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Theory of time-dependent intense-field collisional resonance fluorescence

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The time-dependent theory of Courtens and Szöke is generalized using the approach of Burnett *et al.* to derive time-dependent spectral intensities of resonance fluorescence from atoms driven by a pulsed laser in the presence of collisions. These results are valid both for laser detunings inside and outside the usual impact region of the spectrum, including Zeeman degeneracy effects. We apply this theory to a simple but important example ($J=0$ to $J=1$) to obtain quantitative predictions for the observable scattered-light spectrum which can be directly compared with recent experiments.

I. INTRODUCTION

In a previous paper[1] (hereinafter paper I) we developed a formal theory of collisional redistribution valid for arbitrary laser field strengths and detunings both inside and outside the "impact" region, and which included the effects of Zeeman degeneracy. We required the validity of the binary collision approximation (BCA), the secular and the steady-state approximations, and assumed we were only interested in *scattered*-light intensities in the "impact" region (with respect to the dressed atom) of the scattered spectrum. We found, under these conditions, that the master equation for the reduced density matrix in the dressed-atom basis has a Markoffian form, and that the dressed-level populations can be obtained from simple rate equations (the collision rates being evaluated by an S-matrix calculation in the dressed frame). Furthermore, for scattering in the "impact" region the quantum fluctuation regression "theorem" holds and the correlation function satisfies the same equation of motion as the reduced density operator.

This theory applies to the steady state (as do most theories of the near-resonant collisional redistribution of light).[1-4] Many experiments in this field, however, require the use of intense pulsed lasers.[5,6] This is often a blessing since it can enable one to study kinetics at intensities that would create complete ionization in the steady state. Moreover, the development of a spectrum in the time domain is an interesting problem in its own right, and its solution, as we shall see, presents us with a most useful probe of the collision dynamics in the presence of intense laser fields. In this paper we generalize this theory to include the effects of time-dependent (pulsed) laser excitation using an adiabatic approximation.

A theory of time-dependent spectra has been developed by Courtens and Szöke[7] (CS) and our work is an elaboration and extension of the work of these two authors (see also Eberly and Wodkiewicz[8]). The work of CS (Ref. 7) was based on a two-state atom and used the Bloch-vector approach to evaluate the integrated intensities of the scattered features in the usual impact regime (with respect to the atomic states). The latter regime excludes any frequency or intensity dependence of the optical collision cross sections—precisely the type of effects that we wish to study. In the impact approximation they obtained an equation of motion for the dressed-frame Bloch vector, as well as dressed-frame damping rates, by a unitary transformation of the impact theory, atomic frame, variables. (The emission intensities were derived from a somewhat heuristic Fermi golden rule argument.) Those results are then expected to be valid for $\Delta,\Omega \ll \tau_c^{-1}$ where $\Delta=\omega_L-\omega_0$ is the detuning of the laser frequency ω_L from the atomic transition frequency ω_0 and τ_c is the duration of a typical collision.

Here we use the approach and notation of paper I (but with explicit emphasis on the time dependence

of the laser pulse) to rigorously justify and clarify some of the assumptions of CS,[7] and to generalize their results to the nonimpact regime. Specifically we derive a Markoffian master equation for the density matrix in the dressed frame with time-dependent relaxation rates and consider under what conditions it reduces to rate equations for the dressed-atom populations. Using the definition of the "physical" time-dependent spectrum developed by Eberly and Wodkiewicz,[8] and independently by Courtens and Szöke,[7] we write the observable spectrum in terms of the dipole correlation function. We then derive an equation of motion for the correlation function and discuss the validity of the quantum fluctuation regression "theorem" in this time-dependent case.

Finally, we choose a simple but important example, a $J_g=0$ to $J_e=1$ transition (where g denotes the ground state and e the excited state), and solve the equations explicitly to obtain quantitative predictions for the observable scattered-light intensities which can be directly compared with recent experiments.[9]

II. MASTER EQUATION

We consider an atom with ground and excited levels having angular momenta J_g and J_e, respectively, being driven by a laser with classical electric field amplitude $\mathscr{E}_0(t)$ and detuning $\Delta=\omega_L-\omega_0$. $\mathscr{E}_0(t)$ is in general a complex time-varying amplitude of the form

$$\mathscr{E}(t)=E_0(t)e^{i\phi(t)}\,, \tag{1}$$

where $E_0(t)$ is a "slowly varying" (adiabatic) pulse envelope and $\phi(t)$ is a fluctuating phase.

Following paper I we transform the Hamiltonian [paper I, Eq. (9)] to a "rotating frame" by the unitary transformation

$$\hat{U}_R(t)=\exp\{i[\omega_L t+\phi(t)]\tfrac{1}{2}(\hat{S}_A^z-1)\}\,, \tag{2}$$

where $\hat{S}_A^z$ is the inversion operator. Note here that $\hat{U}_R$ explicitly includes the phase-varying term. In the rotating-wave approximation (RWA) the rotating-frame Hamiltonian is

$$\hat{H}^{\rm rot}_{\rm atom/rad}(t)=-\tfrac{1}{2}\hbar\hat{S}_A^z[\Delta+\dot{\phi}(t)] +\tfrac{1}{2}\hbar(\omega_0+\omega_L) -[\hat{D}_A^+E_0(t)+\hat{D}_A^-E_0(t)]\,. \tag{3}$$

The $\dot{\phi}(t)$ term arises from laser bandwidth, and provided there are no Fourier components of the laser which overlap the absorption line (i.e., provided $\dot{\phi}$ is slowly varying such that $\dot{\phi}\ll\Delta$) this term can be neglected and we write the Hamiltonian as [paper I, Eq. (12)]

$$\hat{H}^{\rm rot}_{\rm atom/rad}(t)=-\tfrac{1}{2}\hbar\hat{S}_A^z\Delta+\tfrac{1}{2}\hbar(\omega_0+\omega_L) -[\hat{D}_A^+E_0(t)+\hat{D}_A^-E_0(t)]\,. \tag{4}$$

Here again $E_0(t)$, and hence $\hat{H}^{rot}_{\rm atom/rad}(t)$, is time dependent. [The effect of fast (nonadiabatic) phase fluctuations which give rise to Fourier components in the laser wings that may overlap the absorption line can be included later by perturbation theory if they are "small."] The inversion operator $\hat{S}_A^z$ and dipole operator $\hat{D}_A^{\pm}$ are defined as in paper I:

$$\hat{S}_A^z=\sum_{m_e m_g}(|m_e\rangle\langle m_e|-|m_g\rangle\langle m_g|)\,, \tag{5}$$

$$\hat{D}_A^+=\sum_{m_e m_g}|m_e\rangle\langle m_e|\vec{d}\cdot\vec{\epsilon}_L|m_g\rangle\langle m_g|\,, \tag{6}$$

$$\hat{D}_A^-=\sum_{m_e m_g}|m_g\rangle\langle m_g|\vec{d}\cdot\vec{\epsilon}_L^{\,*}|m_e\rangle\langle m_e|\,, \tag{7}$$

where m_e and m_g are angular momentum projections associated with the excited and ground states, respectively, and $\vec{\epsilon}_L$ is the laser polarization vector.

We define the time-dependent Rabi frequency for any two states that are coupled by the field, thus

$$\Omega(t)=\frac{E_0(t)}{\hbar}\langle m_e|\vec{d}\cdot\vec{\epsilon}_L|m_g\rangle \tag{8}$$

and the corresponding generalized Rabi frequency, thus

$$\Omega'(t)\equiv[\Delta^2+\Omega^2(t)]^{1/2}\,, \tag{9}$$

where Ω' has the sign of Δ. Following Courtens and Szöke[7] we transform Eq. (4) to the dressed frame:

$$\hat{H}_{\rm D}(t)\equiv\hat{H}_{\rm AD}(t)+\hat{\dot{\theta}}(t) \tag{10}$$

with

$$\hat{H}_{\rm AD}(t)=\hat{U}_D(t)\hat{H}^{\rm rot}_{\rm atom/rad}(t)\hat{U}_D^{\dagger}(t) \tag{11}$$

and

$$\hat{\dot{\theta}}(t)=i\frac{\partial\hat{U}_D(t)}{\partial t}\hat{U}_D^{\dagger}(t)\,. \tag{12}$$

Here, $\hat{U}_D(t)$ is the unitary transformation which diagonalizes $\hat{H}^{\rm rot}_{\rm atom/rad}(t)$, at each instant t, and $\hat{H}_{\rm AD}(t)$ is the adiabatically diagonal Hamiltonian of the atom-laser system [i.e., $\hat{H}_{\rm AD}(t)$ is diagonal when $\alpha(t)\equiv\dot{\Omega}(t)/\Omega(t)\equiv0$], and $\hat{\dot{\theta}}$ (see below) is an *off-*

diagonal contribution proportional to $\alpha(t)$ and due to the time-changing field amplitude.

The *adiabatic* eigenstates (dressed states) $\hat{H}^{\rm rot}_{\rm atom/rad}(t)$ are expressed in terms of the rotating-frame states $|a\rangle$ and $|b\rangle$ as

$$|{\rm II}\rangle = b_2(t)|a\rangle + b_1(t)|b\rangle\ , \tag{13}$$

$$|{\rm I}\rangle = b_1(t)|a\rangle - b_2(t)|b\rangle \tag{14}$$

with

$$b_1(t) = \left[\frac{\Omega'(t)+\Delta}{2\Omega'(t)}\right]^{1/2},$$

$$b_2(t) = \left[\frac{\Omega'(t)-\Delta}{2\Omega'(t)}\right]^{1/2}, \tag{15}$$

and for rotating-frame states $|c\rangle$ which are not coupled to the laser

$$|c\rangle_D = |c\rangle\ . \tag{16}$$

The adiabatic transformation $\hat{U}_D(t)$ can be written explicitly in terms of the rotating-frame states as

$$\begin{aligned}
&\langle a|\hat{U}_D(t)|a\rangle = \langle b|\hat{U}_D(t)|b\rangle = b_1(t)\ ,\\
&\langle a|\hat{U}_D(t)|b\rangle = -\langle b|\hat{U}_D(t)|a\rangle = b_2(t)\ ,\\
&\langle c|\hat{U}_D(t)|d\rangle = \delta_{cd}\ ,
\end{aligned} \tag{17}$$

where $|d\rangle$ is any state (coupled to the laser or not). In addition, for the dressed states $|{\rm I}\rangle$ and $|{\rm II}\rangle$ we have explicitly

$$\langle {\rm I}|\hat{\dot{\theta}}(t)|{\rm II}\rangle = -\frac{1}{2}\left[\frac{\Omega(t)\Delta}{\Omega'(t)^2}\right]\dot{\alpha}(t)\ . \tag{18}$$

(Note $\langle {\rm I}|\hat{\dot{\theta}}|{\rm I}\rangle = \langle {\rm II}|\hat{\dot{\theta}}|{\rm II}\rangle \equiv 0$.) The semiclassical dressed levels have adiabatic energies

$$\begin{aligned}
E_{\rm II} &= \hbar\omega_0 - \hbar\delta_D(t)\ ,\\
E_{\rm I} &= \hbar\omega_L + \hbar\delta_D(t)\ ,
\end{aligned} \tag{19}$$

where $\delta_D(t) \equiv \frac{1}{2}[\Omega'(t)-\Delta]$ is the ac Stark shift. Note the eigenstates Eqs. (13)–(16) as well as the energies (19) are now functions of time, defined in terms of the instantaneous Rabi frequency. [A quantized laser-field treatment gives a ladder of dressed doublets, the rungs on the ladder separated by the energy corresponding to an integral number (n) of photons $n\hbar\omega_L$.[10] The limit of large photon numbers (high intensities) corresponds to the classical field results used here.]

We write the equation of motion density matrix for the complete system [paper I, Eq. (13)] of atom plus perturbers *in the dressed frame* in the form

$$\partial_t\hat{\rho}_D(t) = [\tilde{L}^A_D(t) + \tilde{V}_D(t) + \tilde{L}_p + \tilde{\dot{\theta}}(t)]\hat{\rho}_D(t)\ , \tag{20}$$

where [as paper I, Eqs. (14)–(18)] the Liouville superoperators are defined by

$$\tilde{L}^A_D(t)\hat{\rho}_D = -i[\hat{H}_{\rm AD}(t),\hat{\rho}_D]\ , \tag{21}$$

$$\tilde{L}_p\hat{\rho}_D = -i[\hat{H}_p,\hat{\rho}_D]\ , \tag{22}$$

$$\tilde{V}_D(t)\hat{\rho}_D = -i[\hat{V}_D(t),\hat{\rho}_D]\ , \tag{23}$$

$$\tilde{\dot{\theta}}(t)\hat{\rho}_D = -i[\hat{\dot{\theta}}(t),\hat{\rho}_D]\ . \tag{24}$$

Here, $\hat{H}_p$ is the Hamiltonian for the free perturbers and $\hat{V}_D$ is the interaction between atom and perturbers (the perturber-perturber interaction is ignored) in the dressed-state basis, i.e.,

$$\begin{aligned}
\hat{V}_D(t) &= \hat{U}_D(t)\hat{U}_R(t)\left[\sum_l \hat{V}_{Al}\right]\hat{U}_R^{-1}(t)\hat{U}_D^{-1}(t)\\
&= \hat{U}_D(t)\sum_l \hat{V}_{Al}\hat{U}_D^{-1}(t)\ .
\end{aligned} \tag{25}$$

(As in paper I, $\hat{V}_{Al}$ is the collisional interaction between the atom and the lth perturber.) The last form follows if collisions do not directly couple upper and lower states. Note that, due to $\hat{U}_D(t)$, $\hat{V}_D(t)$ is now explicitly time dependent. The derivation continues exactly as in paper I with the replacement $\tilde{L}^A_D \to \tilde{L}^A_D(t) + \tilde{\dot{\theta}}(t)$ and also $\tilde{V}_D \to \tilde{V}_D(t)$. Then, in the binary collision approximation,[1,4] and assuming the laser field is turned on adiabatically, starting at $t=0$, we obtain the following equation for the reduced density matrix $\hat{\sigma}_D$ of the atomic system:

$$\begin{aligned}
\partial_t\hat{\sigma}_D(t) &= [\tilde{L}^A_D(t) + \tilde{\dot{\theta}}(t)]\hat{\sigma}_D(t)\\
&\quad + \int_0^\infty d\tau\,\mathscr{C}(t,t-\tau)\hat{\sigma}_D(t-\tau)
\end{aligned} \tag{26}$$

(as usual $\hat{\sigma}_D(t) = {\rm Tr}_{\rm pert}[\hat{\rho}_D(t)]$) with

$$\mathscr{C}(t,t-\tau) = N\,{\rm Tr}_{1\,\rm pert}\left[\hat{\rho}^D_{\rm pert}(1)\exp\left[\int_{t-\tau}^{t}\tilde{L}^A_D(t')dt'\right][\tilde{V}^{1\rm I}_D(\tau;t)\tilde{U}^1_D(\tau,0;t)\tilde{V}^{1\rm I}_D(\tau;t) + \tilde{V}^{1\rm I}_D(\tau;t)\delta(\tau)]\right]\ , \tag{27}$$

where

$$\tilde{V}_D^{1\mathrm{I}}(\tau;t)=\exp\left[-\int_{t-\tau}^{t}[\tilde{L}_D^A(\tau')+\tilde{L}_p^1]d\tau'\right]\tilde{V}_D^1(t)\exp\left[+\int_{t-\tau}^{t}[\tilde{L}_D^A(\tau')+\tilde{L}_p^1]d\tau'\right] \tag{28}$$

and

$$\tilde{U}_D^1(\tau,0;t)=T\exp\left[\int_0^\tau \tilde{V}_D^1(t'';t)dt''\right] \tag{29}$$

(where T is the time-ordering operator). The superscripts 1 refer to single perturber variables. Note that $\tilde{V}_D^{1\mathrm{I}}(\tau;t)$ is still an implicit function of time through the dependence of $\tilde{V}_D^1(t)$ on $\Omega(t)$. In deriving (26) and (27) we have also assumed $\dot{\theta}\tau_c\sim\alpha\tau_c\ll 1$ so that we are working in adiabatic eigenstates. This assumption, in physical terms, means that the field amplitude, although still a function of time, must be varying slowly enough such that the adiabatic eigenstates do not change appreciably during a collision.

We now consider the adiabatic approximation leading to Eq. (26). For off-diagonal couplings (noting that $\dot{\theta}$ has no diagonal couplings and no couplings to levels not connected by the field) this approximation can be expressed as

$$\tilde{L}_D^A(t)\gg\tilde{\dot{\theta}}(t)\ . \tag{30}$$

In this form the assumption is seen to be similar to the secular approximation of paper I which required $\tilde{L}_D^A\gg\tilde{\gamma}_D^c(t)$. That is, we require that the nondegenerate splittings $\Omega'(t)$ of the laser-coupled levels in the dressed frame be large compared to their ac Stark broadened widths [due to the time-changing field amplitude, $\alpha(t)=\dot{\Omega}(t)/\Omega(t)$]. Thus, as CS (Ref. 7) note, the "turn-on" of the laser pulse should be slow enough such that the Fourier components introduced do not overlap the absorption line. In the context of the secular approximation, the dressed levels connected by the field should be "well separated," $\Omega'(t)\gg\dot{\theta}(t)\sim\alpha(t)$. (Thus the initial turn-on will be adiabatic provided $\Delta\gg\alpha$.)

Applying this adiabatic-secular approximation (usual secular[10] and adiabatic approximation) we find that only the elements of the density matrix that have degenerate frequencies are coupled. [Actually, in weak fields during turn-on, $\tilde{L}_D^A(t)\sim\delta_D(t)\gg\tilde{\gamma}_D^c(t)$ is not necessarily true for Zeeman degenerate components. However, when the field is weak, the spherical symmetry of the collisional interaction again ensures that for determining populations only diagonal elements of the density matrix are coupled.[11]] The off-diagonal elements uncouple from the diagonal elements, and (as in paper I) one has only to solve Markoffian equations for the dressed-frame density operator. It is important to recall, however, that the damping operators are still functions of the instantaneous field strength (and hence t). Thus we can write Eqs. (26) as

$$\partial_t\hat{\sigma}_D(t)=\tilde{L}_D^A(t)\hat{\sigma}_D(t)+\tilde{\gamma}_D^c(t)\hat{\sigma}_D(t) \tag{31}$$

with the time-dependent collisional operator given by

$$\tilde{\gamma}_D^c(t)\equiv N\,\mathrm{Tr}_{1\,\mathrm{pert}}\left[\int_0^\infty d\tau[\tilde{V}_D^{1\mathrm{I}}(\tau;t)\tilde{U}_D^1(\tau,0;t)+\tilde{V}_D^{1\mathrm{I}}(0;t)+\tilde{V}_D^{1\mathrm{I}}(\tau;t)\delta(\tau)]\hat{\rho}_{\mathrm{pert}}^D(1)\right] \tag{32}$$

valid for observation times long compared to τ_c. This is the central result of this section; it is analogous to paper I, Eq. (32), with the adiabatic replacement $\tilde{L}_D^A\rightarrow\tilde{L}_D^A(t)$ and where $\tilde{\gamma}_D^c\rightarrow\tilde{\gamma}_D^c(t)$ is now a function of time [through $\Omega(t)$]. In the impact limit [$\Delta,\Omega(t)\ll\tau_c^{-1}$] and two-state case it reduces exactly to the results of CS.[7] Outside the impact limit the collision operator can be evaluated from an S-matrix approach in the dressed frame and will in general be a complicated function of Δ and $\Omega(t)$.[1,12]

In the classical path approximation the matrix elements of $\tilde{\gamma}_D^c(t)$ can be expressed in terms of "time-dependent" S^D matrix elements in the dressed frame,[1] thus

$$\langle\langle ij\,|\,\tilde{\gamma}_D^c(t)\,|\,lm\,\rangle\rangle=[S_{il}^D(t)S_{mj}^{+D}(t)-\delta_{il}\delta_{mj}]_{\mathrm{av}}\ , \tag{33}$$

where the average is over impact parameters, velocities, and directions relative to the laser field. Here

$$S^D(t)=T\exp\left[-i\int_{-\infty}^{\infty}\tilde{V}_1^1(\tau,t)d\tau\right]\ .$$

Note that while $S^D(t)$ is a function of time, for $\alpha\tau_c\ll 1$, t acts only as a parameter defining the field strength which is fixed for a given collision.

III. CORRELATION FUNCTION

The "physical" spectrum as defined by Eberly and Wodkiewicz[8] can be written as the two-time Fourier transform of the atomic dipole correlation function:

$$I_{\epsilon'}(\omega,t) \propto \Gamma_0 \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt'' e^{-(\Gamma_0 - i\omega)(t-t')} e^{-(\Gamma_0 + i\omega)(t-t'')} \langle\langle D_{\epsilon'}^{+}(t') D_{\epsilon'}^{-}(t'') \rangle\rangle \ . \tag{34}$$

Here $I_{\epsilon'}(\omega,t)$ is the time-dependent intensity (in units of photons $\sec^{-1}$ atom^{-1}) at frequency ω, and polarization ϵ', as measured with a spectrometer of bandwidth Γ_0. As usual, in terms of raising and lowering operators

$$D_{\epsilon'}^{+}(t) \equiv \vec{D}^{+}(t) \cdot \vec{\epsilon}' \tag{35}$$

and

$$D_{\epsilon'}^{-}(t) \equiv \vec{D}^{-}(t) \cdot \epsilon'^{*} \ .$$

$\vec{D}(t)$ is the atomic dipole operator in the Heisenberg picture, and the double angular brackets in Eq. (34) refer to an ensemble average over perturber variables and a trace over the relevant atomic states. Specifically the dipole correlation function is

$$\begin{aligned} \langle\langle D_{\epsilon'}^{+}(t') D_{\epsilon'}^{-}(t'') \rangle\rangle &= \mathrm{Tr}[\hat{\rho}_H D_{\epsilon'}^{+}(t') D_{\epsilon'}^{-}(t'')] \\ &\equiv C_{\epsilon'}(t',t'') \ , \end{aligned} \tag{36}$$

where $\hat{\rho}_H$ is the time-dependent density matrix of the total system in the Heisenberg picture. Now we want to be able to write a Markoffian equation of motion for the autocorrelation function of the atomic dipole. We know from paper I that this will be possible for arbitrary laser intensities (and scattered frequencies within the impact regime in the dressed frame) *only* if we write the autocorrelation function in terms of dressed basis operators. So we start with the expression for the dipole autocorrelation function in the atomic basis in the form

$$C_{\epsilon'}(t_2,t_1) = \mathrm{Tr}[G_A(t_2,t_1) D_{\epsilon'}^{-}] \ , \tag{37}$$

where the trace is taken over the atom plus perturbers, the correlation matrix $G_A(t,t_1)$ satisfies $\partial_t G_A(t,t_1) = \tilde{L}(t) G_A(t,t_1)$, and $\tilde{L}(t)$ is the time-dependent Liouville operator for the complete system of atom plus perturbers. The alternative form of $C_{\epsilon'}(t_2,t_1)$ in the dressed frame is written thus

$$C_{\epsilon'}(t_2,t_1) = \mathrm{Tr}[G_D(t_2,t_1) D_{D\epsilon'}^{-}(t_2)] \ , \tag{38}$$

where the trace is taken over the dressed states at t_2 plus the perturbers and

$$D_{D\epsilon'}^{\pm}(t) = \hat{U}_D(t) \hat{U}_R(t) D_{\epsilon'}^{\pm} \hat{U}_R^{\dagger}(t) \hat{U}_D^{\dagger}(t) \tag{39}$$

is the adiabatic dressed-frame Schrödinger picture operator.

The derivation proceeds exactly as in paper I. We make the binary collision approximation, and for emission detunings in the dressed frame $\Delta\omega_D \ll \tau_c^{-1}$, we can again neglect the "destruction term" which represents emission during a collision. Following the techniques outlined for the reduced density matrix we make the adiabatic-secular approximations and, assuming $\alpha \ll \tau_c^{-1}$, the quantum fluctuation regression theorem will be valid in the dressed frame, i.e., the time evolution of the correlation function will be determined by a Markoffian equation exactly analogous to Eqs. (31) and (32). This equation for the correlation matrix in the dressed basis is written thus

$$\partial_t \hat{G}_D(t,t') = \tilde{L}_D^{A}(t) \hat{G}_D(t,t') + \tilde{\gamma}_D^{c}(t) \hat{G}_D(t,t') \ , \tag{40}$$

and is solved using the initial conditions

$$\hat{G}_D(t',t') = \hat{\sigma}_D(t') \hat{D}_{D\epsilon'}^{+}(t') \ . \tag{41}$$

Solution of this set of equations gives us the time-dependent spectrum when inserted in Eq. (34).

We now outline the solution of these equations for the form of the integrated scattered spectral components in a specific case, namely a linearly polarized laser incident on a $J_g = 0$ to $J_e = 1$ transition. This problem has been treated in detail by Cooper *et al.*,[3] including the effect of collisions, in the steady-state and impact regime. In that case they predict a triplet in the scattered spectrum polarized parallel to the incident laser and a doublet in the scattered spectrum polarized perpendicular to the incident laser. In the parallel (π) polarization the three peaks correspond to Rayleigh scattering from the ground or $m_J = 0$ substate of the excited level, three-photon scattering followed by fluorescence, and collision-induced fluorescence. These features are exactly those associated with nonlinear scattering from an idealized two-state atom in the presence of collisions.[2,3,5,7] In the perpendicular polarization the two peaks correspond to collisional excitation of the $m_J = \pm 1$ sublevels of the excited state followed by fluorescence to the ground state, or an inverse Raman excitation of the $m_J = 0$ excited substate. In the dressed-atom picture these processes can be

represented as collisional transfer and radiative decays between the dressed levels of the atom (Fig. 1). This polarized scattering spectrum has been observed by Kleiber *et al.*[6] We expect qualitatively similar results in this time-dependent, nonimpact theory.

The dressed states for a $J_g=0$ to $J_e=1$ transition with a linearly polarized incident laser are given by Eqs. (15) and (16):

$$|\mathrm{II}\rangle = b_2(t)\,|00\rangle - b_1(t)\,|10\rangle\ , \tag{42}$$

$$|\mathrm{I}\rangle = b_1(t)\,|00\rangle + b_2(t)\,|10\rangle\ , \tag{43}$$

$$|\pm 1\rangle = |1\pm 1\rangle\ . \tag{44}$$

Here, the rotating-frame atomic states are labeled $|Jm_j\rangle$ by their angular momentum quantum numbers. It is obvious that the dressed states are still eigenfunctions of the component of angular momentum along the driving field's polarization direction. In this case the average collisional operators have cylindrical symmetry which means they can only couple elements of the density matrix with the same value of (m_j-m_i), where the density-matrix elements are labeled $\sigma^D_{m_j m_i}$.[11] This leads to a considerable simplification in Eq. (26).

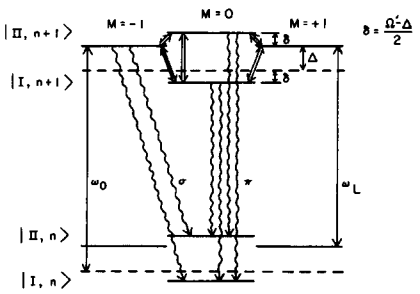

FIG. 1. Collisional, $\Longleftrightarrow$, and radiative, $\leftsquigarrow$, transitions in the dressed frame.

Using Eq. (38) the correlation functions for the two polarizations can be written

$$C_{\parallel}(t,t') = b_1(t)b_2(t)[G^D_{\mathrm{II\,II}}(t,t') - G^D_{\mathrm{II}}(t,t')] + b_1^2(t)G^D_{\mathrm{III}}(t,t') - b_2^2(t)G^D_{\mathrm{I\,II}}(t,t') \tag{45}$$

and

$$C_{\perp}(t,t') = \frac{1}{\sqrt{2}}\{b_2(t)[G^D_{1\,\mathrm{II}}(t,t') + G^D_{-1\,\mathrm{II}}(t,t') + b_1(t)[G^D_{1\mathrm{I}}(t,t') + G^D_{-1\mathrm{I}}(t,t')]\}\ , \tag{46}$$

where $\hat{G}^D(t,t')$ is the solution to (40) with initial conditions (41). Each of the terms in (45) and (46) correspond to different peaks in the scattered spectrum.

Using the cylindrical symmetry and the adiabatic-secular approximation we can write the equations for the relevant elements of the correlation matrix as

$$[\dot{G}^D_{11}(t,t') + \dot{G}^D_{-1-1}(t,t')] = -\Gamma_{11}(t)[G^D_{11}(t,t') + G^D_{-1-1}(t,t')] + \Gamma_{1\,\mathrm{II}}(t)G^D_{\mathrm{II\,II}}(t,t') + \Gamma_{1\mathrm{I}}(t)G^D_{\mathrm{II}}(t,t')\ , \tag{47}$$

$$\dot{G}^D_{\mathrm{II\,II}}(t,t') = -\Gamma_{\mathrm{II\,II}}(t)G^D_{\mathrm{II\,II}}(t,t') + \Gamma_{\mathrm{II}\,1}(t)[G^D_{11}(t,t') + G^D_{-1-1}(t,t')] + \Gamma_{\mathrm{II\,I}}(t)G^D_{\mathrm{II}}(t,t')\ , \tag{48}$$

$$\dot{G}^D_{\mathrm{II}}(t,t') = -\Gamma_{\mathrm{II}}(t)G^D_{\mathrm{II}}(t,t') + \Gamma_{\mathrm{I}\,1}(t)[G^D_{11}(t,t') + G^D_{-1-1}(t,t')] + \Gamma_{\mathrm{I\,II}}(t)G^D_{\mathrm{II\,II}}(t,t')\ , \tag{49}$$

$$\dot{G}^D_{\mathrm{I\,II}}(t,t') = -i\Omega'(t)G^D_{\mathrm{I\,II}}(t,t')\ , \tag{50}$$

$$[\dot{G}^D_{1\mathrm{I}}(t,t') + \dot{G}^D_{-1\mathrm{I}}(t,t')] = i\left[\frac{\Omega'(t)+\Delta}{2}\right][G^D_{1\mathrm{I}}(t,t') + G^D_{-1\mathrm{I}}(t,t')]\ , \tag{51}$$

$$[\dot{G}^D_{1\,\mathrm{II}}(t,t') + \dot{G}^D_{-1\,\mathrm{II}}(t,t')] = -i\left[\frac{\Omega'(t)-\Delta}{2}\right][G^D_{1\,\mathrm{II}}(t,t') + G^D_{-1\,\mathrm{II}}(t,t')] \tag{52}$$

with

$$\Gamma_{11}(t) = \tfrac{1}{2}[\langle\langle 11\,|\,\tilde{\gamma}^c_D(t)\,|\,11\rangle\rangle + \langle\langle -1-1\,|\,\tilde{\gamma}^c_D(t)\,|\,-1-1\rangle\rangle] + \gamma_N\ , \tag{53}$$

$$\Gamma_{\mathrm{II\,II}}(t) = \langle\langle \mathrm{II\,II}\,|\,\tilde{\gamma}^c_D(t)\,|\,\mathrm{II\,II}\rangle\rangle + \gamma_N^{\mathrm{II}}(t)\ , \tag{54}$$

$$\Gamma_{\mathrm{III}}(t)=\langle\langle \mathrm{III}\,|\,\tilde{\gamma}_D^c(t)\,|\,\mathrm{II}\rangle\rangle+\gamma_N^{\mathrm{III}}(t)\,, \tag{55}$$

$$\Gamma_{\mathrm{I\,III}}(t)=\langle\langle \mathrm{II}\,|\,\tilde{\gamma}_D^c(t)\,|\,\mathrm{III}\rangle\rangle+\gamma_N^{\mathrm{I\,III}}(t)\,, \tag{56}$$

$$\Gamma_{\mathrm{II}}(t)=\langle\langle \mathrm{II}\,|\,\tilde{\gamma}_D^c(t)\,|\,\mathrm{II}\rangle\rangle+\gamma_N^{\mathrm{III}}(t)\,, \tag{57}$$

$$\Gamma_{1\,\mathrm{III}}(t)=\langle\langle 11\,|\,\tilde{\gamma}_D^c(t)\,|\,\mathrm{III}\rangle\rangle+\langle\langle -1-1\,|\,\tilde{\gamma}_D^c(t)\,|\,\mathrm{III}\rangle\rangle\,, \tag{58}$$

$$\Gamma_{11}(t)=\langle\langle 11\,|\,\tilde{\gamma}_D^c(t)\,|\,\mathrm{II}\rangle\rangle+\langle\langle -1-1\,|\,\tilde{\gamma}_D^c(t)\,|\,\mathrm{II}\rangle\rangle\,, \tag{59}$$

$$\Gamma_{\mathrm{III}1}(t)=\tfrac{1}{2}[\langle\langle \mathrm{III}\,|\,\tilde{\gamma}_D^c(t)\,|\,11\rangle\rangle+\langle\langle \mathrm{III}\,|\,\tilde{\gamma}_D^c(t)\,|\,-1-1\rangle\rangle]+\gamma_N^{\mathrm{III}1}(t)\,, \tag{60}$$

$$\Gamma_{\mathrm{II}1}(t)=\tfrac{1}{2}[\langle\langle \mathrm{II}\,|\,\tilde{\gamma}_D^c(t)\,|\,11\rangle\rangle+\langle\mathrm{II}\,|\,\tilde{\gamma}_D^c(t)\,|\,-1-1\rangle\rangle]+\gamma_N^{\mathrm{II}1}(t)\,, \tag{61}$$

$$\gamma_N^{\mathrm{III}}(t)=b_2^4(t)\gamma_N\,, \tag{62}$$

$$\gamma_N^{\mathrm{I\,III}}(t)=b_1^4(t)\gamma_N\,, \tag{63}$$

$$\gamma_N^{\mathrm{II}}(t)=b_1^2(t)\gamma_N\,, \tag{64}$$

$$\gamma_N^{\mathrm{II}\,1}(t)=b_2^2(t)\gamma_N\,, \tag{65}$$

where γ_N is the spontaneous decay rate for the upper level.

Solving (50)–(52) with the initial conditions (41) [with (39)] we find (omitting the constant dipole moment factor d_{eg}^{+})

$$G_{\mathrm{III}}^D(t,t')=\exp\left[+i\int_{t'}^{t}\Omega'(\tau)d\tau\right][b_1^2(t')\sigma_{\mathrm{III}}(t')]\,, \tag{66}$$

$$G_{\mathrm{III}}^D(t,t')=\exp\left[-i\int_{t'}^{t}\Omega'(\tau)d\tau\right][-b_2^2(t')\sigma_{\mathrm{II}}^D(t')] \tag{67}$$

and

$$[G_{1\,\mathrm{III}}^D(t,t')+G_{-1\,\mathrm{II}}^D(t,t')]=\exp\left[-i\int_{t'}^{t}\left[\frac{\Omega'(\tau)-\Delta}{2}\right]d\tau\right]\left[b_2(t')\frac{\sigma_{11}^D(t')+\sigma_{-1-1}^D(t')}{\sqrt{2}}\right], \tag{68}$$

$$[G_{11}^D(t,t')+G_{-1\,1}^D(t,t')]=\exp\left[+i\int_{t'}^{t}\left[\frac{\Omega'(\tau)+\Delta}{2}\right]d\tau\right]\left[b_1(t')\frac{\sigma_{11}^D(t')+\sigma_{-1-1}^D(t')}{\sqrt{2}}\right]. \tag{69}$$

Since the spectrum is related to the Fourier transform of (66)–(69) we see that each term contributes to a different spectral peak. Thus G_{III}^D contributes to a peak near $\omega\sim-\Omega'$ and can be identified with the π fluorescence while G_{III}^D contributes near $\omega\sim+\Omega'$ and can be identified with the three-photon peak. Similarly (66) and (67) can be identified with the inverse Raman and σ fluorescence, respectively. [Note that these features are evaluated in a "rotating" frame; in the laboratory atomic frame these (π) features will be located at $\omega_L\pm\Omega'(t)$ and similarly for the σ spectrum at $\omega_L+\delta_D(t)$ and $\omega_0-\delta_D(t)$.] In deriving (66)–(69) we have also used the fact that $\sigma_{\mathrm{III}}^D(0)=0$ for an adiabatic pulse which turns on at $t=0$ (since if $\Omega'>\dot{\theta}$ then there will be no coupling to this density matrix element).

The quantity $G_{\mathrm{III}}^D(t,t')-G_{\mathrm{II}}^D(t,t')$ is much more difficult to evaluate due to interference effects caused by near degeneracy; this term leads to the central (Rayleigh) peak in the three-peak parallel spectrum. The contributions to this feature come from two terms, Rayleigh scattering off the ground state and Rayleigh scattering off the $m_J=0$ excited state. Alternatively in the quantized field dressed-level picture, both decays $|\,\mathrm{II},n+1\rangle\rightarrow|\,\mathrm{II},n\rangle$ and $|\,\mathrm{I},n+1\rangle\rightarrow|\,\mathrm{I},n\rangle$ contribute to the central peak. Interference between these processes makes the intensity *spectrum* of the central peak quite difficult to calculate directly. However, if we are only interested in *integrated intensities* we can substitute the term

$$G_{||}(t,t')=b_1(t)b_2(t)[G_{\mathrm{III}}^D(t,t')-G_{\mathrm{II}}^D(t,t')] \tag{70}$$

into (34) [using (35) and (36)] and integrate ω over a frequency range $\Delta\omega$ ($\Omega'\gg\Delta\omega\gg\tilde{\gamma}_D^c$) about $\omega=\omega_L$. The condition $\Delta\omega\gg\tilde{\gamma}_D^c$ is upheld when the components are well separated (as we have assumed). The integral

$$\int_{-\Delta\omega}^{\Delta\omega} d(\omega-\omega_L)e^{i(\omega-\omega_L)(t'-t'')} \tag{71}$$

then gives a δ function on $(t'-t'')$. If we assume, in addition, that the spectrometer bandwidth Γ_0 is large such that

$$\Omega' \gg \Gamma_0 \gg \tilde{\gamma}_D^c, \alpha \tag{72}$$

(as is the case in many experiments[5,6,9]), then the remaining time integral in (34) can be evaluated by noting that the factor $\exp[-2\Gamma_0(t-t')]$ again acts as a δ function, picking out the value of the integrand at $t'=t$. Thus the integrated intensity in the central (Rayleigh) peak is given by

$$I_R \propto b_1(t)b_2(t)[G^D_{\mathrm{II}\,\mathrm{II}}(t,t)-G_{\mathrm{II}}(t,t)] . \tag{73}$$

Using the initial conditions (41), and again assuming $\sigma^D_{\mathrm{III}}(0)=0$ for an adiabatic pulse which turns on at $t=0$, we find

$$I_R(t) \propto b_1^2(t)b_2^2(t)[\sigma^D_{\mathrm{II}\,\mathrm{II}}(t)+\sigma^D_{\mathrm{II}}(t)] . \tag{74}$$

The integrated intensities of the other peaks in the spectrum can be obtained the same way: integrating over a finite bandwidth, $\Delta\omega$ ($\Omega' \gg \Delta\omega \gg \tilde{\gamma}_D^c$), about each component, and using (72). We then find for the parallel fluorescence and three-photon component (in the parallel polarization),

$$I_{||}(t) \propto b_1^4(t)\sigma^D_{\mathrm{II}\,\mathrm{II}}(t) \tag{75}$$

and

$$I_3(t) \propto b_2^4(t)\sigma^D_{\mathrm{II}}(t) . \tag{76}$$

The total intensity in the π spectrum is

$$I^{\mathrm{tot}}_{||}(t) \propto [b_1^2(t)\sigma_{\mathrm{II}\,\mathrm{II}}(t)+b_2^2(t)\sigma_{\mathrm{II}}(t)] \tag{77}$$

or

$$I^{\mathrm{tot}}_{||}(t) \propto \sigma_{00}(t) , \tag{78}$$

where $\sigma_{00}(t)$ is the population of the $m_j=0$ substate of the excited level. In the perpendicular polarization we find the following expressions for the perpendicular fluorescence and the inverse Raman intensities:

$$I_{\perp}(t) \propto b_1^2(t)\left[\frac{\sigma^D_{11}(t)+\sigma^D_{-1-1}(t)}{2}\right] , \tag{79}$$

$$I_{\mathrm{IR}}(t) \propto b_2^2(t)\left[\frac{\sigma^D_{11}(t)+\sigma^D_{-1-1}(t)}{2}\right] , \tag{80}$$

and the total σ scattered intensity is given by

$$I^{\mathrm{tot}}_{\perp} \propto [\sigma_{11}(t)+\sigma_{-1-1}(t)] . \tag{81}$$

We note that these results are a straightforward adiabatic extension of the results of Cooper *et al.*,[3] and that even in the time-dependent case, the integrated intensities of the spectral lines are proportional to the instantaneous dressed-state populations. This result was pointed out in the two-state case by CS (Ref. 7) but we feel it has not been fully appreciated.

We now solve the master equation (40) for the reduced dressed-atom density matrix elements. In the same approximations the derivation follows exactly that for the correlation matrix and we find (suppressing the argument 't' in all variables)

$$\dot{\sigma}^D_{\mathrm{III}} = -i\Omega'\sigma^D_{\mathrm{III}} , \tag{82}$$

$$\dot{\sigma}^D_{\mathrm{II}\,\mathrm{II}} = -\Gamma_{\mathrm{II}\,\mathrm{II}}\sigma^D_{\mathrm{II}\,\mathrm{II}}+\Gamma_{\mathrm{II}\,\mathrm{I}}(\sigma^D_{11}+\sigma^D_{-1-1}) +\Gamma_{\mathrm{II}\,\mathrm{III}}\sigma^D_{\mathrm{II}} , \tag{83}$$

$$\dot{\sigma}_{\mathrm{II}} = -\Gamma_{\mathrm{II}}\sigma^D_{\mathrm{II}}+\Gamma_{\mathrm{I}\,\mathrm{I}}(\sigma^D_{11}+\sigma^D_{-1-1})+\Gamma_{\mathrm{II}\,\mathrm{III}}\sigma^D_{\mathrm{II}\,\mathrm{II}} , \tag{84}$$

$$(\dot{\sigma}^D_{11}+\dot{\sigma}^D_{-1-1}) = -\Gamma_{11}(\sigma^D_{11}+\sigma^D_{-1-1}) +\Gamma_{1\,\mathrm{II}}\sigma^D_{\mathrm{II}\,\mathrm{II}}+\Gamma_{1\,\mathrm{II}}\sigma^D_{\mathrm{II}} . \tag{85}$$

The solution to (82) is

$$\sigma^D_{\mathrm{III}}(t)=\sigma^D_{\mathrm{III}}(0)\exp\left[-i\int_0^t \Omega'(\tau)d\tau\right] \simeq 0 \tag{86}$$

for $\sigma^D_{\mathrm{III}}(0)=0$ at the beginning of the laser pulse. This is in agreement with the two-state adiabatic results of CS.[7]

The solution to (83)–(85) are complicated in general. Here we cannot neglect the damping rates (both collisional and radiative) as we did in the solution of the diagonal elements of the correlation matrix (which were only required for short times); this is because the damping is obviously quite important in determining the dressed-level *populations* and hence the integrated intensities of the spectral lines.

The damping rates have already been given in Eqs. (53)–(65). They will be in general complicated functions of $\Omega(t)$ and hence complicated functions of time. The damping rates must be evaluated for a given laser pulse shape, and thus the full calculation will be quite tedious. However, for some models, e.g., an adiabatic square pulse, the solution will be straightforward.

The dressed-atom collision rates as a function of Ω were originally calculated for this problem by Light and Szöke.[12] We have repeated those calculations, closely following the approach of Light and

Szöke[12] to define the problem, but using a significantly different numerical algorithm for solving the coupled equations. We also use more stringent error control and more sophisticated numerical techniques for the required averages over impact parameter and collision orientation. Our results are found to have the same qualitative behavior as those of Light and Szöke[12] although certain nonphysical "glitches" in their results (see below) have been smoothed out by our approach.

The calculated cross sections for collisional transfer between dressed levels at $\Delta=17$ cm^{-1} (red) detuning and using $C_6=(4.1\times10^{-31})$ (Ref. 13) are given in Fig. 2 as a function of field strength. There $\tilde{S}_{ij}$ is the cross section for collisional population transfer from dressed state $|j\rangle$ to dressed state $|i\rangle$; it is given by $\tilde{S}_{ij}=\{S^D_{ij}S^{D+}_{ji}\}_{\rm av}$ where the average is over impact parameter and collision orientation and S^D is the usual dressed-frame S matrix.[1] Thus $\langle\langle aa|\tilde{\gamma}^c_D(t)|bb\rangle\rangle=Nv_p\tilde{S}_{ab}$ where N is the perturber density and v_p the relative velocity. (The notation $\tilde{S}$ rather than σ is used for the cross section in order to avoid confusion with density matrix elements.)

These cross sections agree qualitatively with those of Light and Szöke,[12] however, the following points should be noted. (1) Our cross-section results are uniformly a factor of 2 lower than those of Light and Szöke.[12] This is due to a simple difference of a factor of 2 between our definition of the cross section and theirs. (2) The results of Light and Szöke show an anomalous drop in the cross-section ratio $(\tilde{S}_{1\rm I}+\tilde{S}_{-1\rm I})/\tilde{S}_{\rm II}$ at around $\Omega/\Delta=0.5$; there is no such "glitch" in our results. (3) The results of Light and Szöke[12] show an oscillatory structure in the total cross section $\tilde{S}_{\rm tot}=(\tilde{S}_{\rm II}+\tilde{S}_{1\rm I}+\tilde{S}_{-1\rm I})$ which is smoothed out by our approach. (4) Light and Szöke do not evaluate cross sections for transitions $(\tilde{S}_{1\rm II}+\tilde{S}_{-1\rm II})$ [Fig. 2(c)] and these cross sections will be necessary for the detailed solution of the rate equations (83)–(85). (5) In obtaining the cross sections, contrary to Fig. 6 of LS, the integrand as a function of impact parameter was found to oscillate very wildly at small impact parameters (and this was given special attention in our numerical method). (6) The symmetry $P_{ij}(\pi-\alpha,\beta)=P_{ij}(\alpha,\beta)$ used by LS to reduce the angular integrations is not strictly valid, and was not used here.

Again Eqs. (83)–(85) are all functions of time [through $\Omega(t)$] and we choose to numerically solve these rate equations for a given model laser pulse shape. We choose the simple but useful model of an adiabatic square pulse; this model has been discussed by Courtens and Szöke.[7] The pulse shape is essentially "square," but with brief rise and fall times (τ_R) which ensure the adiabatic condition is satisfied. We require for such a model $\Omega'^{-1}\ll\tau_R\sim\alpha^{-1}\ll T_P,\gamma^{-1}$ where T_P is the pulse length. The condition $\tau_R\gg\Omega'^{-1}$ (i.e., $\alpha\ll\Omega'$) en-

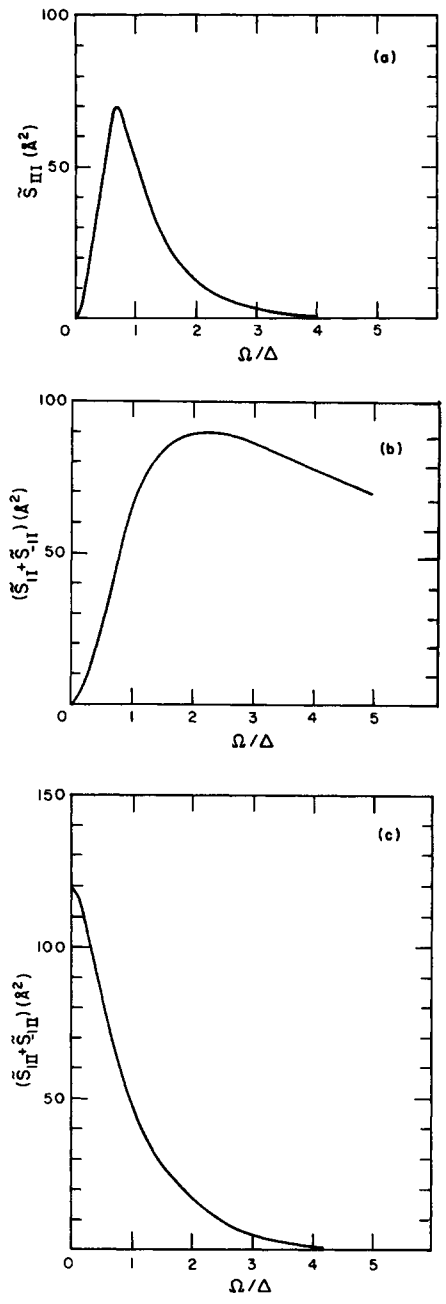

FIG. 2. (a)–(c) Cross sections $\tilde{S}_{ij}$ for population transfers by collisions in dressed frame (see text).

sures that the pulse is adiabatic. The condition $\tau_R \ll \gamma^{-1}$ (where $\gamma \sim \gamma_c, \gamma_N$) ensures that very little population transfer takes place between dressed levels when the laser pulse is turning on (or off) and the intensity is not well defined. The condition $\tau_R \ll T_P$ ensures that the pulse appears "square" and has a well-characterized intensity.

IV. RESULTS

Using the adiabatic square-pulse model and the cross-section results of Fig. 2 we can numerically solve (83)–(85), using

$$\sigma_{\text{II II}}+\sigma_{\text{II}}+(\sigma_{11}+\sigma_{-1-1})=1$$

to obtain results for the integrated intensities of the scattered spectral features (74)–(81). Often, in experiments, the spectral features are time integrated over some interval which is long compared to the system relaxation rates. In the experiments of Kleiber *et al.*[9] a gated, analog integrate and hold circuit was used to collect the total signal from the photomultiplier. In fact, in the adiabatic square-pulse model they experimentally measured the quantity

$$\int_0^{T_P} I_{\epsilon'}(t')dt' .$$

Hence the integrated intensities for a given ac Stark shift (neglecting redistribution in the wings outside the impact region) are

$$I_{||}=\theta b_1^4 \gamma_N \int_0^{T_P} \sigma^D_{\text{II II}}(t')dt' , \tag{87}$$

$$I_3=\theta b_2^4 \gamma_N \int_0^{T_P} \sigma^D_{\text{II}}(t')dt' , \tag{88}$$

$$I_{\text{R}}=\theta 2 b_1^2 b_2^2 \gamma_N \int_0^{T_P} [\sigma^D_{\text{II}}(t')+\sigma^D_{\text{II II}}(t')]dt' \tag{89}$$

in the π spectrum, and

$$I_{\perp}=\theta b_1^2 \gamma_N \int_0^{T_P} \left[\frac{\sigma^D_{11}(t')+\sigma^D_{-1-1}(t')}{2}\right]dt' , \tag{90}$$

$$I_{\text{IR}}=\theta b_2^2 \gamma_N \int_0^{T_P} \left[\frac{\sigma^D_{11}(t')+\sigma_{-1-1}(t')}{2}\right]dt' \tag{91}$$

in the σ spectrum. Here θ is a factor which converts an integrated fluorescence signal for a given component in photon/pulse, into a voltage signal on the storage capacitor. After the intense laser pulse, fluorescence will occur at the unperturbed atomic resonance frequency, but this light is not considered to be part of the observed intensities. The b's are appropriate to the constant pulse intensity. The quantity θ is difficult to measure absolutely, however, Kleiber *et al.*[9] were concerned experimentally with ratios of the above observables, so that the factor θ does not explicitly appear. In particular they were interested in the adiabatic square-pulse model in the depolarization ratio

$$\frac{I_{\perp}}{I_{||}}=\frac{\int_0^{T_P}(\sigma^D_{11}+\sigma^D_{-1-1})dt'}{2b_1^2\int_0^{T_P}\sigma^D_{\text{II II}}dt'} . \tag{92}$$

We can evaluate this ratio to obtain quantitative theoretical predictions which can be directly compared with the experimental results of Kleiber *et al.*[9] The theoretically predicted depolarization, Eq. (92), at $\Delta=17$ cm^{-1} (red) and as a function of (Ω/Δ) are obtained from the solution of the rate equations (83)–(85) using the adiabatic square-pulse model ($T_P=5$ nsec), the cross sections of Fig. 2, and $\gamma_N=2\times10^8$ sec^{-1} (Ref. 14) for the natural decay rate. These results are shown for an argon pressure of 10 Torr in Fig. 3; the comparison with the experimental results has been given elsewhere.[9]

[It should be pointed out that the measured depolarization of Kleiber *et al.*[9] as a function of Stark shift is to be compared with twice the theoretically calculated ratio $I_{\perp}/I_{||}$ of Fig. 3. The origin of the factor of 2 is quite subtle and is explained in detail in Ref. 15. Basically it is due to the fact that the Stark shifts for the perpendicular and parallel fluorescent components differ by a factor of 2 for the same field strength. Physically the same range of intensities is compressed into half as much spectral range in the perpendicular spectrum as in the parallel; this is the origin of the factors of 2 between

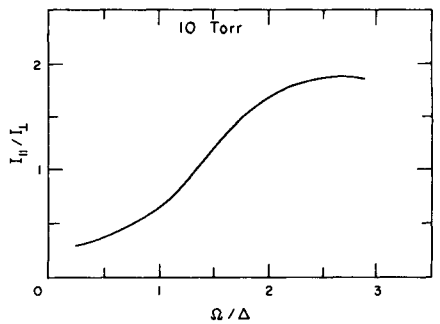

FIG. 3. Depolarization ratio $I_{\perp}/I_{||}$ for conditions given in text.

the measured depolarization as a function of Stark shift given by Kleiber *et al.*[9] and the theoretically evaluated depolarization as a function of field strength given here.]

V. CONCLUSIONS

We have shown how to use the formalism of Burnett *et al.*[1] to generalize the time-dependent theory of Courtens and Szöke[7] to the nonimpact region of the spectrum including arbitrary laser intensities and detunings, with Zeeman degeneracy effects. We have justified the use of Markoffian master equation in the BCA, adiabatic, and secular approximation. We have shown that if, in addition, we are only interested in scattered intensities in the impact region of the spectrum, the equation of motion for the correlation function has the same Markoffian form as that for the density matrix, even in the time-dependent case. We have demonstrated how to apply this theory to a practical calculation where the results can be directly compared with experiment, and we have thus obtained detailed quantitative predictions for physically observable quantities (especially the depolarization) as a function of laser field strength in the adiabatic square-pulse model.

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