Collisional redistribution by laser-induced fluorescence in the Ba-Ar system

M. Belsley* and J. Cooper
Joint Institute for Laboratory Astrophysics, University of Colorado and National Bureau of Standards and Department of Physics, University of Colorado, Boulder, Colorado 80309-0440
(Received 6 October 1986)

The effects due to the presence of a (possibly intense) probe laser, which is used to interrogate the asymptotic excited-m_j-state distribution resulting from the evolution through a half-collision, are systematically studied. Experimental results are obtained which are in good qualitative agreement with predictions based on a recent theoretical treatment. Finally, a simple interpretation of the relevant physical processes involved is suggested by considering the problem within a dressed-state representation.

I. INTRODUCTION

Recent experimental[1] and theoretical[2] investigations have generalized the usual one-photon collisional redistribution experiments to study the scattering of two laser fields by an atom undergoing collisions with a gas of foreign perturbers. In the experimental study undertaken by Alford *et al.*,[1] the first laser induced a transition from the Ba ground state ($6s^2\,{}^1S_0$) to the excited $6s\,6p\,{}^1P_1$ state. As is common in the case of one-photon collisional redistribution experiments, this laser was detuned so far from the atomic transition frequency that absorption occurred only during a strong collision with a rare-gas perturber, on a timescale short compared to the duration of the collision. Under this condition the Markovian description of the collisional interaction breaks down[3] and the details of the collisional processes can be probed. In particular the probability of absorption of light of a given frequency can be directly related to local characteristics of the interatomic potentials.[4] By varying the frequency then, one can vary the internuclear separation at which absorption occurs. Furthermore, the asymptotic m_j-state distribution can provide detailed information about the collisional evolution from the point of absorption to the completion of the collision. Ordinarily this latter information is reported in terms of the polarization properties of the fluorescence from the excited state through the quantity P,

$$P=\frac{I_{||}-I_{\perp}}{I_{||}+I_{\perp}}\ , \tag{1}$$

which is commonly called the polarization. Here $I_{||}$ and $I_{\perp}$ refer to the intensities of the fluorescent light polarized parallel and perpendicular to the polarization of the incident laser. For a $J=0$ to $J=1$ transition, a polarization of unity signifies that the completion of the collision is unimportant. In the extreme opposite limit, a polarization of zero implies that complete mixing occurs as the collision partners fly apart. Determining the polarization as a function of detuning provides a convenient means for studying the mixing of the asymptotic states, due to the collisional dynamics, from a localized point of absorption to the completion of the collision.

Rather than monitor the fluorescence from the excited state directly, Alford *et al.*[1] used a second laser to induce an almost resonant transition from the excited state to a final $6s\,8s\,{}^1S_0$ state. In this manner the excited-^1P-state manifold was probed, and the dependence of the total fluorescence from the final state on the relative polarizations of the two lasers was used to form the polarization P. At the same time problems associated with the degradation of the polarization due to radiative trapping effects[5] were avoided to a large extent. Surprisingly, the polarization was found to be independent of the second laser's intensity, even when the upper transition was saturated. Subsequently, Alber and Cooper[6] derived theoretical expressions in agreement with these observations. Allowing for arbitrary intensities of the second laser within the restriction that the power is never large enough to alter the collision dynamics (i.e., the on-resonance Rabi frequency of either laser was assumed to be much less than the inverse duration of a strong collision), their results predict intensity independent polarization for the redistribution of both linearly and circularly polarized light. A crucial aspect of their calculation was the appearance of a two-photon "Raman-like" term when the two lasers were polarized along the same direction. Although not well understood at the time, the inclusion of this term was necessary to obtain intensity-independent polarization expressions.

In this work we present a physical interpretation of the "Raman" term, which is suggested by viewing the problem in a dressed-state basis. In addition we present a systematic experimental study of the effect of the second laser on the fluorescence from both the final ^1S state and the excited ^1P state. Our data on the polarization dependence of the final-state fluorescence confirm the initial observations of Alford *et al.*,[1] which were made at a single buffer-gas pressure and rather modest detunings (i.e., within the impact limit). The polarization dependence of the excited-state fluorescence is in good qualitative agreement with theoretical expressions which we derive based on the work of Alber and Cooper.[6]

This paper is organized as follows: In Sec. II we present the theoretical expressions of Alber and Cooper[6] and discuss our interpretation of the Raman term. Expressions describing the expected dependence of the excited-state fluorescence are also given in Sec. II. A brief description of the experimental setup is given in Sec. III. The experimental results themselves along with a discussion and some final remarks form the body of Sec. IV.

II. THEORY

From the point of view of scattering theory the one-photon redistribution process is an off-shell event since it is the collision which makes up the energy defect of the absorbed photon with respect to the atomic resonance. In fact it is really a three-body problem, the third body being the incoming or outgoing photon. However, there is the additional complication, over the usual scattering problem, that the creation and decay of the atomic coherences are important. What is needed then is a scattering theory for the density operator and this is what has been developed.[7–10]

Alber and Cooper[2] have generalized the methods of Ref. 7 to allow for two incident laser fields. The approximations they invoke are those common in the field of neutral line broadening. They consider a neutral atom surrounded by N structureless perturbers (usually rare-gas atoms). The binary-collision approximation is assumed valid so that at any given time the atom interacts strongly with at most one perturber. Time-ordered sequences of such collisions are fully accounted for within their formalism. The external radiation fields are taken to be classical, with electric field amplitudes $\mathscr{E}_1$ and $\mathscr{E}_2$, and their interaction with the atomic radiator is described within the dipole and rotating-wave approximations. The radiator is considered to be an effective three-level atom, the possibility of ionization and small ac Stark shifts due to other (nonresonant) levels being specifically neglected. Spontaneous emission is treated within the Markov approximation. A spherically symmetric collisional environment is assumed as well as negligible polarizability of the ground atomic state. In addition, all effects due to the motion of the radiator are neglected, although this point will be the subject of some comment below.

The technique of using a second near-resonant laser, as a probe of the excited-state populations, to obtain polarization information is shown schematically in Fig. 1. The Rabi frequencies of the two lasers Ω_1 and Ω_2 are constrained as follows:

$$|\Omega_1|, |\Omega_2| \ll 1/\tau_c , \tag{2a}$$

$$|\Omega_1|, |\Omega_2| \ll |\Delta_1| , \tag{2b}$$

$$|\Omega_1|^2 \ll |\Delta_1| \max\{|\Delta_2|, \gamma\} , \tag{2c}$$

where γ is a typical spontaneous decay or collisional rate, and τ_c is the duration of a strong collision. The first condition ensures that the influence of either laser field on the collisional process is weak. The second and third assumptions imply that the influence of the first laser on the radiator is also weak and can be treated perturbatively. In addition the detunings of the two lasers are taken to be such that

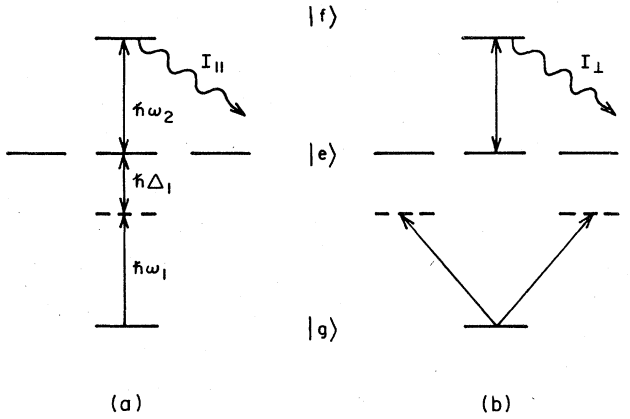

FIG. 1. The pump-probe technique for measuring the polarization in a $J=0$ to $J=1$ collisional redistribution experiment. (a) The total fluorescence from $|f\rangle$ when the pump- and probe-laser polarizations are parallel yields $I_{\|}$. (b) Turning the pump-laser polarization perpendicular to the probe-laser polarization gives $I_{\perp}$.

$$|\Delta_1| \gg 1/\tau_c, \gamma , \tag{3a}$$

$$|\Delta_2| \ll 1/\tau_c . \tag{3b}$$

Thus photons from the first laser field are absorbed only during a collision on a timescale very much shorter than the duration of such a collision; precisely the conditions of one-photon redistribution experiments. The timescale for absorption of photons from the second field is, however, long compared to τ_c, and situations in which the excited- to final-state transition is saturated are explicitly included in the calculation (i.e., $|\Omega_2| \gtrsim \max\{|\Delta_2|, \gamma\}$).

Under these conditions the ground-state population will remain undepleted to a good approximation and the steady-state solution of the equation of motion for the density operator leads to a set of rate equations for the atomic populations.[6] For the case shown in Fig. 1(a) these are, using the corresponding notation of Ref. 6,

$$(\gamma_e + \tfrac{2}{3}\gamma^{(2)} + W)\sigma_{00} = \tfrac{1}{3}\gamma^{(2)}(\sigma_{++} + \sigma_{--}) + (W + \tfrac{1}{3}\gamma_{f\to e})\sigma_{ff} + R + S_0 , \tag{4a}$$

$$(\gamma_e + \tfrac{1}{3}\gamma^{(2)})(\sigma_{++} + \sigma_{--}) = \tfrac{2}{3}\gamma^{(2)}\sigma_{00} + \tfrac{2}{3}\gamma_{f\to e}\sigma_{ff} + 2S_1 , \tag{4b}$$

$$(\gamma_f + W)\sigma_{ff} = W\sigma_{00} - R , \tag{4c}$$

which are graphically shown in Fig. 2. Here $\sigma_{++} = \langle\langle e, m_j = +1; e, m_j = +1 | \sigma(t\to\infty)\rangle\rangle$, etc., with $\sigma(t\to\infty)$ the steady-state density operator;

$$S_0 = \frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle \mathscr{E}_1|^2}{\Delta_1^2}[\gamma_e + \tfrac{2}{3}\gamma_{eg}(\Delta_1) + \tfrac{2}{3}\Gamma_{eg}^{(2)}(\Delta_1)] , \tag{5a}$$

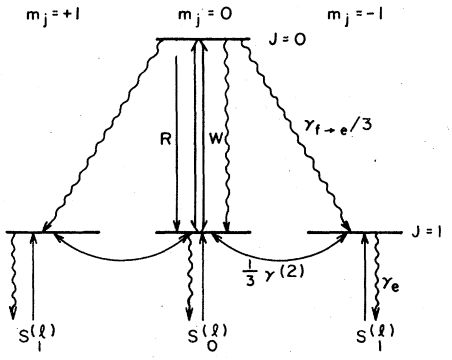

FIG. 2. Rates determining the stationary populations (Ref. 6) for the situation of Fig. 1(a).

$$S_1=\frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{\Delta_1^2}[\tfrac{2}{3}\gamma_{eg}(\Delta_1)-\tfrac{1}{3}\Gamma_{eg}^{(2)}(\Delta_1)] \quad (5b)$$

are the rates associated with populating the excited-state manifold due to the interaction with the first laser field. Because of the large detuning of the first laser, both $\gamma_{eg}(\Delta_1)$ and $\Gamma_{eg}^{(2)}(\Delta_1)$ contain detailed information about the collisional dynamics. The collisionally induced decay rate of the ground-state—excited-state coherence $\gamma_{eg}(\Delta_1)$, contains explicit information about the interatomic different potential near the point of absorption. The quantity $\Gamma_{eg}^{(2)}(\Delta_1)$ describes the instantaneous absorption of a photon from the first laser field during a collision according to the Franck-Condon principle, and the subsequent propagation through the collision to the final excited-m-state distribution giving rise to an asymptotic atomic alignment.[11] The rate of decay of this alignment due to subsequent collisions is given by $\gamma^{(2)}$.

The induced transition rate between the excited and final state is given by

$$W=\frac{1}{3\hbar^2}|\langle f||\mu||e\rangle\mathscr{E}_2|^2 \times\frac{\gamma_e+\gamma_f+2\gamma_{fe}(\Delta_2)}{\Delta_2^2+\left[\frac{(\gamma_e+\gamma_f)}{2}+\gamma_{fe}(\Delta_2)\right]^2}. \quad (6)$$

Here the decay of the excited-state—final-state coherence $\gamma_{fe}(\Delta_2)$, reduces to its (approximately) detuning independent impact value owing to the small detuning of the second laser. $\langle e||\mu||g\rangle$ and $\langle f||\mu||e\rangle$ are reduced dipole matrix elements. γ_e and $\gamma_{f\to e}$ are spontaneous decay rates while γ_f is the total decay rate of the final state.

An additional coupling between the excited and final states,

$$R=W\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{3\hbar^2\Delta_1^2}, \quad (7)$$

is the Raman-type process alluded to in the Introduction. Its presence is crucial to obtaining intensity-independent polarizations. We caution, however, that this term does not represent the Stokes radiation from the usual two-photon Raman process, which absorbs a photon from each field and emits a spontaneous photon to leave the system in its excited state $|e\rangle$. That process has a field dependence of $(|\Omega_1|^2/\Delta_1^2)[|\Omega_2|^2/(\Delta_1+\Delta_2)^2]$, and is down by order $(\Omega/\Delta_1)^2$ compared to terms kept in the calculation of Alber and Cooper.[6]

One can develop some physical insight into the nature of the rate R by considering one-photon collisional redistribution in more detail. This is illustrated for the case of a $J=0$ to $J=1$ transition in Fig. 3. We choose the quantization axis of the system to lie along the direction of polarization of the detected light. The two intensities $I_{\|}$ and $I_{\perp}$ are then obtained by rotating the polarization of the incident laser to be first parallel and then perpendicular to this axis. Even in the absence of collisions there is a small amplitude for being in the excited state which gives rise to Rayleigh scattering. In the usual perturbative interpretation this is described in terms of scattering off of a virtual level [dashed line in Fig. 3(a)]. However, this virtual state is not part of the atomic basis set and its contribution is hidden in the excited-state density matrix elements. When the scattered intensity polarized parallel to the laser's polarization is monitored, the Rayleigh scattering is discriminated against (typically through the use of a monochromator) and the observed signal is proportional to

$$I_{\|}\propto\left[\sigma_{00}-\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{3\hbar^2\Delta_1^2}\right]. \quad (8)$$

The difficulties associated with the virtual level can be circumvented by going over to a dressed-state basis.[12] In this approach one considers as a basis set the compound "atom + laser" system by diagonalizing the interaction of the atom with the radiation field, thereby "dressing" the bare atomic states. For weak fields this basis consists of a ladder of dressed states

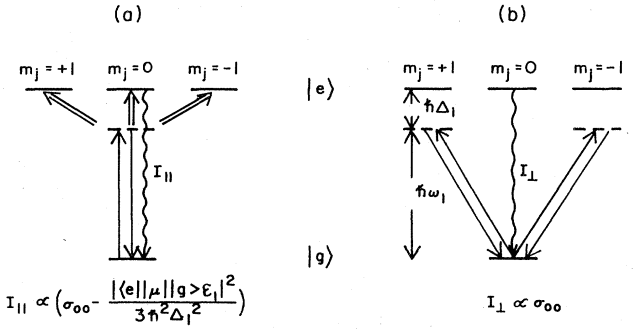

FIG. 3. In a typical one-photon redistribution experiment to fluorescence from the $m_j=0$ excited state is monitored. Double arrows denote collisional interaction. Note that for the conditions of Fig. 3(a) the Rayleigh scattering is discriminated against and $I_{\|}$ is not proportional to the full $m_j=0$ population.

$$|\mathrm{I};N\rangle = |g;N+1\rangle - \Omega_1/2\Delta_1 |e,m_j=0;N-1\rangle \ , \tag{9a}$$

$$|\mathrm{II};N\rangle = \Omega/2\Delta_i |g;N+1\rangle + |e,m_j=0;N-1\rangle \ , \tag{9b}$$

with energies

$$E_{\mathrm{I}} = E_g + (N+1)\hbar\omega_1 \ , \tag{10a}$$

$$E_{\mathrm{II}} = E_g + N\hbar\omega_1 \ . \tag{10b}$$

Here N refers to the number of photons in the applied radiation field. The $m_j = \pm 1$ excited states are not coupled to the ground state by the first laser field for the situation depicted in Fig. 3(a) so that in the transformation to the dressed-state basis they simply become product states of the bare atomic states and the number state of the applied radiation field. Collisions that are taken to be elastic in the bare-state description, couple dressed states within the nearly degenerate manifold of a given N. As shown in Fig. 4, spontaneous emission induces transitions as optical frequencies between adjacent manifolds. In this basis the virtual state of the bare-state description becomes the dressed state $|\mathrm{I};N\rangle$, while collisional excitation from this virtual level to the excited state is the real inelastic transition from $|\mathrm{I};N\rangle$ to $|\mathrm{II};N\rangle$ (see Fig. 4). Fluorescence results from the spontaneous emission of $|\mathrm{II};N\rangle$ down to $|\mathrm{I};N-1\rangle$, while Rayleigh scattering corresponds to the transition $|\mathrm{I};N\rangle$ to $|\mathrm{I};N-1\rangle$. (The other possible transitions $|\mathrm{II};N\rangle \rightarrow |\mathrm{II};N-1\rangle$ and $|\mathrm{I};N\rangle \rightarrow |\mathrm{II};N-1\rangle$ are negligible for weak fields.) Thus the dressed-state description conveniently separates the two different processes which combine to form the total bare-state population σ_{00}.

Viewed in terms of the above statements the net effect of the Raman rate R is clear. It reduces the effective population probed by the second laser from the total $m_j = 0$ state population to an effective population

$$\sigma_{00}^{\mathrm{eff}} = \left[\sigma_{00} - \frac{|\langle e||\mu||g\rangle \mathscr{E}_1|^2}{3\hbar^2\Delta_1^2}\right] . \tag{11}$$

This effective population is of course the $|\mathrm{II}\rangle$ dressed-state population and is precisely that which gives rise to the monitored signal in the one-photon redistribution experiment. In fact, if we consistently replace σ_{00} by $\sigma_{00}^{\mathrm{eff}}$ in Eq. (4) we obtain the following set of rate equations:

$$(\gamma_e + \tfrac{2}{3}\gamma^{(2)} + W)\sigma_{00}^{\mathrm{eff}} = \tfrac{1}{3}\gamma^{(2)}(\sigma_{++} + \sigma_{--}) + (W + \tfrac{1}{2}\gamma_{f\rightarrow e})\sigma_{ff} + S_0^D \ , \tag{12a}$$

$$(\gamma_e + \tfrac{1}{3}\gamma^{(2)})(\sigma_{++} + \sigma_{--}) = \tfrac{2}{3}\gamma^{(2)}\sigma_{00}^{\mathrm{eff}} + \tfrac{2}{3}\gamma_{f\rightarrow e}\sigma_{ff} + 2S_1^D \ , \tag{12b}$$

$$(\gamma_f + W)\sigma_{ff} = W\sigma_{00}^{\mathrm{eff}} \ . \tag{12c}$$

This corresponds to a "half-dressed" picture (as shown in Fig. 5) in which we dress the radiator in the field of the first laser only. S_0^D and S_1^D are simply the dressed-state collisional rates

$$S_0^D = \langle\langle \mathrm{II}\,\mathrm{II}\,|\,\gamma_c\,|\,\mathrm{II}\rangle\rangle\sigma_{\mathrm{II}} = \frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{\Delta_1^2} \times \{\tfrac{2}{3}\gamma_{eg}(\Delta_1) + \tfrac{2}{3}[\Gamma_{eg}^{(2)}(\Delta_1) - \gamma^{(2)}]\} \ , \tag{13a}$$

$$S_1^D = \langle\langle 1\,1\,|\,\gamma_c\,|\,\mathrm{II}\rangle\rangle\sigma_{\mathrm{II}} = \langle\langle -1\,-1\,|\,\gamma_c\,|\,\mathrm{II}\rangle\rangle\sigma_{\mathrm{II}} = \frac{1}{3\hbar^2}\frac{|\langle e||\mu||g\rangle\mathscr{E}_1|^2}{\Delta_1^2} \times \{\tfrac{2}{3}\gamma_{eg}(\Delta_1) - \tfrac{1}{3}[\Gamma_{eg}^{(2)}(\Delta_1) - \gamma^{(2)}]\} \ , \tag{13b}$$

where σ_{II} is assumed undepleted. The probe laser then induces a near-resonant transition from the dressed state $|\mathrm{II}\rangle$ to the bare state $|f\rangle$ with a rate given by W (see Fig. 5).

With this insight one can now return to the calculation of Alber and Cooper in the bare-state representation. One finds that the Raman rate R does not really correspond to an observable at all. Rather, it represents the destructive interference between two possible pathways leading to the creation of the coherence σ_{fe}. In the absence of collisions these two contributions $\sigma_{gg} \rightarrow \sigma_{eg} \rightarrow \sigma_{ee} \rightarrow \sigma_{fe}$ and

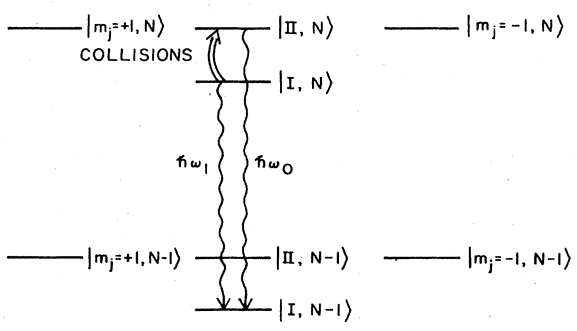

FIG. 4. The dressed-state interpretation of the situation depicted in Fig. 3(a).

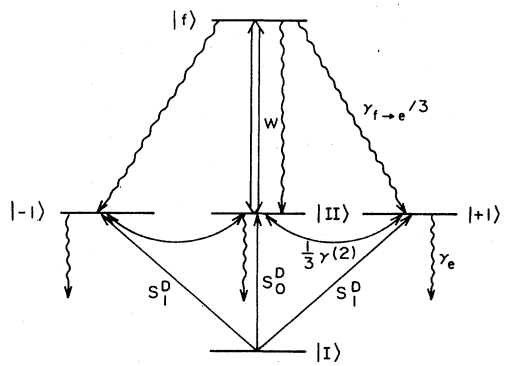

FIG. 5. The half-dressed picture of the rates depicted in Fig. 2.

$\sigma_{gg}\rightarrow\sigma_{eg}\rightarrow\sigma_{fg}\rightarrow\sigma_{fe}$, cancel to order

$$(|\Omega_1|^2/\Delta_1^2)\max\{|\Delta_2/\Delta_1|,\gamma/|\Delta_1|\}$$

and the final-state population is negligibly small. Collisions then are needed to produce a final-state population of the order kept in the calculation, namely of order $(\Omega/\Delta_1)^2$. The physics of the situation is very similar to that present in the pressure-induced extra resonances (PIER) seen in four-wave-mixing experiments.[13]

There are two possible collisional contributions leading to a final-state population. The first, which we call the direct process, involves the absorption of a photon from both laser fields during a single collision. The second process is a stepwise one in which the absorption of the photon from the first laser occurs during a collision resulting in an excited-state population that exists for a time of order γ_e^{-1}. The second photon, since it is near resonant with the $|e\rangle\rightarrow|f\rangle$ transition, can then be absorbed at any point during the excited-state lifetime. The stepwise process is therefore the dominant contribution.

Fortunately this is exactly the situation that one desires. The direct process corresponds to the $|\mathrm{I}\rangle\rightarrow|f\rangle$ transition in the half-dressed picture, while the $|\mathrm{II}\rangle\rightarrow|f\rangle$ rate W represents the stepwise excitation. Since the direct process is negligible for our conditions of small $|\Delta_2|$, the final-state population is directly proportional to the observable in the one-photon redistribution experiment (i.e., the population of the dressed state $|\mathrm{II}\rangle$). Furthermore, this statement remains valid when one turns the polarization of the first laser to arrive at the situation of Fig. 1(b). Of course, the Raman coupling is not present in this second case, due to the opposite polarizations of the two lasers, and the entire population in the bare-$m_j=0$ excited state is probed by the second laser with the same transition rate W. This means that the polarization

$$P=\frac{\sigma_{ff}^{\parallel}-\sigma_{ff}^{\perp}}{\sigma_{ff}^{\parallel}+\sigma_{ff}^{\perp}}\ , \tag{14}$$

which is the ratio of the sum and difference of the final-state populations, is exactly the same as the result of the one-photon experiments, even when the excited- to final-state transition is heavily saturated.

If we choose instead to monitor the fluorescence from the $m_j=0$ excited state to the ground state, the same polarization would be obtained for the configuration shown in Fig. 1. However, if we turn the polarization of the probe laser perpendicular to the quantization axis and continue to monitor the fluorescence from the $m_j=0$ state we would expect the polarization to become intensity dependent as well as pressure dependent. This is because the probe laser now couples the $m_j=\pm1$ states to the final state and therefore the $m_j=0$ state as well (through $\gamma^{(2)}$ and $\gamma_{f\rightarrow e}$). In fact, using the results of Alber and Cooper one can readily find

$$P_{\parallel}=\frac{\sigma_{00}^{\parallel,\parallel}-\frac{R}{W}-\sigma_{00}^{\perp,\parallel}}{\sigma_{00}^{\parallel,\parallel}-\frac{R}{W}+\sigma_{00}^{\perp,\parallel}}$$

$$=P_0\ , \tag{15a}$$

$$P_{\perp}=\frac{\sigma_{00}^{\parallel,\perp}-\frac{R}{W}-\sigma_{00}^{\perp,\perp}}{\sigma_{00}^{\parallel,\perp}-\frac{R}{W}+\sigma_{00}^{\perp,\perp}}$$

$$=P_0\left[1+\frac{\frac{2}{3}[\gamma_f\gamma^{(2)}-\gamma_{f\rightarrow e}(\gamma_e+\gamma^{(2)})]}{\gamma_e\left[\gamma_f+(\gamma_e+\gamma^{(2)})\left[1+\frac{\gamma_f}{W}\right]\right]}\right]\ , \tag{15b}$$

where the first and second superscripts on the population σ_{00} refer, respectively, to the polarization of the first and second laser relative to the quantization axis. P_0 is the polarization of the one-photon experiments and $P_{\parallel}$ and $P_{\perp}$ refer to the polarizations measured from the excited-state fluorescence with the probe laser polarization parallel and perpendicular to the quantization axis.

III. EXPERIMENT

The experimental setup is essentially the same as that used by Alford and co-workers to measure the redistribution of circularly polarized light.[1] An Ar^+ laser is used to pump two standing-wave dye lasers. The excitation laser passes through a Pockels cell to give either vertically or horizontally polarized light and is then weakly focused into a heated oven containing Ba vapor and Ar buffer gas. Its detuning from the ground- ($6s^2\,{}^1S_0$) to excited- ($6s\,6p\,{}^1P_1$) state transition is monitored by means of a reference monochromator. The probe laser, whose polarization is determined by another Pockels cell, is sent counterpropagating into the oven with a beam waist slightly larger than that of the excitation laser. With a Mach-Zehnder étalon, a birefringent tuner, and a thin étalon as tuning elements it runs predominantly in four to five cavity modes spaced approximately 100 MHz apart. Passively stabilized, it is tuned to the excited-to-final ($6s\,8s\,{}^1S_0$) transition.

All signals were observed at right angles to the propagation direction of the two lasers and spectrally resolved using a 0.2-m monochromator. The polarizations measured via the final-state population were determined by monitoring the fluorescence from the final state down to the $6s\,6p\,{}^3P_1$ state at 4601 Å. The fluorescence from the excited state was first sent through a linear polarizer before being resolved from the Rayleigh scattered light by the monochromator. The orientation of this polarizer remains fixed while the laser polarizations are varied using the Pockels cells. A heated sapphire window is placed in the interaction region to reduce the optical depth through which the excited-state fluorescence must travel. Details of this arrangement have been discussed elsewhere.[14] Although the arrangement of the beam sizes used is not ideal for minimizing radiation trapping effects (to do so, the probe beam should be slightly smaller than the pump beam[15]), it does enable us to saturate more of the excited-state population which was initially excited in the spatial wings of the first laser. The density of Ba vapor is kept to levels such that the effects of radiation trapping are negligible.

IV. RESULTS AND DISCUSSION

The above claim of the independence of the polarizations, as measured via the final-state population, with respect to the probe-laser intensity was experimentally verified using circularly polarized light for the case of a single detuning and pressure by Alford *et al.*[1] A more systematic study using linearly polarized light is presented in Fig. 6. It substantiates this claim for a variety of detunings and buffer-gas pressures. In all cases a nonlinear dependence of the total signal on the intensity of the probe laser was observed. For the highest intensities and lowest pressures this dependence went roughly as the intensity to the one-tenth power.

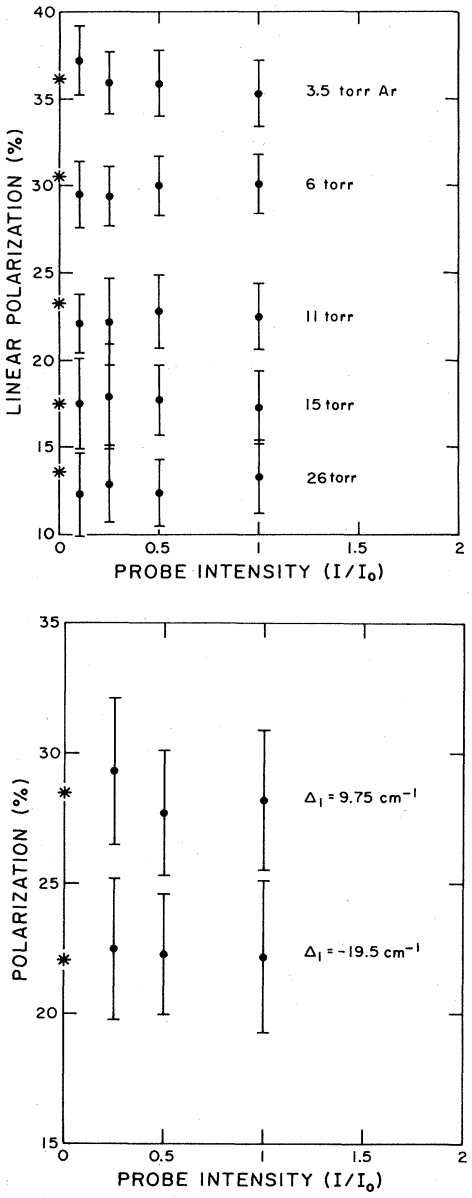

FIG. 6. The linear polarization as measured by the pump-probe technique vs the intensity of the probe laser. *denotes the one-photon measurements of Ref. 18. I_0 is approximately 10 W/cm^2. (a) As a function of Ar pressure. (b) As a function of the detuning Δ_1.

To confirm our understanding of the problem we have also measured the polarization of the excited-state fluorescence in the presence of a saturating probe laser. Our experimental results, shown in Fig. 7, are in good qualitative agreement with Eqs. (15a) and (15b). Of particular interest is the dependence on the intensity of the probe laser. The probe parallel polarizations $P_{||}$, are seen to be roughly equal to the one-photon measurements over an order of magnitude variation in the probe intensity. Equation (15b), however, predicts an intensity dependence for the probe perpendicular case $P_\perp$. The high- and low-intensity limits of this expression are

$$P_\perp \rightarrow P_0 \left[1+\frac{\frac{2}{3}[\gamma_f\gamma^{(2)}-\gamma_{f\rightarrow e}(\gamma_e+\gamma^{(2)})]}{\gamma_e(\gamma_f+\gamma_e+\gamma^{(2)})} \right] \text{ as } W\rightarrow\infty \,, \tag{16a}$$

$$P_\perp \rightarrow P_0 \text{ as } W\rightarrow 0 \,. \tag{16b}$$

Although we are unable to reach high enough intensities such that the polarizations are independent of the probe power, as the probe power is dropped the measurements tend toward the correct (one-photon) limit. The pressure dependence is also of some interest. Again the probe parallel polarizations agree well with the one-photon measurements. As the buffer-gas pressure is decreased, the probe perpendicular polarizations decrease monotonically toward the one-photon measurements. In fact, for pressures lower than those experimentally feasible, the probe perpendicular polarizations should lie below the one-photon measurements as indicated by the following limits:

$$P_\perp \rightarrow P_0 \left[1+\frac{\frac{2}{3}(\gamma_f-\gamma_{f\rightarrow e})}{\gamma_e\left[1+\frac{\gamma_f}{W}\right]} \right] \text{ as } n_p\rightarrow\infty \,, \tag{17a}$$

$$P_\perp \rightarrow P_0 \left[1-\frac{\frac{2}{3}\gamma_{f\rightarrow e}}{\gamma_f+\gamma_e\left[1+\frac{\gamma_f}{W}\right]} \right] \text{ as } n_p\rightarrow 0 \,, \tag{17b}$$

where n_p is the perturber number density.

We should briefly comment on the qualitative effects to which the motion of the radiator might give rise. As the first laser is strongly detuned from the resonance, no particular velocity class of the radiator is excited by the first laser transition. However, because the detuning of the second laser is small compared to the width of the Doppler profile, each separate velocity class in the excited state sees an effective detuning of $(\Delta_2-\mathbf{k}\cdot\mathbf{v})$ in the field-induced transition rate W [see Eq. (6)]. If the effects of velocity-changing collisions are negligible, then the rate equations of Alber and Cooper remain valid for each velocity class and the final- and excited-state populations would simply need to be integrated over the Doppler profile. This would leave the polarization as measured from the final-state fluorescence and $P_{||}$ unaffected since the

dependence on W appears as a common factor in the populations $\sigma_{ff}^{\parallel}$, $\sigma_{ff}^{\perp}$, $\sigma_{00}^{\parallel,\parallel}$, and $\sigma_{00}^{\perp,\parallel}$. The effect of $P_{\perp}$ would be somewhat more complicated.

One would, in fact, expect that the effect of velocity-changing collisions will be negligible for the excited-state populations. Hubeńy and Cooper[16] show that the rate of velocity-changing collisions should be roughly an order of magnitude smaller than the rates associated with the destruction of alignment and orientation. Since we work at pressures such that these latter rates are roughly equal to the radiative decay rate of the excited state, velocity-changing collisions should have a small effect on the excited-state populations. The case for the final-state populations is somewhat different. Using the estimates described below (a mean-squared final-state radius of 170 Å^2 and a radiative lifetime of 6.1 ns) we would expect an average of 1–2 velocity-changing collisions during a final-state lifetime for our pressures. Invoking a strong collision model then allows us to treat the final-state velocity distribution as Maxwellian. Within this model the separate velocity classes of the final-state population are again independent and the same conclusion as that for the case of no velocity-changing collisions is reached regarding the polarizations.

A quantitative analysis of the data is considerably hindered by the lack of knowledge of the atomic parameters relevant to the problem. Nonetheless, we shall be able to draw a few tentative conclusions. We start by determining an absolute value of the ratio γ_f/W for each of the data points in Fig. 7(c) by comparing the saturation

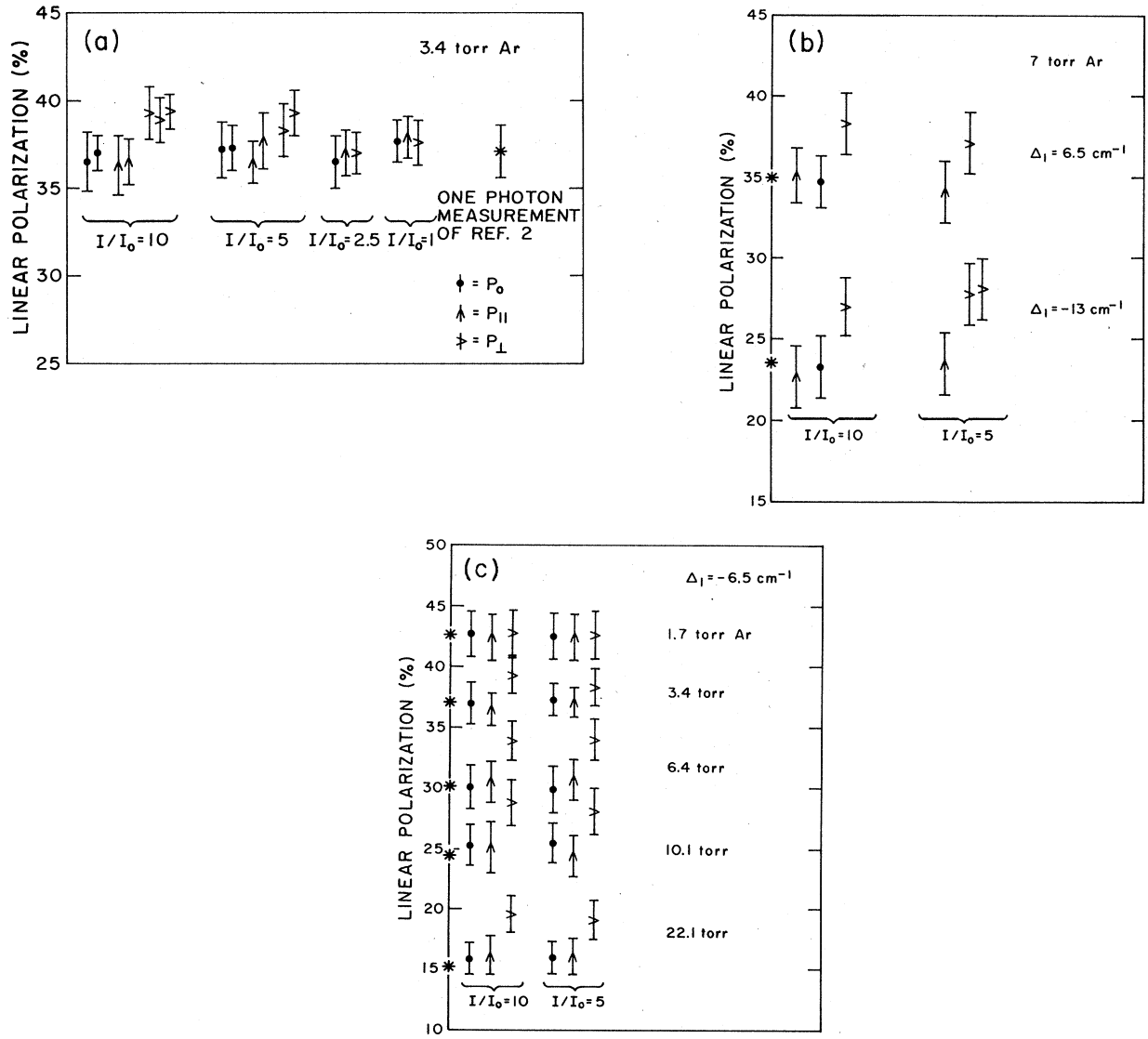

FIG. 7. The linear polarization of the excited-state fluorescence. Here $I_0=1$ W/cm^2. (a) The intensity dependence, $\Delta_1=-6.5$ cm^{-1}, (b) detuning dependence, (c) Ar pressure dependence.

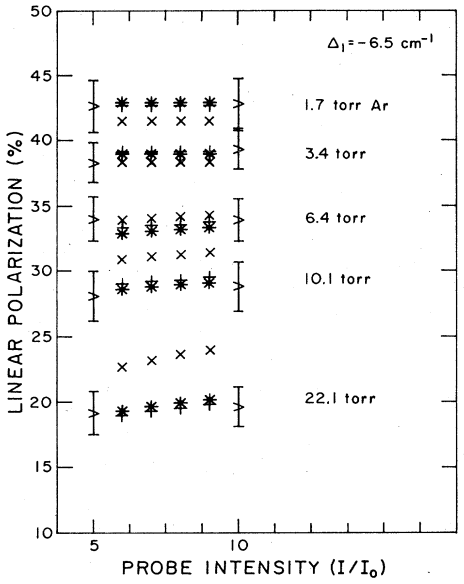

FIG. 8. The predicted variation of $P_\perp$ with probe-laser intensity using the model described in the text: * denotes the overall best fit, + is the best fit with $\tau_f^{\rm rad}$ constrained to 6.1 ns, × is the fit for $\tau_f^{\rm rad}=6.1$ ns and a collisional transfer cross section of 200 Å^2.

behavior of the final- and excited-state fluorescence signals ($I_{||}+2I_\perp$). We thereby ignore any dependence of the induced transition rate W on the motion of the radiator. Within the core of the Gaussian spatial profile of the probe beam, the transition is heavily saturated, and this should be a good approximation. However, a significant portion of our signal undoubtedly comes from atoms probed by the spatial wings of the second laser, as evidenced by our inability to reach the high-intensity limit of Eq. (17a). Rather than try to take this into account, we simply call γ_f/W an effective saturation parameter fit to the data in the manner prescribed. Although this is a fairly crude approximation we feel that the uncertainties in the polarization measurement do not warrant the additional effort entailed in a more exact treatment.

When these values are substituted into Eq. (15b) a set of six equations linear in the parameters γ_f/γ_e and $\gamma_{f\to e}/\gamma_e$ can be derived from the data of Fig. 7(c). The analysis is complicated somewhat by the fact that in reality the barium system is not very well approximated by a three-level atom. The final state lies within thermal energies of several other states and the possibility of collisional transfer from and back to the final state must be considered. Unfortunately, this has the effect of making the set of equations nonlinear and a grid search method was used to find the best least-squares fit to the data. We, in fact, find that the probability for collisional transfer back to the final state is rather small and the best-fit parameters are the following: a radiative lifetime of 13 ns for the excited state, a radiative branching ratio for decay from the final state down to the excited state of 0.18, and a collisional transfer cross section from the final state of approximately 75 Å^2. To the best of our knowledge only a preliminary value[17] of 6.1 ± 0.4 ns is available for the radiative lifetime of the final state. Given the uncertainties involved in our analysis, we hesitate to claim that our results contradict this value. Indeed there is a fair amount of interplay amongst the parameters and if we constrain the final-state radiative lifetime to 6.1 ns we obtain a reasonable fit to the data for a branching ratio of 0.16 and a collisional transfer cross section of 36 Å^2 (see Fig. 8). These values for the collisional transfer cross section are not unreasonable. Using an effective Coulomb approximation, the mean-squared radius for the final state can be estimated to be 170 Å^2 and the best candidate level for collisional transfer is the $6p^2\,{}^1D$ state nearly 2 kT away, so that a cross section somewhat smaller than 170 Å^2 is to be expected. Indeed a collisional transfer cross section of 200 Å^2 is inconsistent with our data (fixing the radiative lifetime of the final state of 6.1 ns, see Fig. 8).

The important result of the experiment is not the determination of these parameters, however. It is a relatively poor means of doing so owing to the rather large uncertainties of the polarization measurements compared to the size of the effect in the probe perpendicular case. Rather what is of primary importance is that the polarization measured by monitoring the total fluorescence from the final state leads to the same results as the one-photon measurements. This statement should be true regardless of the degeneracy of the final state, provided that the monitored signal is proportional to the total final-state population, since it is always the effective population of the excited state which is coupled to the final state by the probe laser. There would seem to be many applications of this technique in the field of collisional redistribution, from allowing one to monitor strongly allowed signals in experiments on metastable states and forbidden transitions, to the minimization of radiation trapping effects in resonance line experiments.

ACKNOWLEDGMENTS

The authors thank Dr. G. Alber, Dr. J. Alford, and Dr. A. Streater for invaluable assistance at various stages in this project. This work was supported by National Science Foundation Grant No. PHY82-00805. One of us (J.C.) also received support from the Atomic and Plasma Radiation Division of the National Bureau of Standards.

*Present address: Department of Physics/Astronomy, CSU Long Beach, 1250 Bellflower Blvd., Long Beach, CA 90840.

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