplified expression for the last three terms in Eq. (1), which describe the overall dynamics of the system in the interaction representation, of the form

$$\mathscr{L}_{RWA}=\begin{pmatrix} -(\Gamma_{13}+\Gamma_{23})/2 & 0 & 0 \\ 0 & -\Gamma_{12}/2 & -(\gamma+\gamma'+\Gamma_{12})/2\sqrt{2} \\ 0 & -\Gamma_{12}/2\sqrt{2} & -(\gamma+\gamma'+\Gamma_{12})/4 \end{pmatrix}$$

$$\oplus\begin{pmatrix} -(\gamma_{12}+w)/2-\Gamma_{13}/4-\Gamma_{23}/4-ig_{\Lambda}/2 & -i\delta_s/2 \\ -i\delta_s/2 & -\Gamma_{12}/2-\Gamma_{13}/4-\Gamma_{23}/4-ig_{\Lambda}/2 \end{pmatrix}$$

$$\oplus\begin{pmatrix} -\Gamma_{12}/2-\Gamma_{13}/4-\Gamma_{23}/4+ig_{\Lambda}/2 & i\delta_s/2 \\ i\delta_s/2 & -(\gamma_{12}+w)/2-\Gamma_{13}/4-\Gamma_{23}/4+ig_{\Lambda}/2 \end{pmatrix}$$

$$\oplus(-3\gamma/8-3\gamma'/8-\Gamma_{12}/8-\Gamma_{13}/4-\Gamma_{23}/4-ig_{\Lambda})$$

$$\oplus(-3\gamma/8-3\gamma'/8-\Gamma_{12}/8-\Gamma_{13}/4-\Gamma_{23}/4+ig_{\Lambda}),$$

where the total detuning is $\delta_s=2\delta+\delta_R=\omega_L+\omega_L'-\omega_{13}-\omega_{23}$. The corresponding eigenvalues are given by

$$\{\lambda_\kappa\}=\left\{\begin{matrix} 0 \\ -(\gamma+\gamma'+3\Gamma_{12})/4 \\ -(\Gamma_{13}+\Gamma_{23})/2 \\ \mu_1 \\ \mu_2 \\ \mu_1^* \\ \mu_2^* \\ (-3\gamma-3\gamma'-\Gamma_{12}-2\Gamma_{13}-2\Gamma_{23}-8ig_{\Lambda})/8 \\ (-3\gamma-3\gamma'-\Gamma_{12}-2\Gamma_{13}-2\Gamma_{23}+8ig_{\Lambda})/8 \end{matrix}\right\}, \tag{14}$$

where

$$\mu_{1,2}=\frac{1}{4}\Big[-\gamma_{12}-w-\Gamma_{12}\pm i\sqrt{4\delta_s^2-(\gamma_{12}+w+\Gamma_{12})^2} -\Gamma_{13}-\Gamma_{23}-2ig_{\Lambda}\Big].$$

Here γ_{12} and w are the rates of relaxation and pumping of the lower level system, Γ_{12} is the dephasing rate in this system, γ and γ' are the rates of relaxation from the excited states, and Γ_{13} and Γ_{23} are the corresponding dephasing rates.

Let us now discuss the eigenvalues λ_k in detail.

Note that because the relaxation operator is not self-adjoint, each eigenvalue corresponds to two eigenvectors, one of which describes the operators acting on the physical variables, while the other describes the density matrix. The eigenvalue $\lambda_0=0$ corresponds to the stationary state $\hat{\rho}_{st}\rightarrow\langle 0|$ and the operator $\hat{I}\rightarrow|0\rangle$, which has no dynamical significance. This eigenvalue determines the coherent line in the fluorescence spectrum. The eigenvalues $\lambda_{1,2}$ describe the nonoscillatory dynamics of the system and determine the Rayleigh scattering of the fields which excite the system. The four eigenvalues $\lambda_{3,4,5,6}$ determine oscillations at half the Rabi frequency $g_{\Lambda}/2=g/\sqrt{2}$ and describe the contribution of field-induced resonances to the fluorescence spectrum. The last two eigenvalues $\lambda_{7,8}$ determine oscillations at the Rabi frequency g_{Λ} and describe the ordinary $1\leftrightarrow 3$ and $2\leftrightarrow 3$ nutations in the weak-field limit.

The preceding analysis shows that in a strong field only the total detuning $\delta_s=2\delta+\delta_R$ appears among the eigenfrequencies of the system, and not the Raman detuning δ_R. Here for $\delta=0$, relaxation in the system of lower levels does not contribute to the oscillations at half the Rabi frequency:

$$\mu_{1,2}=-\Gamma_{13}/4-\Gamma_{23}/4-ig_{\Lambda}/2.$$

This effect can, in principle, be used to study the contribution of the lower level system to the fluorescence spectrum in an experiment where the spectra are measured as a function of the detuning δ for different intensities of the laser line. The measured width of the spectral components located at half the Rabi frequency is then determined directly by the relaxation rate in the lower level system.

For the case of an exact resonance $(\delta,\delta_R=0)$, we can obtain an analytic expression for the fluorescence spectrum in the rotating wave approximation. The major difference compared to the spectra from two independent two-level systems, however, is that in the case of the Λ-system the general coefficient in Eq. (11), which determines the intensity of the spectral components, differs from the corresponding coefficient for the case of a two-level atom, which is simply proportional to γ. In coherent population trapping, this coefficient for a Λ-system and, therefore, the intensity of the spectral components decrease by roughly a factor of Γ_{12}/γ, which is a small parameter. For the cesium atom,[12] as an example, it is $\approx 1.6\times 10^{-3}$, while for sodium[2] it can be estimated to be $\approx 4.9\times 10^{-3}$ using published parameters.[13]

3.2. Fine structure of the fluorescence spectrum

For simplicity let us again consider the case of an exact resonance. Using the equations from Sec. 3.1 together with Eq. (12), we obtain the following expressions for the coefficients c_k in front of the exponential factors:

$$c_0=\frac{\gamma^2/\Gamma_{12}^2}{\sqrt{(3+2\gamma/\Gamma_{12})^3}},\quad c_1=\frac{9}{8}\,\frac{1+2\gamma/\Gamma_{12}}{\sqrt{(3+2\gamma/\Gamma_{12})^3}},$$

$$c_{2,3,5}=0,\quad c_{4,6}=\frac{1}{4}\,\frac{1+\gamma/\Gamma_{12}}{\sqrt{3+2\gamma/\Gamma_{12}}},$$

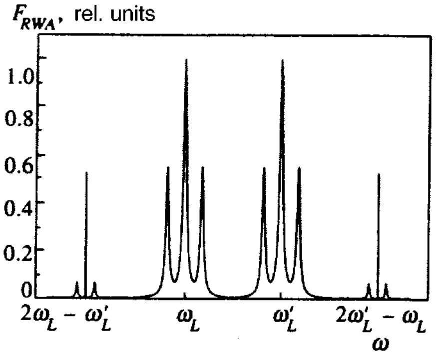

FIG. 4. The resonance fluorescence spectrum of a Λ-system (the ${}^2S_{1/2} \to {}^2P_{3/2}$ transition in the cesium atom) excited by two intense laser fields into a coherent population trapping state.

$$c_{7,8}=\frac{1}{16}\frac{1}{\sqrt{3+2\gamma/\Gamma_{12}}}, \tag{15}$$

These coefficients multiplied by the common factor $\gamma g_\Lambda^2/4\omega_{12}^2$ determine the intensity of the fine structure components of the fluorescence spectrum (12).

The complete fluorescence spectrum of a Λ-system, including the structure in the rotating wave approximation as well as the fine structure calculated above, is shown in Fig. 4. Equation (15) implies that for the typically large values of the ratio γ/Γ_{12} only the coefficients c_0 and $c_4=c_6$ are proportional to the large values of order $\sqrt{\gamma/\Gamma_{12}}$. As a result, only three lines show up in each of the two fine structure features of the fluorescence spectrum (Fig. 4). One of them is coherent (i.e., has zero width) with an intensity proportional to c_0, while the other two are broadened lines with an intensity proportional to c_4 shifted to the left and right of the coherent line at half the Rabi frequency $g_\Lambda/2$.

4. EFFECT OF FOUR-PHOTON INTERACTIONS ON COHERENT POPULATION TRAPPING

Using Appendix B we can easily calculate the contribution to the relaxation of the ground state from the relaxation contributions of the dipole transitions. The nature of the relaxation processes involves an interaction of the dipole moments of the $1\leftrightarrow 3$ and $2\leftrightarrow 3$ transitions with the vacuum fluctuations of the electromagnetic field. These interactions are described by the Hamiltonian

$$\hat{\mathscr{H}}_\xi=\frac{\hbar}{2}\left(\hat{\xi}_1^-\hat{\sigma}_{13}^+ + \hat{\xi}_2^-\hat{\sigma}_{23}^+\right)+\text{H.c.}, \tag{16}$$

where

$$\hat{\xi}_1^-=\frac{1}{\hbar}\int \mathbf{d}_{13}(\mathbf{r})\hat{\mathbf{E}}_0(\mathbf{r}), \quad \hat{\xi}_2^-=\frac{1}{\hbar}\int \mathbf{d}_{23}(\mathbf{r})\hat{\mathbf{E}}_0(\mathbf{r})$$

are the components of the vacuum electromagnetic field $\hat{\mathbf{E}}_0(\mathbf{r})$ with negative frequency amplitudes integrated over the spatial distribution $\mathbf{d}_{k3}(\mathbf{r})$ of the dipole moments $(k=1,2)$.

Isolating the contribution to $\hat{\sigma}_{13}^+$ and $\hat{\sigma}_{23}^+$ owing to four-photon interactions outside the range of applicability of the rotating wave approximation, we can write Eq. (16) in the form

$$\hat{\mathscr{H}}_\xi=\hat{\mathscr{H}}_\xi^0+\hat{\mathscr{H}}_\xi^{4ph},$$

$$\hat{\mathscr{H}}_\xi^{4ph}=\frac{\hbar}{2}\left(\hat{\xi}_1^-\,\delta S_0\hat{\sigma}_{13}^+ + \hat{\xi}_2^-\,\delta S_0\hat{\sigma}_{23}^+\right)+\text{H.c.},$$

where $\hat{\mathscr{H}}_\xi^0$ determines the standard interaction, $\hat{\mathscr{H}}_\xi^{4ph}$ describes the additional contribution owing to four-photon interactions, and δS_0 is the four-photon contribution to the dynamic transformation of the Λ-system. Using the final formulas of Appendix B for the transformed operators $\hat{\sigma}_{13}^+$ and $\hat{\sigma}_{23}^+$, we obtain the following formula for the four-photon contribution to the Hamiltonian:

$$\begin{aligned}\hat{\mathscr{H}}_\xi^{4ph}=\frac{g_\Lambda}{2\Delta}\frac{\hbar}{2}\{&\hat{\xi}_1^-[\hat{\tau},\hat{\sigma}_{13}^+]\exp[-i(\omega_L-\Delta)t]\\&+\hat{\xi}_2^-[\hat{\tau}^+,\hat{\sigma}_{23}^+]\exp[-i(\omega'_L+\Delta)t]\}+\text{H.c.}\end{aligned} \tag{17}$$

By calculating the commutators in Eq. (17), we can write down the four-photon contribution with the aid of the transition operators for the low level subsystem as

$$\begin{aligned}\hat{\mathscr{H}}_\xi^{4ph}=\frac{\hbar}{4\Delta}\{&g'\hat{\xi}_1^-(t)\exp[-i(\omega_L-\Delta)t]\hat{\sigma}_{12}^-\\&+g\hat{\xi}_2^-(t)\exp[-i(\omega'_L+\Delta)t]\hat{\sigma}_{12}^+\}+\text{H.c.}\end{aligned} \tag{18}$$

This implies that the vacuum electromagnetic field interacts with the lower level subsystem through four-photon processes. The efficiency of this interaction depends on the interaction constant of the laser fields with the dipole transitions of the Λ-system. The distinctive feature of this interaction is that the emission of a vacuum photon by the $1\leftrightarrow 3$ transition is accompanied by the absorption of a $1\leftrightarrow 2$ transition photon, while the emission of a vacuum photon by the $2\leftrightarrow 3$ transition is accompanied by the emission of a $1\leftrightarrow 2$ transition photon. Conservation of energy in these processes is ensured by the four-photon interaction of the vacuum field with the laser fields, and this is reflected in the exponential terms in Eq. (18).

Following Ref. 10, we can write the relaxation operator for the low level subsystem, which corresponds to Eq. (18), in its customary form in terms of the operator basis $\{\hat{n}_1,\hat{n}_2,\hat{\sigma}_1,\hat{\sigma}_2\}$:

$$\mathscr{L}_{12}$$

$$=\begin{pmatrix}-\gamma_{12} & \gamma_{12} & 0 & 0\\ w_{12} & -w_{12} & 0 & 0\\ 0 & 0 & -(\gamma_{12}+w_{12})/2 & 0\\ 0 & 0 & 0 & -(\gamma_{12}+w_{12})/2\end{pmatrix},$$

where γ_{12} is the relaxation rate of the lower level subsystem. Given that the term $g'\hat{\xi}_1^-\hat{\sigma}_{12}^-$ in Eq. (18) describes a relaxation transition from level 1 to level 2 and the term $g\hat{\xi}_2^-\hat{\sigma}_{12}^+$ describes a transition in the opposite direction, we obtain the following expression for the relaxation owing to the contribution of four-photon processes:

$$\mathscr{L}_{12}^{4ph}=\frac{g_\Lambda^2}{4\Delta^2}\begin{pmatrix} -c^2\gamma_{13} & c^2\gamma_{13} & 0 & 0 \\ c'^2\gamma_{23} & -c'^2\gamma_{23} & 0 & 0 \\ 0 & 0 & -(c^2\gamma_{13}+c'^2\gamma_{23})/2 & 0 \\ 0 & 0 & 0 & -(c^2\gamma_{13}+c'^2\gamma_{23})/2 \end{pmatrix}, \tag{19}$$

where $c=g/g_\Lambda$, $c'=g'/g_\Lambda$, and $c^2+c'^2=1$.

Equation (19) implies that the contribution of four-photon processes to the relaxation rate constant of the lower level subsystem, Γ_{12} is given by

$$\Gamma_{12}^{4ph}=\frac{g_\Lambda^2}{8\Delta^2}\,(c'^2\gamma_{13}+c^2\gamma_{23})\approx\frac{g_\Lambda^2}{4\Delta^2}\,\gamma_{13}/2.$$

This contribution leads to a fundamental lower bound on Γ_{12}. As an example, for the Cs atom and laser field intensities of ~ 1 W/cm^2, we have $\Gamma_{12}^{4ph}\sim 10^{-3}\gamma_{13}/2\approx 10^4$ s^{-1}.

5. THE ROLE OF FOUR-PHOTON INTERACTIONS IN THE FORMATION OF AN ABSORPTION RESONANCE AND DISPERSION

The simplest experimental possibility for observing the dark resonance is to measure the transmission and/or dispersion (in atomic vapor) of exciting laser waves, whose independent detection is made easier by the relatively large difference Δ of the corresponding frequencies ω_L and ω_L' in the neighborhood of the resonance: $\Delta \gg \Gamma$. The real and imaginary parts of the corresponding refractive indices are expressed in an obvious way in terms of the operators for the corresponding dipole transitions (assuming that macroscopic volume averaging is valid) as

$$n_k''=\frac{\hbar c g_k N_0}{I_k}\,\mathrm{Im}\langle|k\rangle\langle 3|\rangle, \tag{20}$$

and

$$n_k'-1=\frac{\hbar c g_k N_0}{I_k}\,\mathrm{Re}\langle|k\rangle\langle 3|\rangle, \tag{21}$$

where g_k and I_k are the corresponding Rabi frequencies and intensities of the fields, with $k=1$ corresponding to frequency ω_L and $k=2$ to ω_L' (here $g_1=g$ and $g_2=g'$). To calculate Eqs. (20) and (21) in the stationary case it is necessary to find the stationary density matrix in the rotating wave approximation; this matrix is represented by the corresponding zero eigenvector $\langle 0|$ determined from the equation

$$\langle 0|\mathscr{L}_{RWA}=0.$$

When the contribution of four-photon processes to resonance dephasing is taken into account, the relaxation operator in the expression for $\mathscr{L}_{RWA}$ must include the field-dependent correction (19). Then the results of averaging in Eqs. (20) and (21) with a natural choice of basis for the vector representation of the density matrix (see Appendix A1) are described simply by the corresponding components of the vector $\langle 0|$.

Performing the corresponding analytic calculations and expressing the concentration of active atoms in terms of the pressure p, we obtain

$$n_k'-1=-0.0289p\lambda^3\,\frac{\tilde{\gamma}^2}{\tilde{g}_k^2}\,n_3\left[\tilde{\delta}-\frac{(\tilde{g}_1^2-\tilde{g}_2^2)\tilde{\Gamma}_{12}\tilde{\delta}+\tilde{g}_\Lambda^2\tilde{\delta}_R}{\tilde{g}_\Lambda^2\tilde{\Gamma}_{12}+4\tilde{\Gamma}_{12}^2+4\tilde{\delta}_R^2}\right], \tag{22}$$

and

$$n_k''=0.0289p\lambda^3\,\frac{\tilde{\gamma}^2}{\tilde{g}_k^2}\,n_3. \tag{23}$$

Here n_3 describes the population of the excited state calculated according to the formula

$$n_3=\Bigg[3+\frac{2\tilde{\gamma}\tilde{g}_\Lambda^2}{\tilde{g}_1^2\tilde{g}_2^2}\,(1+\tilde{\delta}^2)+\frac{\tilde{\gamma}}{2} \\ \times\frac{\tilde{g}_\Lambda^4\tilde{\Gamma}_{12}+\tilde{g}_\Lambda^6/4-(\tilde{g}_2^2-\tilde{g}_1^2)^2\tilde{\Gamma}_{12}\tilde{\delta}^2+2\tilde{g}_\Lambda^2(\tilde{g}_2^2-\tilde{g}_1^2)\tilde{\delta}_R\tilde{\delta}}{\tilde{g}_1^2\tilde{g}_2^2(\tilde{\delta}_R^2+\tilde{\Gamma}_{12}^2+\tilde{\Gamma}_{12}\tilde{g}_\Lambda^2/4)}{}_{-1},$$

where the tilde means that the corresponding variables are normalized to Γ. The arguments δ and δ_R depend on the velocity of the atom owing to the single-photon and residual Doppler effect, while the damping Γ_{12} in the lower level system is determined by the reciprocal time of flight of the atom (for a cuvette with pure vapor). Thus, in order to obtain computational data which model the experimental situation, Eqs. (22) and (23) must be averaged over a Maxwellian velocity distribution, which is done numerically.

Figure 5 shows calculated resonance absorption curves for ${}^2S_{1/2}\rightarrow{}^2P_{3/2}$ transitions in cesium and potassium for intense pump and weak probe fields. Although four-photon dephasing is not very important for cesium, in the case of

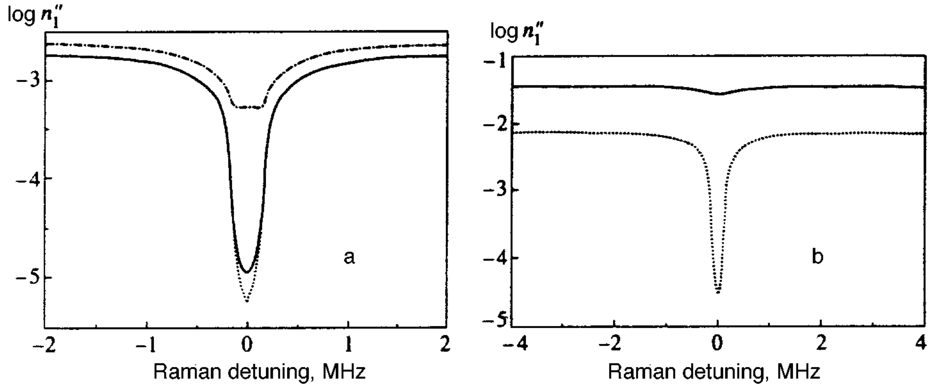

FIG. 5. Absorption resonances in cesium (a) and potassium (b) vapor including (smooth curve) and neglecting (dotted line) four-photon dephasing. The dot-dashed curve in Fig. a corresponds to a calculation taking four-photon dephasing into account where the separation between the lower levels was specially reduced by a factor of 10. The field intensities in the calculations were $I_1 = 0.01$ mW/cm^2 and $I_2 = 10$ mW/cm^2.

potassium, for which the splitting of the ground state for the isotope ^{41}K is only 0.25 GHz, i.e., almost two orders of magnitude smaller than the splitting in cesium, with a pump field intensity of 10 mW/cm^2 the resonance in the absence of a magnetic field is essentially unobservable. Thus, the four-photon mechanism for dephasing of the Λ-resonance establishes a fundamental limit of ''observability'' for the absorption resonance in strong fields, by imposing a limit either on the pump field intensity or on the magnitude of the splitting in the ground state.

6. CONCLUSION

Four-photon interactions, therefore, play a fundamental role in the formation of fluorescence spectra, as well as of the absorption spectra and/or dispersion of a resonance Λ-system during coherent population trapping.

A typical arrangement of a possible experiment for detecting a resonance fluorescence spectrum employing an atomic beam is shown in Fig. 1b. Experiments of this sort using an atomic beam and an atom trap have been described in detail by Gauthier *et al.*[14] and Stalgies *et al.*,[15] respectively. The directions of the atomic and laser beams are chosen to be mutually perpendicular so as to avoid the ordinary Doppler effect. The fluorescence spectrum is analyzed with a Fabry-Perot interferometer. Calculations for the fluorescence spectrum of two-level atoms show that for an atomic beam with 10^9 atoms/s·mm^2, using a 5-millimeter Fabry-Perot cavity with $Q \sim 10^4$ one can expect more than 10^5 photons/s from a volume of diameter $\sim 100\ \mu$m. For a Λ-system, four-photon interactions, on one hand, reduce the fluorescence intensity relative to that of a two-level atoms by a factor of $(g_\Lambda/2\omega_{12})^2$, and, on the other, increase it by a factor of $(\gamma/\Gamma_{12})^{1/2}$. As an example, for the Cs atom a saturation intensity of 1.1 mW/cm and the corresponding parameter $g_\Lambda = 10^2\gamma$ are already achieved for a laser power of 30 mW at a wavelength of 852 nm focussed into a spot with a diameter of about 1 mm. For these experimentally easily realized parameters, we may expect, as the calculations show, a reduction in the scattering intensity of the Λ-system compared to two-level atoms by a factor of 2×10^3 and, therefore, to detect fewer than ~ 100 photons/s, which is not a problem for modern detection systems.

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APPENDIX A

Dynamic superoperator of a Λ-system

Let us consider a Λ-configuration of the quantum mechanical levels of an atom (Fig. 1) acted on by two laser fields with frequencies close to a Raman resonance which is described by a Hamiltonian of the form

$$\hat{\mathscr{H}}_\Lambda = \hat{\mathscr{H}}_a + \hat{\mathscr{H}}_I, \tag{A1}$$

where

$$\hat{\mathscr{H}}_a = -\hbar\omega_{12}|2\rangle\langle 2| + \hbar\omega_{13}|3\rangle\langle 3|$$

is the intrinsic Hamiltonian of the atom (the energy of level 1 is taken to be zero, so that the projection operator $|1\rangle\langle 1|$ is absent in the Hamiltonian) and

$$\hat{\mathscr{H}}_I = \hbar g\ \cos(\omega_L t + \varphi)(|1\rangle\langle 3| + |3\rangle\langle 1|) + \hbar g' \times\cos(\omega_L' t + \varphi')(|2\rangle\langle 3| + |3\rangle\langle 2|)$$

is the Hamiltonian of the interaction of the atomic system with two light fields having frequencies ω_L and ω_L', including the dependence of the excitation on the phase of the field. The interaction constants, i.e., the Rabi frequencies, depend on the amplitudes A_{ω_L} and $A_{\omega_L'}$ of the external field an on the dipole matrix elements d_{13} and d_{23}:

$$g = \frac{1}{\hbar}\, d_{13}A_{\omega_L}, \quad g' = \frac{1}{\hbar}\, d_{23}A_{\omega_L'}. \tag{A2}$$

Only the case when a single-photon resonance is present is of interest, i.e., when ω_L and ω_L' are close, respectively, to ω_{13} and ω_{23}. We can rewrite $\hat{\mathscr{H}}_a$ in the form

$$\hat{\mathscr{H}}_a = \hat{\mathscr{H}}_0 + \hat{\mathscr{H}}_\delta, \tag{A3}$$

where the ‘‘unperturbed’’ Hamiltonian

$$\hat{\mathscr{H}}_0=\hbar(\omega_L|3\rangle\langle 3|-\Delta|2\rangle\langle 2|), \tag{A4}$$

including the biharmonic frequency detuning $\Delta=\omega_L'-\omega_L\approx\omega_{12}$, describes free precession with the two laser frequencies. The ‘‘perturbing’’ Hamiltonian $\hat{\mathscr{H}}_b$ can be written in the form

$$\hat{\mathscr{H}}_\delta=-\hbar\delta|3\rangle\langle 3|+\hbar\delta_R|2\rangle\langle 2|,$$

where

$$\delta=\omega_L-\omega_{13},\quad \delta_R=\omega_L'-\omega_L-\omega_{12}$$

describe the single-photon detuning for the $|1\rangle\to|3\rangle$ transition and the two-photon Raman detuning, respectively. Both detunings can be zero with a suitable choice of laser frequencies.

The dynamics of an atomic system with the Hamiltonian (A3) can be characterized as a combination of fast (owing to $\hat{\mathscr{H}}_0$) and slow (owing to $\hat{\mathscr{H}}_\delta$) precessions, so it is appropriate to shift to a representation of the interaction with the unperturbed unitary transformation

$$\mathscr{U}_0(t)=\exp\left[-\frac{i}{\hbar}\hat{\mathscr{H}}_0 t\right].$$

In the rotating wave approximation[6] we can neglect the rapidly oscillating terms, so that the Hamiltonian (A1) takes the form

$$\hat{\mathscr{H}}_\Lambda=\hat{\mathscr{H}}_\delta+\hat{\mathscr{H}}_p=\hbar[-\delta|3\rangle\langle 3|+\delta_R|2\rangle\langle 2|+(g_\Lambda/2)(|c\rangle\times\langle 3|+\text{H.c.})] \tag{A5}$$

and is the effective Hamiltonian in this approximation. Here we have introduced the bound state ($|c\rangle$) and the unbound state ($|n\rangle$) orthogonal to it:

$$|c\rangle=g_\Lambda^{-1}(ge^{-i\varphi}|1\rangle+g'e^{-i\varphi'}|2\rangle),$$

$$|n\rangle=-g_\Lambda^{-1}(g'e^{-i\varphi}|1\rangle-ge^{-i\varphi'}|2\rangle). \tag{A6}$$

The state $|c\rangle$ is associated with excitation of the level e with an effective coupling constant of $g_\Lambda=\sqrt{g^2+g'^2}$. For zero Raman detuning $(\delta_R=0)$, it is easy to see that the Hamiltonian (A5) describes a two-level system. This can be demonstrated most clearly by substituting the expression $|2\rangle\langle 2|$ corresponding to the inverse transformation (A6) in Eq. (A5):

$$|1\rangle=g_\Lambda^{-1}(g'e^{i\varphi'}|c\rangle+ge^{-i\varphi}|n\rangle),$$

$$|2\rangle=g_\Lambda^{-1}(ge^{i\varphi}|c\rangle-g'e^{-i\varphi'}|n\rangle),$$

which leads to the result

$$\begin{aligned}\hat{\mathscr{H}}_\Lambda=\hbar\Bigg[\left(\delta+\frac{\delta_R g^2}{g_\Lambda^2}\right)|c\rangle\langle c|+\left(\delta+\frac{\delta_R g'^2}{g_\Lambda^2}\right)|n\rangle\langle n|\Bigg]\\ +\hbar\delta_R\frac{gg'}{g_\Lambda^2}[e^{i(\varphi-\varphi')}|c\rangle\langle n|+\text{H.c.}]\\ +\frac{\hbar g_\Lambda}{2}(|c\rangle\langle 3|+\text{H.c.}).\end{aligned}$$

In the basis $\{|3\rangle,|c\rangle,|n\rangle\}$ the corresponding matrix has the form

$$\hat{\mathscr{H}}_\Lambda=\begin{pmatrix}0 & \hbar g_\Lambda/2 & 0\\ \hbar g_\Lambda/2 & \hbar\delta & 0\\ 0 & 0 & \hbar\delta\end{pmatrix}+\frac{\hbar\delta_R}{g_\Lambda^2}\begin{pmatrix}0 & 0 & 0\\ 0 & g^2 & gg'e^{i(\varphi-\varphi')}\\ 0 & gg'e^{-i(\varphi-\varphi')} & g'^2\end{pmatrix}$$

and, for $\delta_R=0$, can quickly be expanded in terms of the 2×2 matrix of a two-level system ‘‘dressed’’ with the atomic field and the 1×1 matrix of a single unbound state, i.e., the excited and bound states form an effective two-level system $|e\rangle\oplus|c\rangle$. For simplicity we redefine $|1\rangle$ as $\exp(-i\varphi)|1\rangle$ and $|2\rangle$ as $\exp(-i\varphi')|2\rangle$, so that we can rewrite Eq. (A6) in the form

$$|c\rangle=g_\Lambda^{-1}(g|1\rangle+g'|2\rangle),\quad |n\rangle=-g_\Lambda^{-1}(g'|1\rangle-g|2\rangle),$$

which does not contain the phase factors explicitly.

With this representation of the Hamiltonian in the rotating wave approximation, the corresponding dynamic part of the Liouvillian has the form

$$\mathscr{L}_\Lambda=\frac{i}{\hbar}[\hat{\mathscr{H}}_\Lambda,\odot]. \tag{A7}$$

The complete Liouvillian $\mathscr{L}_{RWA}$ also contain a relaxation operator which is specified phenomenologically here.

A1. Transformation of the Liouvillian in the rotating wave approximation

The initial representation of the Liouvillian in the rotating wave approximation is a matrix in the nonhermitian basis $\{\hat{e}_k\}=\hat{P}_{\alpha,\beta}=|\alpha\rangle\langle\beta|$, where $k=(\alpha,\beta)$ and $\alpha,\beta=1,2,3$, which can be represented by the following complex matrix elements using Eqs. (A7):

$$\mathscr{L}_{RWA}=\begin{pmatrix} -\gamma-\gamma' & \gamma & \gamma' & 0 & 0 & -\frac{ig}{2} & \frac{ig}{2} & -\frac{ig'}{2} & \frac{ig'}{2} \\ 0 & -\gamma_{12} & \gamma_{12} & 0 & 0 & \frac{ig}{2} & -\frac{ig}{2} & 0 & 0 \\ 0 & w & -w & 0 & 0 & 0 & 0 & \frac{ig'}{2} & -\frac{ig'}{2} \\ 0 & 0 & 0 & i\delta_R-\Gamma_{12} & 0 & \frac{ig'}{2} & 0 & 0 & -\frac{ig}{2} \\ 0 & 0 & 0 & 0 & -i\delta_R-\Gamma_{12} & 0 & -\frac{ig'}{2} & \frac{ig}{2} & 0 \\ -\frac{ig}{2} & \frac{ig}{2} & 0 & \frac{ig'}{2} & 0 & -i\delta-\Gamma_{13} & 0 & 0 & 0 \\ \frac{ig}{2} & -\frac{ig}{2} & 0 & 0 & -\frac{ig'}{2} & 0 & i\delta-\Gamma_{13} & 0 & 0 \\ -\frac{ig'}{2} & 0 & \frac{ig'}{2} & 0 & \frac{ig}{2} & 0 & 0 & -i\delta-\Gamma_{23} & 0 \\ \frac{ig'}{2} & 0 & -\frac{ig'}{2} & -\frac{ig}{2} & 0 & 0 & 0 & 0 & i\delta-\Gamma_{23} \end{pmatrix}.$$

For converting to the more convenient Hermitian bases, we can introduce two transformations V_c and V_{c1} of the form

$$V_c=\begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & g/g_\Lambda & 0 & g'/g_\Lambda & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & g/g_\Lambda & 0 & g'/g_\Lambda \\ 0 & 0 & 0 & 0 & 0 & -g'/g_\Lambda & 0 & g/g_\Lambda & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -g'/g_\Lambda & 0 & g/g_\Lambda \end{pmatrix},$$

$$V_{c1}=\begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1/\sqrt{2} & 1/\sqrt{2} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -i/\sqrt{2} & i/\sqrt{2} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1/\sqrt{2} & 1/\sqrt{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -i/\sqrt{2} & i/\sqrt{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1/\sqrt{2} & 1/\sqrt{2} \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -i/\sqrt{2} & i/\sqrt{2} \end{pmatrix}. \qquad \text{(A8)}$$

The transformation V_c introduces two pairs of polarization operators $\hat{P}_c, \hat{P}_c^+$ and $\hat{P}_n, \hat{P}_n^+$ for transitions to the excited level from the bound and unbound states, while V_{c1} introduces the Hermitian cosine-sine operators (analogs of the coordinates and momenta or the Pauli matrices $\hat{\sigma}_1$, $\hat{\sigma}_2$ in a two-level system):

$$\hat{q}_g=(\hat{P}_{12}+\hat{P}_{21})/\sqrt{2}, \quad \hat{p}_g=-i(\hat{P}_{12}-\hat{P}_{21})/\sqrt{2},$$

$$\hat{q}_c=(\hat{P}_c+\hat{P}_c^+)/\sqrt{2}, \quad \hat{p}_c=-i(\hat{P}_c-\hat{P}_c^+)/\sqrt{2},$$

$$\hat{q}_n=(\hat{P}_n+\hat{P}_n^+)/\sqrt{2}, \quad \hat{p}_n=-i(\hat{P}_n-\hat{P}_n^+)/\sqrt{2}. \qquad \text{(A9)}$$

Here the subscripts g, c, and n correspond to the ground $(1\leftrightarrow 2)$, bound $(c\leftrightarrow 3)$, and unbound $(n\leftrightarrow 3)$ subsystems.

After applying the transformations (A8) to the Liouvillian $\mathscr{L}_{RWA}$ in the rotating wave approximation, for the transformed operator $L_{RWA}=V_{c1}V_cL_{RWA}V_c^{-1}V_{c1}^{-1}$ we obtain

$$\mathscr{L}_{RWA}=\left(\begin{array}{ccc|cc|cc|cc}
-\gamma-\gamma' & \gamma & \gamma' & 0 & 0 & 0 & \frac{g_\Lambda}{\sqrt{2}} & 0 & 0 \\
0 & -\gamma_{12} & \gamma_{12} & 0 & 0 & 0 & -\frac{\xi^2 g_\Lambda}{\sqrt{2}} & 0 & \frac{\xi\eta g_\Lambda}{\sqrt{2}} \\
0 & w & -w & 0 & 0 & 0 & -\frac{\eta^2 g_\Lambda}{\sqrt{2}} & 0 & -\frac{\xi\eta g_\Lambda}{\sqrt{2}} \\
\hline
0 & 0 & 0 & -\Gamma_{12} & -\delta_R & 0 & -\xi\eta g_\Lambda & 0 & -(\xi^2-\eta^2)\,\frac{g_\Lambda}{2} \\
0 & 0 & 0 & \delta_R & -\Gamma_{12} & 0 & 0 & -\frac{g_\Lambda}{2} & 0 \\
\hline
0 & 0 & 0 & 0 & 0 & -\Gamma_c & \delta & \Delta\Gamma & 0 \\
-\frac{g_\Lambda}{\sqrt{2}} & \frac{\xi^2 g_\Lambda}{\sqrt{2}} & \frac{\eta^2 g_\Lambda}{\sqrt{2}} & \xi\eta g_\Lambda & 0 & -\delta & -\Gamma_c & 0 & \Delta\Gamma \\
\hline
0 & 0 & 0 & 0 & \frac{g_\Lambda}{2} & \Delta\Gamma & 0 & -\Gamma_n & \delta \\
0 & -\frac{\xi\eta g_\Lambda}{\sqrt{2}} & \frac{\xi\eta g_\Lambda}{\sqrt{2}} & (\xi^2-\eta^2)\,\frac{g_\Lambda}{2} & 0 & 0 & \Delta\Gamma & -\delta & -\Gamma_n
\end{array}\right), \qquad \text{(A10)}$$

where we have used the following notation

$$\xi=g/g_\Lambda\,, \quad \eta=g'/g_\Lambda\,,$$

$$\Gamma_c=\xi^2\Gamma_{13}+\eta^2\Gamma_{23}\,, \quad \Gamma_n=\eta^2\Gamma_{13}+\xi^2\Gamma_{23}\,,$$

$$\Delta\Gamma=\xi\eta(\Gamma_{13}-\Gamma_{23}).$$

The block structure of the transformed dynamic superoperator L_{RWA} indicated by the continuous lines in Eq. (A10) is discussed in more detail in Appendix A2.

As opposed to the initial complex representation, the transformed operator $\mathscr{L}_{RWA}$ has real matrix elements, since it corresponds to the Hermitian basis $\{\hat{e}_k\}$.

For a symmetric Λ-system, with $\Gamma_{13}=\Gamma_{23}$, $g=g'$, and, therefore, $\Delta\Gamma=0$ and $\xi=\eta$, Eq. (A10) takes the form

$$L_{RWA}=\left(\begin{array}{ccc|cc|cc|cc}
-\gamma-\gamma' & \gamma & \gamma' & 0 & 0 & 0 & g_\Lambda/\sqrt{2} & 0 & 0 \\
0 & -\gamma_{12} & \gamma_{12} & 0 & 0 & 0 & -g_\Lambda/2\sqrt{2} & 0 & g_\Lambda/2\sqrt{2} \\
0 & w & -w & 0 & 0 & 0 & -g_\Lambda/2 & 0 & -g_\Lambda/2\sqrt{2} \\
\hline
0 & 0 & 0 & -\Gamma_{12} & -\delta_R & 0 & -g_\Lambda/2 & 0 & 0 \\
0 & 0 & 0 & \delta_R & -\Gamma_{12} & 0 & 0 & -g_\Lambda/2 & 0 \\
\hline
0 & 0 & 0 & 0 & 0 & -\Gamma & \delta & 0 & 0 \\
-g_\Lambda/\sqrt{2} & g_\Lambda/2\sqrt{2} & g_\Lambda/2 & g_\Lambda/2 & 0 & -\delta & -\Gamma & 0 & 0 \\
\hline
0 & 0 & 0 & 0 & g_\Lambda/2 & 0 & 0 & -\Gamma & \delta \\
0 & -g_\Lambda/2\sqrt{2} & g_\Lambda/2\sqrt{2} & 0 & 0 & 0 & 0 & -\delta & -\Gamma
\end{array}\right).$$

A2. The block structure of the dynamic superoperator in the rotating wave approximation

The physical significance of the superoperator L_{RWA} defined by Eq. (A10) becomes most transparent on examining its block structure. It is convenient to break the matrix (A10) up into blocks in accordance with a definite set of polarization variables (A9). By analogy, the matrix blocks can be numbered with a subscript p corresponding to the populations and by the subscripts g, c, and n corresponding to the polarizations of ground $(1\leftrightarrow 2)$, bound $(1\leftrightarrow 3)$, and unbound $(1\leftrightarrow 2)$ subsystems. In this notation, the matrix (A10) appears as

$$L_{RWA}=\begin{pmatrix} L_{pp} & 0 & -L_{cp}^T & -L_{np}^T \\ 0 & L_{gg} & -L_{cg}^T & -L_{ng}^T \\ L_{cp} & L_{cg} & L_{cc} & -L_{nc}^T \\ L_{np} & L_{ng} & L_{nc} & L_{nn} \end{pmatrix}. \tag{A11}$$

It consists of nine nonzero independent blocks. The diagonal block L_{pp} describes the dynamics of the populations n_3, n_c, and n_n, and the blocks L_{gg}, L_{cc}, and L_{nn}, the polarization dynamics, respectively, of the ground state and of the bound and unbound subsystems. The five nonzero nondiagonal matrices describe the dynamics of the Λ-system owing to coupling among the above basis variables. The antisymmetry of these five blocks is a consequence of the purely oscillatory character of the dynamics resulting from the interaction with the external field, while the inner dynamics includes relaxation so it is also represented by the matrix elements which yield nonzero real components in the eigenvalues of the matrix $\mathscr{L}_{RWA}$.

It is easy to see from the block structure of (A11) that there is no connection between the populations and polarizations of the ground state, since $\mathscr{L}_{gp}=0$. This reflects the fact that the exciting field acts directly only on transitions into the excited state, while single-photon excitation of the ground state is absent.

The block representation given here for the dynamic superoperator in the rotating wave approximation is convenient for qualitative discussions of the effect of the parameters of the Λ-system on its dynamics, since it reduces to changes in only the inner structure of the blocks in the representation (A11).

APPENDIX B

Superoperator calculation of the general formula for the fine structure of the spectrum

Let us calculate the two-time correlation function (10) that determines the atomic fluorescence spectrum:

$$\mathscr{K}(\tau)=\langle\hat{\rho}_0 S(0,t)|\hat{\sigma}^-(t)[S(t,t+\tau)\hat{\sigma}^+(t+\tau)]\rangle. \tag{B1}$$

Here the total evolution superoperator (see Eq. (2)) has the form

$$S(0,t)=S_{RWA}(t)\widetilde{S}_0(t), \tag{B2}$$

where $S_{RWA}(t)$ is the superoperator in the rotating wave approximation and $\widetilde{S}_0(t)$ is the superoperator for the perturbed evolution owing to the nonresonant interaction. The superoperator $\widetilde{S}_0(t)$ describes the transformation of an initial system Hamiltonian $\hat{\mathscr{H}}(t)$ of the form

$$\hat{\mathscr{H}}(t)\rightarrow\hat{\mathscr{H}}_0+\delta\hat{\mathscr{H}}(t)=\hat{\mathscr{H}}_0+\frac{\hbar g_\Lambda}{2}(\hat{\tau}e^{i\Delta t}+\hat{\tau}^+e^{-i\Delta t}), \tag{B3}$$

where the operator $\hat{\tau}$ is defined by Eq. (9). In first-order perturbation theory we can introduce a superoperator $\widetilde{S}_0(t)$, corresponding to the Hamiltonian (B3), in the form

$$\widetilde{S}_0(t)=S_0(t)+\delta S_0(t)=S_0(t)+\int_0^t \delta\mathscr{L}_p(\tau)d\tau S_0(t),$$

where the perturbation Liouvillian has the form

$$\delta\mathscr{L}_p(\tau)=\frac{i}{\hbar}[\delta\hat{\mathscr{H}}(\tau),\odot]. \tag{B4}$$

Integrating $\delta S_0(t)$ with respect to τ and using Eq. (B4) together with Eq. (B3), we obtain

$$\delta S_0(t)=\frac{g_\Lambda}{2\Delta}[\hat{\tau}e^{i\Delta t}-\hat{\tau}^+e^{-i\Delta t},\odot]S_0(t), \tag{B5}$$

which describes oscillations at a frequency Δ.

The superoperator $S_0(t)$ in Eq. (B5) describes the unperturbed dynamics represented by the Hamiltonian (A4), which accounts for the free precession of all the Λ-system transitions. The latter is represented in the form

$$\begin{aligned} S_0(t)=&S_{12}^+e^{-i\Delta t}\oplus S_{12}^+e^{i\Delta t}\oplus S_{13}^+e^{-i\omega_L t}\oplus S_{13}^+e^{i\omega_L t}\\ &\oplus S_{23}^+e^{-i\omega_L' t}\oplus S_{23}^+e^{i\omega_L' t}\oplus P_0, \end{aligned} \tag{B6}$$

where the matrices $S_{kl}^\pm$ for the corresponding superoperators are the one-dimensional eigen-projectors on the corresponding intrinsic precession of the variables and P_0 is the projector on the three-dimensional subspace of the nonoscillatory variables, i.e., the populations. After substituting Eq. (B6) in Eq. (B5), we obtain

$$\begin{aligned} \delta S_0(t)=\frac{g\Lambda}{2\Delta}\{&[\hat{\tau},\odot]S_{13}^+\exp[-i(\omega_L-\Delta)t]\\ &-[\hat{\tau}^+,\odot]S_{23}^+\exp[-i(\omega_L'+\Delta)t]\\ &-[\hat{\tau}^+,\odot]S_{13}^-\exp[i(\omega_L-\Delta)t]\\ &+[\hat{\tau},\odot]S_{23}^-\exp[i(\omega_L'+\Delta)t]\}. \end{aligned} \tag{B7}$$

Then substituting Eq. (B7) into Eq. (B2) and using Eq. (B1) together with the relation $\hat{\rho}_0 S(0,t)\rightarrow\langle 0|$ for $t\rightarrow\infty$ because of the damping of all the eigen-oscillations corresponding to the nonzero eigenvalues, we can finally write the correlation function in the form

$$\mathscr{K}(t,t+\tau)=\langle 0|\delta S_0(t)\hat{\sigma}^-\cdot\exp(\mathscr{L}_{RWA}\tau)\delta S_0(t+\tau)\hat{\sigma}^+\rangle, \tag{B8}$$

where the symbol "$\cdot$" denotes the product of transformed operators and $\sigma^\pm=\sigma_{13}^\pm+\sigma_{23}^\pm$ is the sum of the complex amplitudes oscillating at the optical frequencies.

If we then apply δS_0 to $\hat{\sigma}^\pm$ and recall that $\hat{\sigma}_{13}^\pm$ and $\hat{\sigma}_{23}^\pm$ are eigenvectors for the eigen-projectors $S_{13}^\pm$ and $S_{23}^\pm$, we obtain

$$\begin{aligned} \delta S_0(t)\hat{\sigma}^-=-\frac{g_\Lambda}{2\Delta}\{&[\hat{\tau}^+,\hat{\sigma}_{13}^-]\exp[i(\omega_L-\Delta)t]\\ &+[\hat{\tau},\hat{\sigma}_{23}^-]\exp[i(\omega_L'+\Delta)t], \end{aligned} \tag{B9}$$

and

$$\begin{aligned} \delta S_0(t+\tau)\hat{\sigma}^+=\frac{g_\Lambda}{2\Delta}\{&[\hat{\tau},\hat{\sigma}_{13}^+]\exp[-i(\omega_L-\Delta)(t+\tau)]\\ &+[\hat{\tau}^+,\hat{\sigma}_{23}^+]\exp[-i(\omega_L'+\Delta)(t+\tau)]\}, \end{aligned} \tag{B10}$$

where the commutators of the complex amplitudes of the $1\leftrightarrow 3$ and $2\leftrightarrow 3$ transitions with $\hat{\tau}$ and $\hat{\tau}^{+}$ are given by

$$[\tau^{+},\hat{\sigma}_{13}^{-}]=-\frac{\hat{\sigma}_{12}^{-}}{\sqrt{2}},\quad [\tau,\hat{\sigma}_{23}^{-}]=-\frac{\hat{\sigma}_{12}^{+}}{\sqrt{2}},$$

$$[\tau,\hat{\sigma}_{13}^{+}]=\frac{\hat{\sigma}_{12}^{+}}{\sqrt{2}},\quad [\tau^{+},\hat{\sigma}_{23}^{+}]=\frac{\hat{\sigma}_{12}^{-}}{\sqrt{2}}.$$

After substitution of Eqs. (B9) and (B10) in Eq. (B8) and leaving out terms which oscillate relative to t, with the dynamical representation in the rotating wave approximation in terms of the eigenvectors and corresponding eigenvalues Eq. (B8) finally takes the form of Eq. (12).

APPENDIX C

Transformation of the time evolution superoperator in the rotating wave approximation

For a Λ-system with a time-independent Hamiltonian $\hat{\mathscr{H}}$, the time evolution superoperator is unitary and is given by an exponential $\mathscr{S}(t)=\exp(\mathscr{L}_{\mathscr{H}}t)$ with a purely dynamic Liouvillian of the type (A7) and can be written in the form

$$\mathscr{L}(t)=\mathscr{U}(t)\odot\mathscr{U}^{-1}(t)=\sum_{\alpha,\beta}\exp[-i(\omega_{\alpha}-\omega_{\beta})t]|\alpha\rangle\langle\alpha|\odot|\beta\rangle\langle\beta|, \tag{C1}$$

where the ω_{α} and $|\alpha\rangle$ are the Bohr eigenfrequencies and the corresponding eigenvectors of the Hamiltonian, while the unitary transformation $\mathscr{U}(t)$ is specified by the relation

$$\mathscr{U}(t)=\exp\left[-\frac{i}{\hbar}\hat{\mathscr{H}}t\right].$$

Subsequently, we can use the interaction representation for the superoperator $\mathscr{L}_{\mathscr{H}}+\mathscr{L}_{r}$, which differs from the purely dynamical Liouvillian corresponding to $\mathscr{S}(t)$ in that it includes the relaxation superoperator $\mathscr{L}_{r}$, and treat $\mathscr{L}_{\mathscr{H}}$ as the Liouvillian for the unperturbed time evolution $\mathscr{S}(t)$. In the interaction representation the time dependent relaxation superoperator has the form

$$\mathscr{L}_{r}^{I}(t)=\sum_{\alpha,\beta;\mu,\nu}\exp\{i[(\omega_{\alpha}-\omega_{\beta})-(\omega_{\mu}-\omega_{\nu})]t\}L_{\alpha\beta,\mu\nu}|\alpha\rangle\langle\mu|\odot|\nu\langle\beta|, \tag{C2}$$

where

$$L_{\alpha\beta,\mu\nu}=\sum_{k,l;m,n}\langle\alpha|k\rangle\langle l|\beta\rangle L_{kl,mn}\langle m|\mu\rangle\langle\nu|n\rangle$$

denotes the matrix elements of the relaxation superoperator in terms of the eigen-basis of the Hamiltonian $\hat{\mathscr{H}}$.

Let us consider the important special case in which the oscillations in Eq. (C.2) are fast compared to the rates of all the relaxation processes, so that it is possible to average over these oscillations. Then the resulting effective (''reduced'') relaxation operator has the form

$$\mathscr{L}_{re}=\sum_{\alpha,\beta}L_{\alpha\alpha,\beta\beta}|\alpha\rangle\langle\beta|\odot|\beta\rangle\langle\alpha| + \sum_{\alpha\neq\beta}L_{\alpha\beta,\alpha\beta}|\alpha\rangle\langle\alpha|\odot|\beta\rangle\langle\beta|, \tag{C3}$$

where it is assumed that all the frequencies $\omega_{\alpha\beta}$ corresponding to the $\beta\rightarrow\alpha$ atomic transitions ($\alpha\neq\beta$) are different. The first term in Eq. (C.3) describes the relaxation of the populations owing to $\beta\rightarrow\alpha$ transitions from other levels ($\beta\neq\alpha$) and radiative decay ($\beta=\alpha$). The second term describes the relaxation of the polarization variables. The corresponding matrix is $n\times n$, where $n=3$ is the number of levels in a Λ-system.

The superoperator (C3) commutes with the dynamical Liouvillian, since they have an eigenbasis in common. Given this circumstance, the relaxation of the atomic oscillations is simply described by the corresponding damping rates

$$\Gamma_{\alpha\beta}=-\operatorname{Re}L_{\alpha\beta,\alpha\beta}.$$

If these quantities are all nonzero, then the stationary (zero-) vector $\langle\hat{\rho}^{st}|$ has nonzero components only in population relaxation space and is actually described by the n-component zero-vector ρ_{α}^{st} of the $n\times n$ submatrix $L_{\alpha\alpha,\beta\beta}$.

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