Observation of effect of stimulated processes on dressed-state collisional kinetics

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We describe our observations of the effect of stimulated and four-wave mixing processes on the kinetics of atomic states dressed by an intense laser field in the presence of collisions. The effects are observed via the forward- and side-scattered light. We suggest that the effects observed by Harter *et al.* are produced by amplification of background laser fluorescence by these processes.

I. INTRODUCTION

The study of collisions in the presence of an intense laser field has received considerable theoretical and experimental attention in recent years.[1,2] One method for studying a prototype of such collisions, an optical collision,[3] is via the near-resonant scattered light spectrum of atoms in an intense laser beam.[4] To understand the observed spectrum in such an experiment, it is most convenient to consider in effect the kinetics of the "dressed"[2] atomic states in the presence of spontaneous emission and collisions.[5] In the very intense radiation fields necessary to observe laser modification of collision dynamics, however, additional strong-field radiative processes can profoundly alter the dressed state kinetics; examples of such processes being the four-wave parametric amplification and "three-photon" gain in a strongly driven atomic system that have been studied qualitatively in sodium by Harter *et al.*[6,7]

Here we report on more detailed observations of these four-wave parametric processes and three-photon gain (as discussed below) in the Sr-λ-460-nm transition. We have studied their dependence on laser power, and buffer gas pressure, and also how they affect the relative intensities of the spontaneous (side-direction) fluorescence and, therefore, the measurement of the dressed state collision rates.

II. THEORETICAL BACKGROUND

The formal theory of dressed state kinetics, including the effect of collisions, has been developed for a two-state system by Reynaud and Cohen-Tannoudji[8] and independently by Burnett *et al.*[9] who also treat the problem including the effects of m-level degeneracy. In the binary collision approximation and secular approximation the evolution of the dressed atom density matrix reduces to a set of rate equations in the dressed frame. The dressed frame kinetics are determined by the transition rates shown in Fig. 1 for the case of a $J=0$ to $J=1$ transition (e.g., the Sr resonance transition). It is these rates that one would like to measure unambiguously.

The spontaneous radiative processes, shown as wavy lines, are discussed theoretically by Cooper *et al.*[10] and have been observed by Kleiber *et al.*[11] The collision rates, shown as double lines, have been calculated in the dressed frame using an S-matrix approach by Light and Szöke,[12] and we have used these in a full calculation of the time-dependent spectrum for this system that is valid when the stimulated effects we discuss here are absent.[13] Extra transitions, due to stimulated radiative ef-

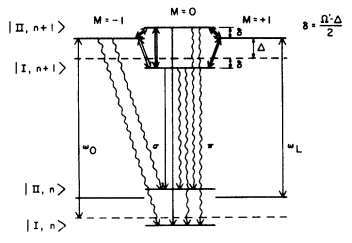

FIG. 1. Dressed level diagram showing the various radiative decay rates (wavy arrows), collisional transfer processes (double arrows), and stimulated radiative rates (straight arrows) between the dressed states.

fects,[14] shown as straight arrows in Fig. 1, may also be important. The theoretical work of Boyd *et al.*[6] predicts the possibility of four-wave parametric amplification: The nonlinear polarization of the medium in the presence of a strong pump wave at frequency ω_L and weak probe waves at frequencies ω_3 and ω_4 (with $\omega_3+\omega_4=2\omega_L$) can induce three-photon gain at ω_3 and parametrically amplify waves at ω_4. Experimental work on the D_2 line of sodium by the same group (Harter *et al.*[7]) has qualitatively verified these effects. They have observed gain in the sidebands symmetrically displaced from the laser frequency by the generalized Rabi frequency, Ω'. At higher densities and laser intensities they also observe self-focusing of the incident laser. In contrast to their experiments, in which the source of the weak radiation being amplified was unknown, we have identified the source to be weak background laser fluorescence.

III. EXPERIMENTAL SETUP

The experimental setup is detailed in Ref. 11. Here, however, the N_2 pumped dye laser has been replaced with a commercial excimer pumped dye laser system (Lambda Physik Model No. EMG102 operated with XeCl and a He buffer gas, and Model No. FL2000 operated with Coumarin 460 laser dye). The dye laser output was typically 10 mJ in a 6-nS pulse, and with a spectral FWHM of 0.03 nm. The laser was tuned to 461 nm, 17 cm^{-1} to the red of the Sr resonance line. As in Ref. 11 the beam was polarized with a glan prism polarizer and focused into the oven to a focal spot of ~ 150 μm diam. The estimated intensity in this spot is ~ 1 GW/cm^2. The laser intensity was changed by inserting neutral density filters before the oven entrance. The oven was operated at 600 °C and contained strontium vapor at 2×10^{14} cm^{-3} and argon buffer gas at 10 Torr.

The forward-scattered spectrum was measured by imaging the oven center onto the slit of a spectrometer in the forward direction. To avoid saturating the detector, a small block ($\frac{1}{4}$ in. diam) is placed on axis after the oven to block the direct laser beam. Polarizers after the oven are used to observe only the parallel scattered spectrum.

IV. EXPERIMENTAL RESULTS

A. Forward direction

Typical spectra are shown in Fig. 2, as a function of increasing laser intensity. A "turn on"

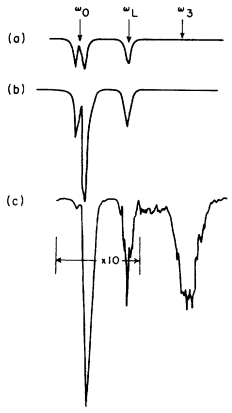

FIG. 2. Forward-scattering spectrum as a function of incident laser intensity. The spectra (a)—(c) show the effect of increasing the laser intensity by factors of 2.

behavior for both the three-photon peak and the Rayleigh peak (laser frequency) is evident. In addition, there appears to be some stimulated behavior in the red wing of the fluorescence (fourth parametric wave), while the blue wing of the fluorescence remains approximately linear. These observations are detailed in Fig. 3, showing the peak intensity of the scattered features versus incident laser intensity. Note that the turn on of the Rayleigh (laser) peak occurs at slightly higher incident intensities than the three-photon or red-wing fluorescence turn on.

We have also studied this nonlinear behavior as a function of buffer gas pressure. In all cases the incident intensity required to turn on the stimulat-

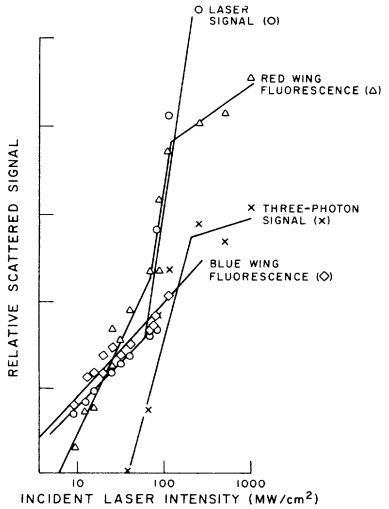

FIG. 3. Relative intensity of the forward-scattered features as a function of incident laser intensity.

ed radiative processes increased with increasing argon pressure. These observations are consistent with requiring higher gain for the stimulated radiative processes with increasing buffer gas pressure to overcome the collisional processes that remove population from the $m_J=0$ doublet that provides the gain.

(Although we have not made detailed studies of the stimulated behavior versus strontium density, we note that at a lower Sr density of $\sim 1\times 10^{13}$ cm^{-3} we were unable to observe any nonlinear radiative effects.)

We believe the mechanism responsible for these effects is stimulated three-photon scattering coupled with phase matched parametric amplification of the "fourth wave" which appears to the red of the unshifted atomic resonance.[6] To test the origin of the weak waves which are amplified, we put the laser through a grating spectrometer before focusing it into the oven. This spectrometer decreased the signal in the laser wings (at $\pm\Delta$ from the laser peak) from $\sim 5\times 10^{-4}$ to $\sim 5\times 10^{-5}$ of the peak laser signal. Under these conditions (peak intensities at the oven center ~ 100 MW/cm^2), the stimulated emissions at both the three-photon peak and the fourth wave peak are no longer apparent. We are uncertain whether the stimulated emission has completely disappeared or is simply too small to observe with our present setup. In any case, the observation of these stimulated processes can be correlated with the broadband fluorescence from the dye laser.

The Rayleigh peak (laser peak) remains linear up to laser intensities ~ 60 MW/cm^2, then it also appears to turn on. This is independent of the presence of the laser wings. We believe this is a geometrical effect associated with the onset of self-defocusing. At low laser intensities the beam travels through the oven without self-defocusing and strikes the beam block after the oven. The linear signal observed under these conditions is the small angle Rayleigh scattering which gets around the block. As the laser intensity is increased, self-defocusing occurs (for the laser turned to the *blue* of resonance we expect self-focusing; to the *red,* self-defocusing), which blows up the laser beam spatially, around the beam block and gives a nonlinear signal at the laser frequency. To verify this we imaged the oven center on a photographic plate (with the beam block present) in the forward direction. The beam is observed to blow up spatially at about the same intensity that the laser peak turns on (~ 60 MW/cm^2). Thus the nonlinear behavior of the laser peak correlates well with the onset of self-defocusing.

B. Side direction

We observed large qualitative differences in the side-direction scattered spectrum as a function of the stimulated emission in the forward direction. Figure 4 shows two spectra taken in the side direction under similar conditions: (a) with the grating spectrometer in place to remove the laser wings, and (b) with the grating replaced by a mirror. Note the Rayleigh peak is, as expected, roughly the same height in both traces. In (a) the spontaneous three-photon peak is quite well pronounced but in (b) it has almost disappeared. In addition, the parallel fluorescence signal grows substantially between (a) and (b). We attribute these effects to the stimulated scattering processes discussed above. When the three-photon peak goes stimulated in the forward direction, the side-direction spontaneous emission drops accordingly. Similarly when the three-photon scattering process turns on, stimulated radiative excitation of the $m=0$ substate of the excited level gives an anomalously large parallel fluorescence signal.

V. CONCLUSIONS

Finally we emphasize that in any experiment making *quantitative* measurements of dressed state collision rates via the intensities of side-scattered light, one must be careful to eliminate the collective effect of the stimulated processes discussed above. At the high laser intensities necessary to

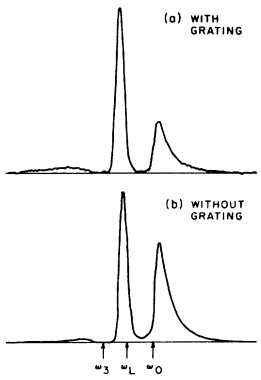

FIG. 4. Side-scattered spectrum (a) with and (b) without the grating arrangement to remove the laser background fluorescence.

observe nonlinear effects in collision dynamics, this may be quite difficult. Note, in particular, in the present case, that by saturating the collision $\Omega/\Delta \gtrsim 1$ (cf. Ref. 12), we also saturate the transition. This implies that the effects we describe here will be present (at sufficient densities) at the interesting field strengths unless care is taken to eliminate the background fluorescence of the laser.

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