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Intensity effects in two-photon collisional redistribution of radiation

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We study laser-intensity effects in two-photon collisional redistribution of radiation. Thereby we concentrate on a special case of recent experimental interest, where the transition from the excited to the final state is saturated by a strong laser field. Both exciting lasers are, however, assumed weak in the sense that the dynamics of a collision between the radiator and a perturber is not significantly influenced by the laser fields. In particular, our treatment explains the intensity independence of the polarization dependence of total fluorescence from the final state which has been measured previously even if the transition from the excited to the final state is saturated.

I. INTRODUCTION

Scattering of light by atoms undergoing collisions with a gas of foreign perturbers is an important tool for studying interatomic interactions.[1–9] In recent years there has been a considerable effort to obtain this type of information from scattering experiments.[10,11] Typically in such an experiment a laser with frequency ω_L excites an atom from its ground state to an excited state and the spectrum of the spontaneously emitted radiation is detected. Of particular interest is the situation where the laser is so far detuned from the atomic transition frequency ω_0, that the photon is absorbed during a time much smaller than the duration of a (strong) collision τ_c, i.e., $1/|\omega_L-\omega_0| \ll \tau_c$. Under this condition details of the intracollisional evolution process are probed and the influence of collisions on the excited atom can no longer be described within the Markov (or impact) approximation.[2] Studying the collisionally redistributed peak of the scattered spectrum, which is centered around the atomic transition frequency ω_0, yields information about interatomic potentials.[11]

Recently, Alford *et al.*[12] generalized the usual one-photon scattering experiments and studied scattering of two-laser fields by an atom undergoing collisions with a gas of foreign perturbers. The first laser thereby excited a barium atom from its ground $^1S_0$ state to an excited $^1P_1$ state. Its detuning from the atomic transition frequency Δ_1 was so large that the laser photon was absorbed during a collision, i.e., $1/|\Delta_1| \ll \tau_c$. The second laser then induced an almost-resonant transition from this excited $^1P_1$ state to some final state of $^1S_0$ symmetry, i.e., $1/|\Delta_2| \gg \tau_c$, and the polarization dependence of the total fluorescence originating from this final state was detected. With this setup they were able to probe the excited-state manifold and hence obtained detailed information about a collision between the barium atom and a noble-gas perturber. At the same time, they avoided to a large extent trapping of the fluorescence radiation. In particular they made the surprising observation that the polarization dependence of the total fluorescence was independent of the intensity of the second laser, even when the transition from the excited $^1P_1$ to the final $^1S_0$ state was saturated.

In a recent paper,[13] we studied theoretically the scattering of two weak laser fields by an atom undergoing collisions with a gas of foreign perturbers. Here we want to generalize these investigations to a case where the laser fields are no longer weak. In general, this is a complicated task, in particular when the laser fields become so intense that they strongly modify the collision process between the atom and a perturber.[14,5,6] However, we shall not consider this situation here. Motivated by the experiment of Alford *et al.*[12] we shall study the case, where the first laser is strongly detuned from resonance in comparison with the inverse duration of a collision, whereas the second laser is almost on resonance. Both laser fields are assumed to be weak in the sense that they do not significantly influence the collision process between the atom and a perturber, i.e., $|\Omega|\tau_c \ll 1$, where Ω is the Rabi frequency (defined below). The first laser field will be assumed weak, in the sense that the ground-state population of the atom is not significantly depleted. However, the second laser field may be so intense that the almost-resonant transition from the excited to the final state is saturated. In particular, we shall consider the final-state population of the excited atom for a $J=0 \to J=1 \to J=0$ type transition for various polarizations of the exciting laser fields. This is the quantity that has been measured by Alford *et al.*[12] in their two-photon experiment. Within the binary-collision approximation we shall derive analytical expressions for the final-state population in the stationary limit valid also for situations where the second laser saturates the transition from the excited to the final state.

The paper is organized as follows. In Sec. II we present the problem under investigation and give a general set of equations that determine the populations of the excited atom in the stationary limit for arbitrary polarizations of both laser fields. Special examples are given in Sec. III, where we focus on the stationary final-state population brought about by linearly and circularly polarized light.

II. PROBLEM AND GENERAL EQUATIONS

We study a neutral atom (radiator) that is surrounded by N structureless perturbers and interacts with a classical electromagnetic field. The density of the perturbers that collide with the radiator is assumed to be small enough that the collisional interaction with the radiator may be treated within the binary-collision approximation (BCA).[2,15] The classical electromagnetic field

$$\mathbf{E}(t)=\sum_{j=1}^{2}\mathbf{e}_j\mathscr{E}_j e^{-i\omega_j t}+\text{c.c.} \tag{1}$$

consists of two monochromatic laser fields of frequencies $\omega_1\omega_2$ and polarizations $\mathbf{e}_1,\mathbf{e}_2$. In particular, we consider a situation where the first laser field $(\omega_1,\mathbf{e}_1)$ excites the radiator from its ground state $|g\rangle$ with total angular momentum $J=0$ to an excited state $|e_i\rangle$ with $J=1$ and the second laser photon $(\omega_2,\mathbf{e}_2)$ induces a transition from this manifold to the final state $|f\rangle$ with $J=0$ (see Fig. 1). This excitation process can be described in the basis of rotating radiator states $\{|g\rangle,\ |\bar{e}_i\rangle=|e_i\rangle e^{-i\omega_1 t},\ |\bar{f}\rangle=|f\rangle e^{-i(\omega_1+\omega_2)t}\}$ by the effective Hamiltonian[13]

$$\begin{aligned}H_{\text{eff}}=&E_g|g\rangle\langle g|+\sum_{i=1}^{3}(E_g-\hbar\Delta_1)|\bar{e}_i\rangle\langle\bar{e}_i|\\&+(E_g-\hbar\Delta_1-\hbar\Delta_2)|\bar{f}\rangle\langle\bar{f}|\\&-\tfrac{1}{2}\hbar\sum_{i=1}^{3}(\Omega^*_{fe_i}|\bar{e}_i\rangle\langle\bar{f}|+\Omega_{fe_i}|\bar{f}\rangle\langle\bar{e}_i|\\&+\Omega^*_{e_ig}|g\rangle\langle\bar{e}_i|+\Omega_{e_ig}|\bar{e}_i\rangle\langle g|)\end{aligned} \tag{2}$$

with the detunings from resonance

$$\Delta_1=(E_g+\hbar\omega_1-E_e)/\hbar\ ,$$

$$\Delta_2=(E_e+\hbar\omega_2-E_f)/\hbar\ ,$$

and the Rabi frequencies

$$\Omega_{e_ig}=(2/\hbar)\langle e_i|\boldsymbol{\mu}\cdot\mathbf{e}_1|g\rangle\mathscr{E}_1\ ,$$

$$\Omega_{fe_i}=(2/\hbar)\langle f|\boldsymbol{\mu}\cdot\mathbf{e}_2|e_i\rangle\mathscr{E}_2\ .$$

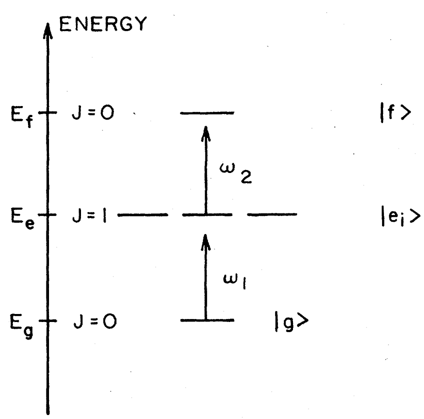

FIG. 1. Schematic representation of the excitation process in the radiator.

Small quadratic Stark shifts due to the other levels in the atom and ionization from the final state have thereby been neglected. Within the BCA the reduced density matrix of the radiator $\sigma(t)=\text{Tr}_{\text{perturbers}}[\rho(t)]$ then obeys the equation of motion[2,13]

$$\frac{d}{dt}\sigma(t)=[L_{\text{eff}}+\Gamma+N\,\text{Tr}_P(V_1\rho_P)]\sigma(t)+\int_0^t dt'\,M(t-t')\sigma(t') \tag{3a}$$

with the collisional memory kernel

$$M(\tau)=N\,\text{Tr}_P[V_1G_1(\tau)V_1\rho_P] \tag{3b}$$

and

$$\frac{d}{d\tau}G_1(\tau)=(L_{\text{eff}}+L_P+V_1+\Gamma)G_1(\tau),\quad \tau\geq 0 \tag{3c}$$

with $G_1(\tau=0)=1$. Thereby we defined

$$L_{\text{eff}}\{\cdots\}=\frac{1}{i\hbar}[H_{\text{eff}},\{\cdots\}]\ ,$$

$$V_1\{\cdots\}=\frac{1}{i\hbar}[V,\{\cdots\}]\ ,$$

$$L_P\{\cdots\}=\frac{1}{i\hbar}[(\hat{p}^2/2M),\{\cdots\}]\ .$$

V is the radiator—single-perturber effective interaction potential, which is assumed to couple only degenerate radiator states, i.e., we neglect inelastic collisions. $\hat{p}$ and M are the momentum and mass of a perturber. ρ_P is the density operator of one perturber, e.g.,

$$\rho_P=\exp(-\beta\hat{p}^2/2M)/\text{Tr}_P[\exp(-\beta\hat{p}^2/2M)]$$

with $\beta=1/kT$. All effects due to the motion of the radiator are neglected in Eqs. (3). Equation (3a) is valid for times much larger than the duration of a collision τ_c as initial radiator-perturber correlations, which decay on a time scale of order τ_c, have been neglected.[2] The tetradic operator Γ describes spontaneous decay of the radiator states in the Markov approximation.[13] Equations (3) determine the population of the radiator atom in a two-photon excitation process for arbitrary relative values of $|\Delta_1|$, $|\Delta_2|$, $|\Omega_{fe_i}|$, $|\Omega_{e_ig}|$, and the inverse duration of a collision $1/\tau_c$ as long as the three-level approximation of Eq. (2) is justified. In particular, they include all the typical saturation effects due to the lasers, which are usually calculated by Bloch equations, and are able to treat cases where the usual Markov treatment of collisions breaks down, because the characteristic time scale of the dynamics of the radiator in the laser fields becomes smaller than the duration of a collision τ_c ($\tau_c\approx 10^{-12}$ s for a van der Waals interaction).[2] It is precisely such a situation that is of great interest in redistribution experiments due to the fact that one becomes able to probe details of the intracollisional evolution process.[9]

Equation (3a) has been discussed in a recent paper in the case of weak fields.[13] Thereby it was assumed that all Rabi frequencies were much smaller than the inverse duration of a collision and also much smaller than all detunings of the lasers from their atomic transition frequencies. The first condition allowed us to treat the influence of the laser fields on the collision between the radiator and a perturber, which is described by $G_1(\tau)$ in Eq. (3c), perturbatively. The second assumption implies that the influence of the laser fields on the radiator is weak so that it can also be treated perturbatively. If these weak-field conditions break down, things become quite complicated. The most difficult situation certainly arises when the Rabi frequencies exceed the inverse duration of a collision so that the laser fields strongly modify the collision process. In particular, degeneracy of radiator states should be properly taken into account in such a case as it may give rise to significant effects.[14,5,6] However, here we shall restrict our study to a less complicated case of recent experimental interest,[12] but in which saturation effects still occur, namely intensities such that

$$|\Omega_{fe_i}|, |\Omega_{e_ig}| \ll 1/\tau_c \, , \tag{4a}$$

$$|\Omega_{fe_i}|, |\Omega_{e_ig}| \ll |\Delta_1| \, , \tag{4b}$$

$$|\Omega_{e_ig}|^2 \ll |\Delta_1| \max\{|\Delta_2|, \gamma\} \, , \tag{4c}$$

and detunings

$$|\Delta_1| \gg 1/\tau_c, \gamma \, , \tag{4d}$$

$$|\Delta_2| \ll 1/\tau_c \, . \tag{4e}$$

γ is thereby a typical spontaneous decay or collisional rate. Equation (4a) implies that the influence of both laser fields on the collision process between the radiator and a perturber is weak so that Eq. (3c) can still be evaluated perturbatively with respect to the laser fields.

According to Eq. (4d) the characteristic time for absorbing the laser photon $(\omega_1, \mathbf{e}_1)$, i.e., $1/|\Delta_1|$, is much less than the duration of a collision τ_c so that this photon is absorbed instantaneously during a collision. But the second laser photon $(\omega_2, \mathbf{e}_2)$ is absorbed on a long time scale in comparison with τ_c [see Eq. (4e)] due to the fact that the detuning Δ_2 is assumed to be small. According to Eqs. (4b) and (4c) the first laser field is effectively weak, but as far as the second laser field is concerned we allow also for situations where the transitions $|e_i\rangle \rightarrow |f\rangle$ is saturated, i.e., $|\Delta_1| \gg |\Omega_{fe_i}| \gtrsim \max\{|\Delta_2|, \gamma\}$. Using the relations of Ref. 13 we find from Eq. (3a) for the stationary populations of the radiator under the conditions of Eqs. (4) the expressions

$$\begin{aligned}
(\gamma_f + W)\sigma_{ff}(t\rightarrow\infty) &= W \sum_{\substack{q_1,q_2\\K,Q}} e_{q_1}^{(2)}(e_{-q_2}^{(2)})^*(-1)^{1+K+q_1}[K]^{1/2} \begin{pmatrix} 1 & 1 & K \\ q_1 & q_2 & -Q \end{pmatrix} \sigma_Q^K(ee, t\rightarrow\infty) \\
&\quad - \frac{1}{3}\frac{1}{\hbar^2} |\mathbf{e}_1\cdot\mathbf{e}_2^*|^2 W \frac{|\langle e||\mu||g\rangle \mathscr{E}_1|^2}{\Delta_1(\Delta_1+\Delta_2)} \, , \\
i(\gamma_e + \gamma^{(K)})\sigma_Q^K(ee, t\rightarrow\infty) &= \sum_{\substack{q_1,q_2,Q',\\K'',Q''}} [K,K'']^{1/2} \begin{pmatrix} 1 & 1 & K \\ Q' & -q_1 & -Q \end{pmatrix} \begin{pmatrix} 1 & 1 & K'' \\ Q' & q_2 & -Q'' \end{pmatrix} \sigma_{Q''}^{K''}(ee, t\rightarrow\infty) \\
&\quad \times \left\{ e_{q_1}^{(2)}(e_{-q_2}^{(2)})^*(-1)^{K+K''} \frac{1}{3\hbar^2} \frac{|\langle f||\mu||e\rangle \mathscr{E}_2|^2}{\Delta_2 + i[(\gamma_e+\gamma_f)/2 + \gamma_{fe}(\Delta_2)]} \right. \\
&\quad \left. + (e_{-q_1}^{(2)})^* e_{q_2}^{(2)}(-1)^{1+Q+Q''} \frac{1}{3\hbar^2} \frac{|\langle f||\mu||e\rangle \mathscr{E}_2|^2}{\Delta_2 - i[(\gamma_e+\gamma_f)/2 + \gamma_{fe}(\Delta_2)]} \right\} \\
&\quad + i \sum_{q_1,q_2} e_{q_1}^{(2)}(e_{q_2}^{(2)})^*(-1)^{1+K+q_2}[K]^{1/2} \begin{pmatrix} 1 & 1 & K \\ q_2 & -q_1 & -Q \end{pmatrix} W\sigma_{ff}(t\rightarrow\infty) \\
&\quad + i\frac{\gamma_{f\rightarrow e}}{\sqrt{3}} \delta_{K,0}\delta_{Q,0}\sigma_{ff}(t\rightarrow\infty) \\
&\quad + (\mathbf{e}_1\cdot\mathbf{e}_2^*)^* \sum_{q_1,q_2} e_{q_1}^{(2)} e_{-q_2}^{(1)}(-1)^K[K]^{1/2} \begin{pmatrix} 1 & 1 & K \\ q_2 & -q_1 & -Q \end{pmatrix} \frac{1}{9}\frac{1}{\hbar^4} \\
&\quad \times \frac{|\langle f||\mu||e\rangle \mathscr{E}_2|^2}{\Delta_2 + i[(\gamma_e+\gamma_f)/2 + \gamma_{fe}(\Delta_2)]} \frac{|\langle e||\mu||g\rangle \mathscr{E}_1|^2}{\Delta_1(\Delta_1+\Delta_2)}
\end{aligned} \tag{5}$$

$$+\mathbf{e}_1\cdot\mathbf{e}_2^* \sum_{q_1,q_2} (e_{q_1}^{(2)})^*(e_{q_2}^{(1)})^*(-1)^{1+Q}[K]^{1/2} \begin{bmatrix} 1 & 1 & K \\ q_2 & q_1 & -Q \end{bmatrix} \frac{1}{9}\frac{1}{\hbar^4}$$

$$\times \frac{|\langle f||\mu||e\rangle \mathscr{E}_2|^2}{\Delta_2 - i[(\gamma_e+\gamma_f)/2+\gamma_{fe}(\Delta_2)]} \frac{|\langle e||\mu||g\rangle \mathscr{E}_1|^2}{\Delta_1(\Delta_1+\Delta_2)}$$

$$-i \sum_{q_1,q_2} e_{q_1}^{(1)}(e_{q_2}^{(1)})^*(-1)^{q_2}[K]^{1/2} \begin{bmatrix} 1 & 1 & K \\ -q_1 & q_2 & -Q \end{bmatrix} \begin{Bmatrix} 1 & 1 & K \\ 1 & 1 & 0 \end{Bmatrix}$$

$$\times \frac{1}{\hbar^2} |\langle e||\mu||g\rangle \mathscr{E}_1|^2 \frac{\gamma_e + \Gamma_{eg}^{(K)}(\Delta_1)}{\Delta_1^2} ,$$

with $\sigma_Q^K(ee, t\to\infty) = \langle\langle ee,KQ \mid \sigma(t\to\infty)\rangle\rangle$ and $\sigma_{ff}(t\to\infty) = \langle\langle ff \mid \sigma(t\to\infty)\rangle\rangle$. $|ee,KQ\rangle\rangle$ is an irreducible tetradic vector with respect to the rotation group.[13] In addition to the conditions of Eqs. (4) we assumed in the derivation of Eqs. (5) a spherically symmetric collision environment and a negligible polarizability of the ground state of the radiator. Equations (5) further assume a steady state, which does not deplete the ground-state population. Terms of higher order in the parameter $\gamma/|\Delta_1| \ll 1$ have also been neglected in Eqs. (5). γ_e and γ_f are the total decay rates of states $|e_i\rangle$ and $|f\rangle$ due to spontaneous emission of photons. $\gamma_{f\to e}$ is the branching rate from $|f\rangle$ to $|e_i\rangle$. The collisionally induced decay rate of the optical coherence between states $|e_i\rangle$ and $|g\rangle$ is given by

$$\gamma_{eg}(\Delta_1) = \tfrac{1}{3}N \int d^3p_1 \int d^3p_2\, \rho(p_2) \sum_{\mu} \mathrm{Re} \left[i(\Delta_1 + \tfrac{1}{2}i\gamma_e) \int_0^\infty d\tau \exp[i(\Delta_1+\tfrac{1}{2}i\gamma_e)\tau] \right.$$
$$\left. \times \langle\langle j_e\mu\mathbf{p}_1, j_g 0\mathbf{p}_1 \mid U_I(\tau,0) V_1^I(0,0) \mid j_e\mu\mathbf{p}_2, j_g 0\mathbf{p}_2\rangle\rangle \right] . \tag{6a}$$

$U_I(\tau,0)$ is the tetradic time-evolution operator for the collision between the radiator and one perturber in the interaction picture (for details see Appendix D of Ref. 13). Due to the fact that the first laser is strongly detuned from resonance, i.e., $|\Delta_1|\tau_c \gg 1$, this rate is frequency dependent, which indicates the breakdown of a Markov- (impact) type treatment of the influence of the perturbers on the radiator. The laser photon $(\omega_1,\mathbf{e}_1)$ is absorbed instantaneously during a collision at the Franck-Condon point and $\gamma_{eg}(\Delta_1)$ therefore contains detailed information about the interatomic difference potential and the interaction dynamics around this point. The collisional decay rate $\gamma_{fe}(\Delta_2)$ of the optical coherence between states $|f\rangle$ and $|e_i\rangle$ is defined in an analogous way. However, the second laser photon $(\omega_2,\mathbf{e}_2)$ is absorbed on a long time scale in comparison with the duration of a collision τ_c, i.e., $1/|\Delta_2| \gg \tau_c$, and this quantity has therefore to be evaluated in the impact limit and becomes (approximately) frequency independent. $\gamma^{(K)}$ is a frequency-independent collision rate describing the decay of the orientation ($K=1$) and the alignment ($K=2$) of the excited-state manifold $|e_i\rangle$.[13] As we are neglecting inelastic collisions we have $\gamma^{(K=0)}=0$.[2] The quantity

$$\Gamma_{eg}^{(K)}(\Delta_1) = -\sum_{\substack{\mu_1,\mu_2,\ldots,\mu_5,\\ Q}} (-1)^{\mu_2+\mu_4} \begin{bmatrix} 1 & 1 & K \\ \mu_2 & -\mu_1 & Q \end{bmatrix} \begin{bmatrix} 1 & 1 & K \\ \mu_4 & -\mu_3 & Q \end{bmatrix} N$$

$$\times \int d^3p_1 \int d^3p_2 \int d^3p_3 \int d^3p_4\, \rho(p_4)$$

$$\times 2\Delta_1 \mathrm{Im} \left[\langle\langle j_e\mu_1\mathbf{p}_1, j_e\mu_2\mathbf{p}_1 \mid U_I(\infty,0) \mid j_e\mu_5\mathbf{p}_2, j_e\mu_4\mathbf{p}_3\rangle\rangle \right.$$

$$\times \int_0^\infty d\tau \exp\{i[\Delta_1 + (E(\mathbf{p}_3)-E(\mathbf{p}_2))/\hbar + i\tfrac{1}{2}\gamma_e]\tau\}$$

$$\left. \langle\langle j_e\mu_5\mathbf{p}_2, j_g 0\mathbf{p}_3 \mid U_I(\tau,0) V_1^I(0,0) \mid j_e\mu_3\mathbf{p}_4, j_g O\mathbf{p}_4\rangle\rangle \right] , \tag{6b}$$

describes the instantaneous absorption of the laser photon $(\omega_1,\mathbf{e}_1)$ during the collision according to the Franck-Condon principle (because $1/|\Delta_1| \ll \tau_c$) and the subsequent completion of the collision, which leaves the radiator in that particular linear combination of excited states $|e_i\rangle$, which gives rise to a multiple component K. This quantity has been discussed in detail by Cooper.[7] $\langle e||\mu||g\rangle$ and $\langle f||\mu||e\rangle$ are reduced dipole matrix elements and $e_q^{(i)}$, $i=1,2$ are the spherical components of the laser polarization vectors $\mathbf{e}_i$. The field-induced transition rate between states $|e_i\rangle$ and $|f\rangle$ due to the second laser field is given by

$$W=\frac{1}{3\hbar^2}\,|\langle f||\mu||e\rangle \mathscr{E}_2|^2\frac{\gamma_e+\gamma_f+2\gamma_{fe}(\Delta_2)}{\Delta_2^2+[(\gamma_e+\gamma_f)/2+\gamma_{fe}(\Delta_2)]^2}\;. \tag{6c}$$

Note that there is an additional field-induced coupling between states $|e_i\rangle$ and $|f\rangle$, which is proportional to $\mathbf{e}_1\cdot\mathbf{e}_2^*$ and involves the ground state of the radiator through a stimulated Raman process. If $\mathbf{e}_1\cdot\mathbf{e}_2^*=0$ this stimulated Raman process is forbidden and the corresponding transition rate vanishes.

III. EXAMPLES

In order to demonstrate the influence of the intensity of the second laser field on the stationary final-state population of the radiator, which has been measured in a recent experiment,[12] we discuss now the set of Eqs. (5) for two special types of laser polarizations.

A. Linear polarizations

1. $\mathbf{e}_1=\mathbf{e}_2$

Using a quantization axis parallel to $\mathbf{e}_1$, Eqs. (5) for the stationary reduced density-matrix elements reduce to the following equations for the stationary populations:

$$\begin{aligned}
&(\gamma_e+\tfrac{2}{3}\gamma^{(2)}+W)\sigma_{00}(t\to\infty)\\
&\quad=\tfrac{1}{3}\gamma^{(2)}[\sigma_{++}(t\to\infty)+\sigma_{--}(t\to\infty)]\\
&\qquad+(W+\tfrac{1}{3}\gamma_{f\to e})\sigma_{ff}(t\to\infty)+R+S_0^{(l)}\;,\\[1em]
&(\gamma_e+\tfrac{1}{3}\gamma^{(2)})[\sigma_{++}(t\to\infty)+\sigma_{--}(t\to\infty)]\\
&\quad=\tfrac{2}{3}\gamma^{(2)}\sigma_{00}(t\to\infty)+\tfrac{2}{3}\gamma_{f\to e}\sigma_{ff}(t\to\infty)+2S_1^{(l)}\;,\\[1em]
&(\gamma_f+W)\sigma_{ff}(t\to\infty)=W\sigma_{00}(t\to\infty)-R
\end{aligned} \tag{7a}$$

with $\sigma_{++}(t\to\infty)=\langle\langle e,m_j=+1;e,m_j=+1|\sigma(t\to\infty)\rangle\rangle$, etc. This set of equations is graphically shown in Fig. 2.

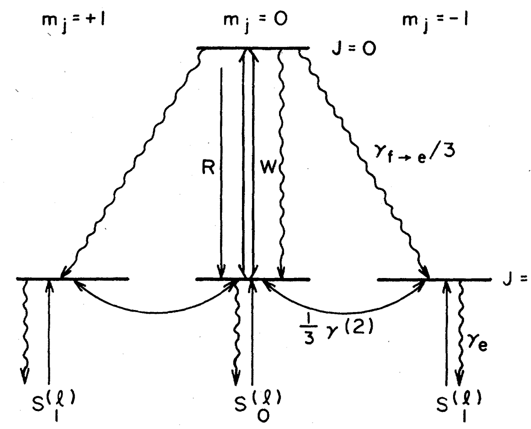

FIG. 2. Rates determining the stationary populations for linear polarizations and $\mathbf{e}_1=\mathbf{e}_2$.

The effect of the first (weak) laser is to populate the excited states $|e_i\rangle$ with the rates

$$S_0^{(l)}=\frac{1}{3\hbar^2}\,|\langle e||\mu||g\rangle\mathscr{E}_1|^2\frac{\gamma_e+\frac{2}{3}\gamma_{eg}(\Delta_1)+\frac{2}{3}\Gamma_{eg}^{(2)}(\Delta_1)}{\Delta_1^2}$$

and

$$S_1^{(l)}=\frac{1}{3\hbar^2}\,|\langle e||\mu||g\rangle\mathscr{E}_1|^2\frac{\frac{2}{3}\gamma_{eg}(\Delta_1)-\frac{1}{3}\Gamma_{eg}^{(2)}(\Delta_1)}{\Delta_1^2}\;. \tag{7b}$$

Through $\Gamma_{eg}^{(2)}(\Delta_1)$ and $\gamma_{eg}(\Delta_1)$ these rates contain detailed information about the collision process due to the fact that the first laser photon $(\omega_1,\mathbf{e}_1)$ is absorbed instantaneously during a collision, i.e., $1/|\Delta_1| \ll \tau_c$.[7,11–13] States $|f\rangle$ and $|e,m_j=0\rangle$ are coupled by the field-induced transition rate W [see Eq. (6c)] due to the second laser. However, there is an additional coupling between these two states described by the rate

$$R=W\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{\Delta_1^2}\;. \tag{7c}$$

It is due to a stimulated Raman process involving the ground state of the radiator and is crucial for our following discussion. Besides these field-induced transition rates the steady-state populations are also determined by the spontaneous decay rates γ_e, γ_f, and $\gamma_{f\to e}$ and the collisional mixing rate $\gamma^{(2)}$ acting within the excited-state manifold $|e_i\rangle$. The stationary final-state population is given by

$$\sigma_{ff}^{\parallel}(t\to\infty)=\left[\gamma_f+W\frac{\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{1}{3}\gamma^{(2)}}(\gamma_e-\frac{1}{3}\gamma_{f\to e})}{\gamma_e\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{1}{3}\gamma^{(2)}}+W}\right]^{-1}\frac{W}{\gamma_e\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{1}{3}\gamma^{(2)}}+W}$$

$$\times\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{\Delta_1^2}\frac{2}{3}\left[\gamma_{eg}(\Delta_1)\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{1}{3}\gamma^{(2)}}+[\Gamma_{eg}^{(2)}(\Delta_1)-\gamma^{(2)}]\frac{\gamma_e}{\gamma_e+\frac{1}{3}\gamma^{(2)}}\right]. \tag{8}$$

As soon as $W\gtrsim\gamma_e$ the final-state population will exhibit a *nonlinear* dependence on the intensity of the second laser due to the fact that the transition from $|e,m_j=0\rangle$ to $|f\rangle$ is saturated.

2. $\mathbf{e}_l\perp\mathbf{e}_2$

Again we choose our quantization axis parallel to $\mathbf{e}_1$ but $\mathbf{e}_2$ is now perpendicular to this direction. From Eqs. (5) we find for the stationary populations the set of equations

$$(\gamma_e+\tfrac{2}{3}\gamma^{(2)})\sigma_{00}(t\to\infty)$$
$$=\tfrac{1}{3}\gamma^{(2)}[\sigma_{++}(t\to\infty)+\sigma_{--}(t\to\infty)]$$
$$+\tfrac{1}{3}\gamma_{f\to e}\sigma_{ff}(t\to\infty)+S_0^{(l)},$$
$$(\gamma_e+\tfrac{1}{3}\gamma^{(2)}+\widetilde{W})[\sigma_{++}(t\to\infty)+\sigma_{--}(t\to\infty)]$$
$$=\tfrac{2}{3}\gamma^{(2)}\sigma_{00}(t\to\infty)$$
$$+2(\tfrac{1}{3}\gamma_{f\to e}+\widetilde{W})\sigma_{ff}(t\to\infty)+2S_1^{(l)}, \tag{9a}$$

$$(\gamma_f+2\widetilde{W})\sigma_{ff}(t\to\infty)=\widetilde{W}[\sigma_{++}(t\to\infty)+\sigma_{--}(t\to\infty)],$$

which are graphically represented in Fig. 3.

The excited states are again populated by the weak first laser with rates $S_0^{(l)}$ and $S_1^{(l)}$. The effective stimulated transition rate due to the second laser,

$$\widetilde{W}=\tfrac{1}{2}W\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\gamma^{(2)}+\frac{1}{2}W}, \tag{9b}$$

connects the final state with the excited states $|e,m_j=\pm1\rangle$. Contrary to the case of parallel polarizations there is now no additional field-induced coupling R between the final the excited states due to the fact that a stimulated Raman transition involving the ground state is forbidden because $\mathbf{e}_1\cdot\mathbf{e}_2^*=0$. The stationary final-state population is given by

$$\sigma_{ff}^{\perp}(t\to\infty)=\left[\gamma_f+W\frac{\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{1}{3}\gamma^{(2)}}(\gamma_e-\frac{1}{3}\gamma_{f\to e})}{\gamma_e\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{1}{3}\gamma^{(2)}}+W}\right]^{-1}\frac{W}{\gamma_e\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{1}{3}\gamma^{(2)}}+W}$$

$$\times\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{\Delta_1^2}\frac{2}{3}\left[\gamma_{eg}(\Delta_1)\frac{\gamma^{(2)}+\gamma_e}{\gamma_e+\frac{1}{3}\gamma^{(2)}}-\frac{\frac{1}{2}\gamma_e}{\gamma_e+\frac{1}{3}\gamma^{(2)}}[\Gamma_{eg}^{(2)}(\Delta_1)-\gamma^{(2)}]\right]. \tag{10}$$

It is interesting to note that the polarization-dependent quantity

$$p_l=\frac{\sigma_{ff}^{\parallel}(t\to\infty)-\sigma_{ff}^{\perp}(t\to\infty)}{\sigma_{ff}^{\parallel}(t\to\infty)+\sigma_{ff}^{\perp}(t\to\infty)}=\frac{3\alpha^{(2)}(\Delta_1)}{2(1+\gamma^{(2)}/\gamma_e)+\alpha^{(2)}(\Delta_1)} \tag{11}$$

is *completely independent of the intensity of the second laser,* even if the transition $|e_1\rangle\to|f\rangle$ is saturated. Note that $\alpha^{(K)}(\Delta_1)$ is defined by the relation

$$\Gamma_{eg}^{(K)}(\Delta_1)-\gamma^{(K)}=2\gamma_{eg}(\Delta_1)\alpha^{(K)}(\Delta_1). \tag{12}$$

The neglect of inelastic collisions implies $\alpha^{(K=0)}(\Delta_1)=1$. If the completion of a collision were unimportant, i.e., $U_I(\infty,0)=1$ in Eq. (6b), $\alpha^{(K)}(\Delta_1)=1$ and the polarization dependence p_l is completely determined by subsequent collisions, i.e., by the ratio $\gamma^{(2)}/\gamma_e$. In the extreme opposite limit, where $\alpha^{(K)}(\Delta_1)=0$, the completion of a collision is extremely important and leads to $p_l=0$ (see the discussion by Cooper in Ref. 7).

B. Circular polarizations

1. $\mathbf{e}_l = \mathbf{e}_2$

We choose both laser fields to be circularly polarized in the same direction. For σ_+ polarizations we find from Eqs. (5) for the stationary populations of the radiator the following set of equations:

$$
\begin{aligned}
&(\gamma_e+W+\tfrac{1}{2}\gamma^{(1)}+\tfrac{1}{6}\gamma^{(2)})\sigma_{--}(t\to\infty)=(W+\tfrac{1}{3}\gamma_{f\to e})\sigma_{ff}(t\to\infty)+\tfrac{1}{2}(\gamma^{(1)}-\tfrac{1}{3}\gamma^{(2)})\sigma_{++}(t\to\infty)+\tfrac{1}{3}\gamma^{(2)}\sigma_{00}(t\to\infty)+S_-^{(c)}\ ,\\
&(\gamma_e+\tfrac{2}{3}\gamma^{(2)})\sigma_{00}(t\to\infty)=\tfrac{1}{3}\gamma_{f\to e}\sigma_{ff}(t\to\infty)+\tfrac{1}{3}\gamma^{(2)}[\sigma_{--}(t\to\infty)+\sigma_{++}(t\to\infty)]+S_0^{(c)}\ , \qquad (13a)\\
&(\gamma_e+\tfrac{1}{2}\gamma^{(1)}+\tfrac{1}{6}\gamma^{(2)})\sigma_{++}(t\to\infty)=\tfrac{1}{3}\gamma_{f\to e}\sigma_{ff}(t\to\infty)+\tfrac{1}{2}(\gamma^{(1)}-\tfrac{1}{3}\gamma^{(2)})\sigma_{--}(t\to\infty)+\tfrac{1}{3}\gamma^{(2)}\sigma_{00}(t\to\infty)+S_+^{(c)}\\
&(\gamma_f+W)\sigma_{ff}(t\to\infty)=W\sigma_{--}(t\to\infty)\ ,
\end{aligned}
$$

which are graphically represented in Fig. 4. The weak first laser populates the excited states with rates

$$
\begin{aligned}
S_+^{(c)}&=\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathcal{E}_1|^2}{\Delta_1^2}\\
&\quad\times[\gamma_e+\tfrac{2}{3}\gamma_{eg}(\Delta_1)+\tfrac{1}{2}\Gamma_{eg}^{(1)}(\Delta_1)+\tfrac{1}{6}\Gamma_{eg}^{(2)}(\Delta_1)]\ ,\\
S_-^{(c)}&=\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathcal{E}_1|^2}{\Delta_1^2} \qquad (13b)\\
&\quad\times[\tfrac{2}{3}\gamma_{eg}(\Delta_1)-\tfrac{1}{2}\Gamma_{eg}^{(1)}(\Delta_1)+\tfrac{1}{6}\Gamma_{eg}^{(2)}(\Delta_1)]\ ,\\
S_0^{(c)}&=\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathcal{E}_1|^2}{\Delta_1^2}\frac{1}{3}[2\gamma_{eg}(\Delta_1)-\Gamma_{eg}^{(2)}(\Delta_1)]\ .
\end{aligned}
$$

The second laser induces a transition between $|e_i\rangle$ and $|f\rangle$ described by the rate W of Eq. (6c). Due to the circular polarizations of both lasers an orientation is induced in the excited-state manifold $|e_i\rangle$, which decays with a collisional rate $\gamma^{(1)}$. The other incoherent decay rates are the same as in Eq. (7a). For σ_- polarizations, Eqs. (13a) still apply provided we made the replacements

$$
\begin{aligned}
&\sigma_{--}(t\to\infty)\to\sigma_{++}(t\to\infty)\ ,\\
&\sigma_{++}(t\to\infty)\to\sigma_{--}(t\to\infty)\ .
\end{aligned}
$$

From these equations we find for the stationary final-state population the expression

$$
\begin{aligned}
\sigma_{ff}^{||}(t\to\infty)=&\left[\gamma_f+W\frac{(\gamma_e+\gamma^{(1)})\dfrac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}(\gamma_e-\frac{1}{3}\gamma_{f\to e})}{W\left[\gamma_e+\frac{1}{2}\gamma^{(1)}+\frac{1}{6}\gamma^{(2)}\dfrac{\gamma_e}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\right]+(\gamma_e+\gamma^{(1)})\dfrac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\gamma_e}\right]^{-1}\\
&\times\frac{W}{W\left[\gamma_e+\frac{1}{2}\gamma^{(1)}+\frac{1}{6}\gamma^{(2)}\dfrac{\gamma_e}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\right]+(\gamma_e+\gamma^{(1)})\dfrac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\gamma_e}\\
&\times\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathcal{E}_1|^2}{\Delta_1^2}\left[2\gamma_{eg}(\Delta_1)[\tfrac{1}{3}(\gamma^{(1)}+\gamma_e)]\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}-\tfrac{1}{2}[\Gamma_{eg}^{(1)}(\Delta_1)-\gamma^{(1)}]\frac{\gamma_e(\gamma_e+\gamma^{(2)})}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\right.\\
&\left.\qquad+\tfrac{1}{6}[\Gamma_{eg}^{(2)}(\Delta_1)-\gamma^{(2)}]\frac{\gamma_e(\gamma_e+\gamma^{(1)})}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\right]\ , \qquad (14)
\end{aligned}
$$

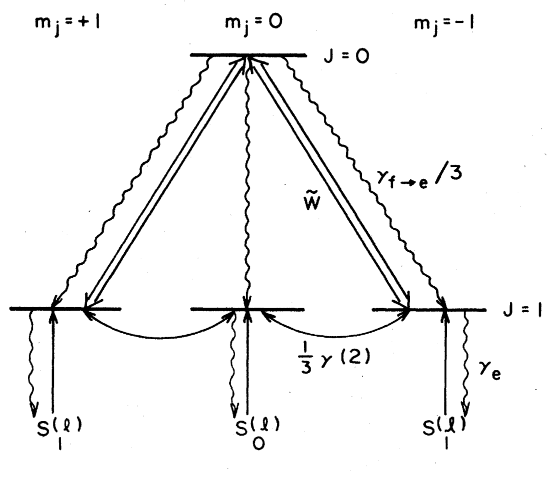

FIG. 3. Same as Fig. 2 but with $e_1 \perp e_2$.

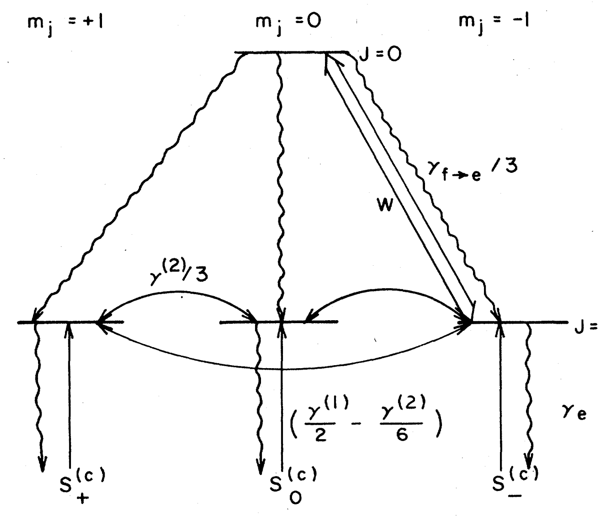

FIG. 4. Rates determining the stationary populations for circular polarizations with e_1 σ_+ polarized and $e_1 = e_2$.

which will exhibit a *nonlinear* dependence on the intensity of the second laser as soon as $W \gtrsim \gamma_e$ and the transition between states $|e, m_j=-1\rangle$ and $|f\rangle$ starts to saturate.

2. $e_2 = e_1^*$

We consider now the case where the helicities of both laser fields are different. Choosing again σ_+ polarization for the first laser we obtain the following equations for the stationary populations of the radiator:

$$\begin{aligned}
&(\gamma_e+W+\tfrac{1}{2}\gamma^{(1)}+\tfrac{1}{6}\gamma^{(2)})\sigma_{++}(t\to\infty) \\
&\quad =(W+\tfrac{1}{3}\gamma_{f\to e})\sigma_{ff}(t\to\infty) \\
&\qquad +\tfrac{1}{2}(\gamma^{(1)}-\tfrac{1}{3}\gamma^{(2)})\sigma_{--}(t\to\infty) \\
&\qquad +\tfrac{1}{3}\gamma^{(2)}\sigma_{00}(t\to\infty)+S_+^{(c)}+R\ , \\
&(\gamma_e+\tfrac{1}{2}\gamma^{(1)}+\tfrac{1}{6}\gamma^{(2)})\sigma_{--}(t\to\infty) \\
&\quad =\tfrac{1}{3}\gamma_{f\to e}\sigma_{ff}(t\to\infty)+\tfrac{1}{2}(\gamma^{(1)}-\tfrac{1}{3}\gamma^{(2)})\sigma_{++}(t\to\infty) \\
&\qquad +\tfrac{1}{3}\gamma^{(2)}\sigma_{00}(t\to\infty)+S_-^{(c)}\ , \\
&(\gamma_e+\tfrac{2}{3}\gamma^{(2)})\sigma_{00}(t\to\infty) \\
&\quad =\tfrac{1}{3}\gamma_{f\to e}\sigma_{ff}(t\to\infty) \\
&\qquad +\tfrac{1}{3}\gamma^{(2)}[\sigma_{++}(t\to\infty)+\sigma_{--}(t\to\infty)]+S_0^{(c)}\ , \\
&(\gamma_f+W)\sigma_{ff}(t\to\infty)=W\sigma_{++}(t\to\infty)-R\ ,
\end{aligned} \tag{15}$$

which are graphically represented in Fig. 5. The significant difference from Eqs. (13a) is the appearance of the Raman coupling between $|f\rangle$ and $|e, m_j=+1\rangle$, which involves the ground state and is described by the rate R of Eq. (7c). If we made the replacements

$$\sigma_{++}(t\to\infty)\to\sigma_{--}(t\to\infty)\ ,$$
$$\sigma_{--}(t\to\infty)\to\sigma_{++}(t\to\infty)\ ,$$

Eqs. (15) would describe the case where the fist laser is σ_- and the second laser σ_+ polarized. The stationary final-state population is given by

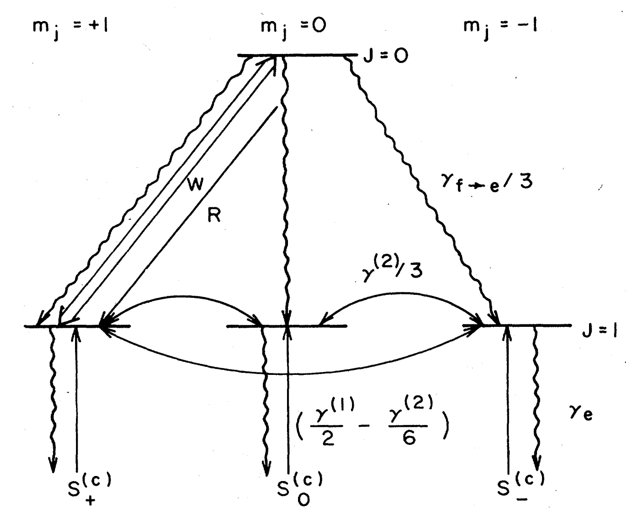

FIG. 5. Same as Fig. 4 but with $e_2 = e_1^*$.

$$\sigma^{\perp}_{ff}(t\to\infty)=\left[\gamma_f+W\frac{(\gamma_e+\gamma^{(1)})\dfrac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}(\gamma_e-\frac{1}{3}\gamma_{f\to e})}{W\left[\gamma_e+\frac{1}{2}\gamma^{(1)}+\frac{1}{6}\gamma^{(2)}\dfrac{\gamma_e}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\right]+(\gamma_e+\gamma^{(1)})\dfrac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\gamma_e}\right]^{-1}$$

$$\times\frac{W}{W\left[\gamma_e+\frac{1}{2}\gamma^{(1)}+\frac{1}{6}\gamma^{(2)}\dfrac{\gamma_e}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\right]+(\gamma_e+\gamma^{(1)})\dfrac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\gamma_e}$$

$$\times\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{\Delta_1^2}\left[2\gamma_{eg}(\Delta_1)[\tfrac{1}{3}(\gamma_e+\gamma^{(1)})]\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}+[\Gamma^{(1)}_{eg}(\Delta_1)-\gamma^{(1)}]\frac{1}{2}\frac{\gamma_e+\gamma^{(2)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\gamma_e\right.$$

$$\left.+[\Gamma^{(2)}_{eg}(\Delta_1)-\gamma^{(2)}]\frac{1}{6}\frac{\gamma_e+\gamma^{(1)}}{\gamma_e+\frac{2}{3}\gamma^{(2)}}\gamma_e\right]. \tag{16}$$

Similarly, as in the case of linear polarizations the polarization-dependent quantity,

$$p_c=\frac{\sigma^{\perp}_{ff}(t\to\infty)-\sigma^{\parallel}_{ff}(t\to\infty)}{\sigma^{\perp}_{ff}(t\to\infty)+\sigma^{\parallel}_{ff}(t\to\infty)}=\frac{3\alpha^{(1)}(\Delta_1)\gamma_e/(\gamma_e+\gamma^{(1)})}{2+\alpha^{(2)}(\Delta_1)\dfrac{\gamma_e}{(\gamma_e+\gamma^{(2)})}} \tag{17}$$

is *completely independent of the intensity of the second laser even if* $W\gtrsim\gamma_e$.

We want to point out that we have neglected terms of higher order in the parameter $\gamma/|\Delta_1|\ll 1$ in Eq. (5). These terms are negligible in Eqs. (8) and (16) for the final-state populations as long as the density of perturbers is not too small, i.e.,

$$\frac{\gamma_{eg}(\Delta_1)}{(\max\{\gamma_s,|\Delta_2|\})^2}\gg\frac{\gamma_s}{\Delta_1^2}, \tag{18}$$

where γ_s is a typical spontaneous decay rate. This condition implies that the dominant contribution to the final-state population comes from the two-step absorption process and the contribution from the direct two-photon absorption process in negligible.[16] Looking at the spontaneously emitted radiation corresponding to the transition of the radiator from its final state $|f\rangle$ to some intermediate state, the two-photon absorption term determines the area of the Raman peak described in Ref. 13, whereas the two-step absorption contribution determines the area of the Rayleigh- and the two-photon redistributed peak.

Alford *et al.*[12] have measured the polarization-dependent quantity p_c of Eq. (17) in their two-step absorption experiment and indeed found p_c to be intensity independent even in cases where the final-state population exhibited a nonlinear dependence on the intensity of the second laser.

IV. CONCLUSIONS

We have studied intensity effects in two-photon collisional redistribution of radiation for a case of recent experimental interest, namely a situation where the first laser is detuned to the quasistatic wing and the second laser is almost on resonance (impact limit). The first laser was effectively weak and the second laser was allowed to saturate the transition from the excited to the final state. Motivated by a recent experiment[12] we discussed the stationary final-state population of the excited atom for various polarizations of both laser fields for a $J=0\to J=1\to J=0$ type transition. In particular, we found that the polarization-dependent quantities p_l and p_c, which are convenient quantities for studying details of the collision between the radiator and a perturber, are independent of the laser intensities even for cases where the transition from the excited to the final state is saturated by the second laser field. This is consistent with the experimental findings of Alford *et al.*,[12] who studied the case of circular polarizations. Our equations further showed that there is an effective coupling between the final and the excited state brought about by a stimulated Raman process, which involves the ground state. This process leads to the fact that the two-step contribution to the stationary final-state population vanishes in the absence of collisions (see also the discussion in Ref. 16 for the nondegenerate case). We stress that if this rate R were not included in the equations, we would have failed to get an intensity-independent quantity p_c or p_l. However, it is not clear at this stage if a similar result also holds for situations where different angular momenta are involved.

In our treatment we have neglected the motion of the radiator. Let us now discuss qualitatively which kind of effects the velocity of the radiator might give rise to. As the first laser is strongly detuned from resonance, no particular velocity class of the radiator is excited by the first laser transition. This also applies for the second excitation step from $|e_i\rangle$ to $|f\rangle$ as long as $\max\{\gamma, |\Delta_2|\} \gg (\omega_2/c)\langle v\rangle$. $\langle v\rangle$ is thereby the mean velocity of the radiator. As soon as our second laser excites a particular velocity class, because $\gamma, |\Delta_2| \lesssim (\omega_2/c)\langle v\rangle$, things become somewhat more complicated, because in general we also have to properly take into account both "hole" burning and velocity-changing collisions.[17,18] However, assuming that effects due to velocity-changing collisions are unimportant in the sense that the rate of velocity-changing collisions is small compared to the radiative decay rates, the only effect due to the velocity of the perturber is then to change $\Delta_2 \rightarrow \Delta_2 \pm (\omega_2/c)v$ in the field-induced transition rate W of Eq. (6c). Though this would affect the velocity-integrated final-state populations for the various polarizations of the laser fields, it would leave the ratios p_l and p_c of Eqs. (11) and (17) unaffected. The investigations of Berman *et al.*[18] also indicate that velocity-changing effects are expected to be small. We note that these effects are unlikely to be important for the experiments performed by Alford *et al.*[12] since there the spontaneous decay rates γ_e and γ_f are comparable to the collisional rates $\gamma_{fe}(\Delta_2)$, $\gamma^{(2)}$, etc. and the velocity-changing rates are expected to be small compared to the destruction of coherence and collisional mixing rates.

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