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Enhancement of four-wave mixing signals due to velocity-changing collisions

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We present experimental data showing the first observation of large enhancement of four-wave mixing signals due to velocity-changing collisions. The results are explained with a simple model which describes the population distributions in the presence of velocity-changing collisions and optical pumping, allowing relative velocity-changing collision rates to be inferred from the data.

The effect of velocity-changing collisions (VCC) by ground-state perturbers on the velocity distribution of atoms interacting with a monochromatic laser beam has been demonstrated by numerous experiments using saturation spectroscopy.[1,2] The purpose of this Communication is to describe a new manifestation of VCC which gives rise to enhancement of four-wave mixing signals. The enhancement is brought about by a collisional redistribution of the velocities which offsets the effects of velocity hole burning due to hyperfine optical pumping. We show how VCC rates may be inferred from the data using a simple model for the equilibrium population distribution.

The principal distinction between four-wave mixing and saturation spectroscopy using cw lasers in a multilevel system is that four-wave mixing signals increase with a pump induced increase in the ground-state number density while saturation spectroscopy signals increase with a pump induced decrease in the ground-state number density. Hence, cw four-wave mixing experiments tend to emphasize transitions which are not optically pumped, while in saturation spectroscopy the largest signals are observed on optically pumped transitions. The above description is true unless the pump beam laser intensity (I) is much less than I'_{sat}, where the saturation intensity I'_{sat} is calculated to account for optical pumping, spatial diffusion, and wall relaxation.[3,4] Typically, I'_{sat} is much less than the usual I_{sat}. In the first cw degenerate four-wave mixing (DFWM) experiments on the D_2 line in atomic sodium, the only strong signals observed using a cw dye laser were obtained on the $3s\,^2S_{1/2}(F=1)-3p\,^2P_{3/2}(F=3)$ transition and on the $3s\,^2S_{1/2}(F=1)-3p\,^2P_{3/2}(F=0)$ transition.[5] Of the six dipole allowed transitions on the D_2 line in atomic sodium, these two transitions are the only transitions that are not optically pumped. The remaining four transitions are indeed observed but only at intensities comparable to I'_{sat}. On these transitions at intensities larger than I'_{sat}, atoms in the $F=2$ ground state are quickly pumped into the $F=1$ ground state while atoms in the $F=1$ ground state are quickly pumped into the $F=2$ ground state. This paper demonstrates that at pump intensities comparable to I_{sat}, the normally very weak signal on the optically pumped transition given by $3s\,^2S_{1/2}(F=1)-3p\,^2P_{3/2}(F=2)$ can be considerably enhanced in the presence of VCC by ground-state perturbers. The physical origin of this enhancement is easily understood by considering the effect of VCC on velocity hole burning caused by the pump beams and optical pumping. In the absence of optical pumping, a pump beam generates a hole in the velocity distribution of the ground state by exciting atoms to the upper level in a very narrow velocity distribution determined by the natural linewidth of the transition. The depth of the hole in the ground-state velocity distribution is equal to the height of the spike in the upper-state velocity distribution and is determined by the strength of the laser-atom coupling. The maximum depth of the velocity hole would be equal to one-half the equilibrium ground-state population in the absence of the laser, corresponding to complete saturation of the transition. However, in an inverted V-type three-level system shown in Fig. 1, where optical pumping is present, it is possible to increase the hole depth in the ground-state population distribution to the point where the depletion of the population of that velocity group is

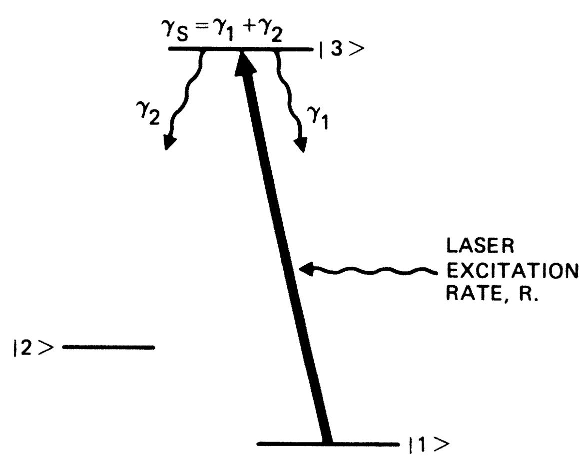

FIG. 1. Inverted V-type three-level system used to describe the effects of VCC on four-wave mixing signals generated on optically pumped transitions.

almost complete. At the same time, the height of the spike in the upper level is also reduced, being determined by the final equilibrium population distribution and the strength of the laser-atom coupling. However, atoms which have not been optically pumped out of level 1 because they are Doppler shifted out of resonance can be shifted into resonance if they experience an appropriate velocity-changing collision. This has the effect of increasing the equilibrium population density of level 1 in the specific velocity group interacting with the laser. As shown in the experiments below, this effect can be very pronounced. A simple analytical solution to the population rate equations has been obtained for this three-level system shown in Fig. 1 where a hard-sphere collision model has been assumed. In this model, the effects of the standing wave in the experiment have been ignored and strong collisions have been assumed, i.e., the velocity distribution is thermal after a collision. Nevertheless, we show that the four-wave mixing signal is an excellent indicator for the resonant population distribution and the VCC rate for various ground-state perturber gases can be estimated from the data.

The experimental approach to demonstrate this effect is based on DFWM in atomic sodium vapor. The geometry for DFWM is discussed in the literature.[6] The cw dye laser was tuned to the 589 nm resonance of the D_2 line. In Doppler-broadened media this interaction is Doppler free since only the zero-velocity group (whose width is determined by the natural linewidth) can interact simultaneously with all three beams.[7] Figure 2 (dotted line) shows the DFWM signal produced as the laser is tuned through the resonances of the D_2 line. As indicated above, while there are six dipole allowed transitions, only two are usually observed at convenient power levels because of optical pumping.[8] However, as a small amount of buffer gas is added, there is a pronounced change in the spectral behavior of DFWM. Figure 2 (solid line) shows that transitions A and B, which are not optically pumped, are reduced due to fine-structure changing collisions,[9] while transitions $3s\,^2S_{1/2}(F=1)$–$3p\,^2P_{3/2}(F=1)$ and $3s\,^2S_{1/2}(F=1)$–$3p\,^2P_{3/2}(F=2)$ (denoted by C in the figure and unresolved in this display) are enhanced. The points in Fig. 3 show the observed dependence of the $3s\,^2S_{1/2}(F=1)$–$3p\,^2P_{3/2}(F=2)$ DFWM signal level as a function of buffer gas pressure for helium. Similar data were also obtained in argon and xenon. As anticipated by our arguments given above, the data show that DFWM signals that are normally weak due to optical pumping are considerably enhanced by the presence of only a few hundred millitorr of buffer gases.

In the absence of pump absorption, DFWM signals vary as the square of the ground-state population density. Hence we expect that a simple rate equation

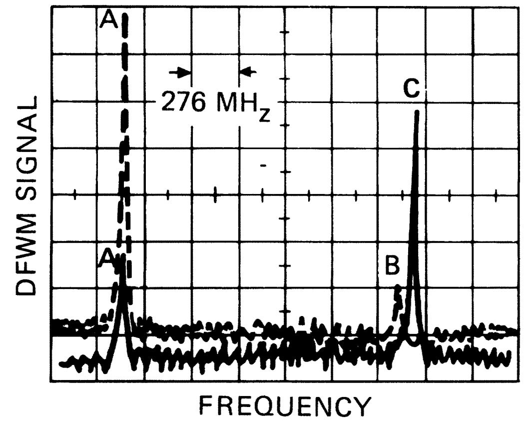

FIG. 2. Spectral structure of the DFWM signal as the cw dye laser is tuned across the D_2 line. The dashed line shows strong signals observed on the $3s\,^2S_{1/2}(F=2)$–$3p\,^2P_{3/2}(F=3)$ transition (A) and the $3s\,^2S_{1/2}(F=1)$–$3p\,^2P_{3/2}(F=0)$ transition (B). The solid line shows the effect of adding 200 mTorr of helium buffer gas. We see that the signal denoted by C in the vicinity of the three dipole allowed transitions originating out of the $F=1$ ground state is considerably enhanced. A high-resolution display of this spectral region shows that, in fact, the signal appearing is due to collisional enhancement of the $3s\,^2S_{1/2}(F=1)$–$3p\,^2P_{3/2}(F=1)$ transition and the $3s\,^2S_{1/2}(F=1)$–$3p\,^2P_{3/2}(F=2)$ transition.

model predicting the equilibrium population densities would explain the DFWM signal level dependence on buffer gas pressure.[10] The rate equations for levels 1 and 3 are shown below:

$$\dot{n}_1(v) = -D(n_1 - n_1^0) - Rn_1 + Rn_3 + \gamma_1 n_3 + \Gamma_{\mathrm{VCC}}[N_1 f(v) - n_1] \ ,$$

$$\dot{n}_3(v) = -Dn_3 - Rn_3 + Rn_1 - \gamma_s n_3 + \Gamma_{\mathrm{VCC}}[N_3 f(v) - n_3] \ .$$

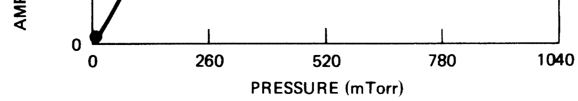

FIG. 3. Comparison between theory ($\Gamma^0_{\mathrm{VCC}} = 1.5 \times 10^7$ sec^{-1} Torr^{-1}) and experiment showing the dependence of the DFWM signal on buffer gas pressure. Comparison is made with the theory by assuming that the DFWM signal is linearly proportional to the square of the calculated population density at $v=0$.

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The effects of velocity-changing collisions are accounted for by assuming the distribution to be thermal after a collision. Such strong collisions are described by a term like $\Gamma_{VCC}[N_i f(v) - n_i(v)]$, where $N_i = \int n_i(v)\, dv$, $f(v)$ is a Maxwellian velocity distribution, and Γ_{VCC} is the pressure dependent velocity-changing collision rate. For simplicity, Γ_{VCC} has been assumed to be the same for both the upper and lower levels of the transition. State-changing collision effects in this pressure range are negligible compared to radiative processes for the transitions under investigation and are not included in this model. In these rate equations, D is the diffusion rate ($\sim p^{-1}$ where p is the buffer gas pressure), γ_1 is the decay rate of level 3 back to level 1, γ_s is the spontaneous emission rate for level 3, n_1^0 is the unperturbed velocity distribution [given by $N_1^0 f(v)$], and R is the velocity dependent laser excitation rate given by $R_0/[1+(\omega-\omega_0-kv)^2/\gamma^2]$, where ω is the laser frequency and kv is the Doppler shift. R_0 is the resonance excitation rate given by $\gamma_s(I/I_{sat})$. These equations may be solved to yield the modified velocity distribution for the ground-state population by assuming only that the natural linewidth and $|\omega-\omega_0|$ are small compared to the Doppler linewidth in order to carry out the velocity integrals analytically:

$$n_1(v) = \frac{N_1^0 f(v)}{1+(R_0/R_1)/(\tau R_0+1)^{1/2}}\left[1+\frac{R_0/R_2}{(\tau R_0+1)^{1/2}}\right.$$
$$\left.\times\left[1-\left(\frac{\gamma_s'-\gamma_1}{D'}\right)\left(\frac{1}{1+(R_0/R_2)/(\tau R_0+1)^{1/2}}\right)\left(\frac{R_0/\gamma_s'}{\tau R_0+(\omega-\omega_0-kv)^2/\gamma^2}\right)\right]\right] ,$$

where

$$R_0=\gamma_s(I_0/I_{sat}),\quad \tau=(\gamma_s'-\gamma_1+D')/(D'\gamma_s') ,$$
$$R_1=(\gamma_s'/F)(D'/\Gamma_{VCC}'),\quad R_2=(\gamma_s'/F)(\gamma_s'-\Gamma_{VCC})/\Gamma_{VCC} ,$$
$$\Gamma_{VCC}'=\Gamma_{VCC}\frac{[\gamma_s'(\gamma_s'-\gamma_1-\Gamma_{VCC})-(\gamma_1-D')(D'-\Gamma_{VCC})]}{(\gamma_s'-\Gamma_{VCC})(D'-\Gamma_{VCC})} ,$$
$$\gamma_s'=\gamma_s+\Gamma_{VCC}+D,\quad D'=D+\Gamma_{VCC} ,$$

and F is proportional to the ratio of the natural linewidth to the Doppler linewidth and is given by $(\pi^{1/2}/2)(\gamma/ku_0)\exp[-(\omega-\omega_0)^2/k_BT]$.

The calculated behavior of this population dependence on Γ_{VCC} for various buffer gas pressures is compared with experiment by plotting $n_1^2(v=0)$ as a function of buffer gas pressure p. Values for the diffusion rate D were taken from the literature.[11] The calculation was multiplied by a constant to enable direct comparison with the DFWM signal. Hence the only adjustable physical parameter was the collison rate Γ_{VCC}^0, where $\Gamma_{VCC}=\Gamma_{VCC}^0 p$. The results from helium are shown in Fig. 3 (solid lines). Similar data and comparison with theory were obtained with argon and xenon. The values used for Γ_{VCC}^0 were 1.5×10^7, 1×10^7, and 0.9×10^7 for helium, argon, and xenon, respectively, in units of $\sec^{-1}\text{Torr}^{-1}$.[12] The value for helium is in surprisingly good agreement with that by Liao and colleagues,[2,13] considering our use of the strong collision model. To the best knowledge of the authors the values for argon and xenon are presented here for the first time. The effect of decreasing the value for Γ_{VCC}^0 is to increase the value of the pressure for maximum signal enhancement. The fit of the model to the data made a 20% change in Γ_{VCC}^0 easily observable. This model is a rate equation approximation and we have not included the effects of VCC on the third order optically induced coherence in degenerate four-wave mixing. In this case, Lam and Berman[14] have shown theoretically that in contrast to our intrinsically intensity dependent collisional enhancement, a small intensity independent collisional enhancement can be observed assuming strong collisions and no optical pumping or diffusion only if Γ_{VCC} for the lower level greatly exceeds Γ_{VCC} for the upper level. If the collision rates are assumed to be equal, as they are in our model, then they predict a small collisionally induced decrease in the signals due to a reduction in the degree of spatial modulation.[14] In fact, experimentally, we do observe a slight collision induced signal reduction at very low pump intensities.

In conclusion, we have demonstrated a new form of collisional enhancement of four-wave mixing signals. This effect has been demonstrated to be a useful signature of velocity-changing collisions and is a totally different approach to the study of VCC from the usual saturation spectroscopy approach.

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[1]C. Brechignac, R. Vetter, and P. R. Berman, Phys. Rev. A 17, 1609 (1978).

[2]P. F. Liao, J. E. Bjorkholm, and P. R. Berman, Phys. Rev. A 21, 1927 (1980).

[3]M. S. Feld, M. M. Burns, T. U. Kuhl, P. G. Pappas, and D. E. Murnick, Opt. Lett. 5, 79 (1980).

[4]P. G. Pappas, M. M. Burns, D. D. Hinshelwook, M. S. Feld, and D. E. Murnick, Phys. Rev. A 21, 1955 (1980).

[5]P. F. Liao, D. M. Bloom, and N. P. Economou, Appl. Phys. Lett. 32, 813 (1978).

[6]R. L. Abrams, J. F. Lam, R. C. Lind, D. G. Steel, and P. F. Liao, in *Phase Conjugation,* edited by Robert Fisher (Academic, New York, 1983).

[7]M. Ducloy and D. Bloch, J. Phys. (Paris) 42, 711 (1981).

[8]As the unfocused pump power is reduced to the 100-μW range, we have no problem observing all six of the dipole allowed transitions and most of the allowed crossovers.

[9]In the vicinity of the D_2 line on sodium, laser excitation of a nonoptically pumped transition [e.g., $3s\,^2S_{1/2}(F=2)-3p\,^2P_{3/2}(F=3)$] can still result in a redistribution of the ground-state equilibrium population density due to state-changing collisions involving the upper state. For example, the fine-structure changing collision $3p\,^2P_{3/2} \rightarrow 3p\,^2P_{1/2}$ enables a net transfer of population from the $3s\,^2S_{1/2}(F=2)$ state to the $3s\,^2S_{1/2}(F=1)$ state.

[10]Similar studies have been performed to examine the efficiency of optically pumped nuclear polarization. P. G. Pappas, R. A. Forber, W. W. Quiver, Jr., R. R. Dasari, M. S. Feld, and D. E. Murnick, Phys. Rev. Lett. 47, 236 (1981).

[11]L. C. Balling, in *Advances in Quantum Electronics,* edited by D. W. Goodwin (Academic, New York, 1975), Vol. 3, p. 1. The diffusion rate is approximated by $D^1 = D_0(2.4/a)^2/p$ where D_0 is tabulated and varies as the square root of the mass ratios. Application of the diffusion approximation places several requirements on the experimental configuration, but most important is the requirement that the beam size be much smaller than the cell size.

[12]The relative accuracy for these numbers seems limited only by the relative accuracy of the diffusion rates given in Ref. 10.

[13]P. R. Berman, P. F. Liao, and J. E. Bjorkholm, Phys. Rev. A 20, 2389 (1979).

[14]J. F. Lam and P. R. Berman (unpublished).