PHYSICAL REVIEW A VOLUME 24, NUMBER 3 SEPTEMBER 1981

Collisionally triggered generation of a coherent signal*

M. Dagenais
Division of Applied Sciences, Harvard University, Cambridge, Massachusetts 02138
and Advance Technology Laboratory, GTE Laboratories, 40 Sylvan Road, Waltham, Massachusetts 02254†
(Received 8 April 1981)

We demonstrate the generation of a coherent signal by an incoherent process. In a first step, population is transferred from the ground state to two excited states by a collisional redistribution process. Then, in a second step, a coherent Raman signal is generated from these two populated excited states. At high pressure, it is predicted that a Raman-type resonance between two equally populated levels should be observed. Information about inelastic-collision cross sections and dephasing coefficients in the excited states is obtained.

I. INTRODUCTION

Usually collisions are thought of as a way of destroying coherence. But recently, by introducing phenomenologically the effect of collisions in the formalism of the third-order susceptibility tensor, it was predicted that collisions could be used to generate coherent Raman signals.[1-3] Such signals would have their origin in the disappearance of the destructive interference between two or more coherent pathways in the presence of collisions. A more intuitive way for understanding this effect would be to realize that at the same time as collisions could supply or take away the extra energy required to access a particular level, as is the case for collisional redistribution,[4,5] a coherence between excited states could be induced by two lasers in the presence of collisions. A slightly different but equivalent way of looking at this problem was recently given by Grynberg[6] who described the collisions as exciting the system in a linear superposition of the eigenstates of the dressed atom. Even when the lasers are tuned away from any one-photon resonance, coherent resonant enhancements are possible and information about level widths of excited states are accessible. Such a phenomenon was very recently observed.[7,8] There, a coherence between excited states in sodium was generated in the presence of collisions by tuning the difference of frequencies of two lasers in correspondence to the separation of the two $3P$ excited states of sodium but none of the lasers were tuned on any one-photon resonance. This gave rise to a Raman type of resonance. Since coherence between two states only exists if both states are populated,[9] it means that in this experiment there was also a real transfer of population from the ground to the two excited states. In the first report of this experiment,[7] the close connection between collisional redistribution and this new pressure-induced coherence effect in the excited states was pointed out for the first time. Shortly after, Bogdan *et al.*[10] also observed a Rayleigh type of resonance in the presence of collisions by doing a degenerate frequency four-wave mixing experiment in Na.

We would like to describe here a new version of these types of experiments which is now based on a two-step process. Contrary to the previously reported experiments,[7,8,10] the coherence in the excited states is not generated at the same time as the population is transferred in these states, but is generated subsequently. This means that the magnitude of the signal goes as the fifth power of the intensity rather than the third power. In our experiment, photons are absorbed off resonance and two nearly excited states in sodium are populated by a collisional redistribution process.[4,5] Then, as the population is increased in these two excited states and in the presence of two lasers of frequencies ω_1 and ω_2, a coherent Stokes-Raman scattering signal (CSRS) of frequency $\omega_{out} = 2\omega_1 - \omega_2$ is generated in the phase-matching direction and in this sense is triggered by collisions. This signal does not exist in the absence of collisions. As the pressure of the buffer gas is increased even more, inelastic collisions start equalizing the population between the two excited states and quantum-mechanical interference between two coherent pathways destroys the coherent signal. At very high pressure, it is predicted that the generated signal should not go anymore as the square of the difference of population between the two nearby excited states but, very peculiarly, as the square of the sum of the two populations. This particular prediction was not checked in our experiment because of a poor signal-to-noise ratio at very high buffer gas pressure. By doing a CSRS type of experiment from an excited state, we demonstrate that one can obtain useful information about dephasing times and inelastic-collision cross sections in the excited states. These measurements can also be extended for obtaining information about interatomic Na-buffer gas poten-

tial without having to deal, for instance, with the usual problems of light trapping faced in the collisional redistribution experiments.[4,5]

II. EXPERIMENTAL SETUP

A 1-MW nitrogen laser with a repetition rate of ten pulses per second is used to pump two dye lasers. One of the dye lasers is used to generate two independently tunable output frequencies (ω_1 and ω_2) separated by about the doublet spacing of the $3P$ state in Na (17 cm^{-1}). This laser is responsible for generating the resonant Raman signal once the $3P$ state is populated. The other laser, called the pump laser, is used only to populate the doublet by a collisionally assisted optical transition and is tuned 24 cm^{-1} above it. This laser is mostly responsible for the transfer of population from the ground state to the $3P$ state because lasers 1 and 2 are tuned about 600 cm^{-1} above the one-photon resonant transition to the doublet. Figure 1 shows the tuning of the lasers and also the interference between two coherent pathways. Lasers 1 and 2 are linearly polarized perpendicular to each other for polarization discrimination in the generated signal. The pump laser is in a different plane from lasers 1 and 2 and the generated signal. The dye laser power is on the order of 1 KW and has a linewidth of about $\frac{1}{4}$ cm^{-1}. The three lasers cross at a small angle (a few milliradians) in the middle of a sodium oven operating at a temperature of 550 K and producing a sodium vapor pressure of 5 mTorr. The generated signal in the phase matching direction is detected with a photomultiplier tube after spatial filtering, polarization discrimination, and frequency filtering. The signal is processed by a boxcar integrator and the dc output is directly proportional to the intensity of the signal generated.

III. THEORY

A model calculation, within the formalism of the density matrix,[11,12] for the intensity of the coherent signal generated versus the buffer gas pressure p will now be given. This treatment will only be valid within the region of validity of the impact approximation for lasers 1 and 2. The following set of equations describes the time evolution of each density matrix element:

$$\frac{\partial\rho_{aa}}{\partial t}=\frac{-i}{\hbar}[V,\rho]_{aa}+\frac{K}{(\omega_{\text{pump}}-\omega_{ag})^2}pI_{\text{pump}}(t)+bp(\rho_{bb}-2\rho_{aa})-\Gamma_{aa}\rho_{aa}+\Gamma_{ca}\rho_{cc}\,, \tag{1a}$$

$$\frac{\partial\rho_{bb}}{\partial t}=\frac{-i}{\hbar}[V,\rho]_{bb}+\frac{2K}{(\omega_{\text{pump}}-\omega_{bg})^2}pI_{\text{pump}}(t)+bp(2\rho_{aa}-\rho_{bb})-\Gamma_{bb}\rho_{bb}+\Gamma_{cb}\rho_{cc}\,, \tag{1b}$$

$$\frac{\partial\rho_{cc}}{\partial t}=\frac{-i}{\hbar}[V,\rho]_{cc}-(\Gamma_{ca}+\Gamma_{cb})\rho_{cc}\,, \tag{1c}$$

$$\frac{\partial\rho_{ab}}{\partial t}=\frac{-i}{\hbar}[H,\rho]_{ab}-\gamma_{ab}\rho_{ab}\,, \tag{1d}$$

$$\frac{\partial\rho_{ac}}{\partial t}=\frac{-i}{\hbar}[H,\rho]_{ac}-\gamma_{ac}\rho_{ac}\,, \tag{1e}$$

$$\frac{\partial\rho_{bc}}{\partial t}=\frac{-i}{\hbar}[H,\rho]_{bc}-\gamma_{bc}\rho_{bc}\,. \tag{1f}$$

$|g\rangle$ refers to the ground state, $|a\rangle$ and $|b\rangle$ to the two $3P$ states, and $|c\rangle$ to the $4D$ state of Na. H is the total Hamiltonian of the system which includes the unperturbed Hamiltonian H_0 of the atomic system and the dipolar interaction $V=-\vec{\mu}\cdot\vec{E}$, where μ is the transition dipole moment and $\vec{E}$ the total applied electric field. The second term on the right-hand side of Eqs. (1a) and (1b) describes the collisionally assisted transfer of population from the ground state to the two P states taking into account the statistical weight of each level. The third term on the right-hand side of Eqs. (1a) and (1b) describes the inelastic transfer of population[13] between the two P states when an excited sodium atom collides with a rare-gas atom in its ground state. bp is the number of inelastic collisions per excited sodium atom per second and is equal to NQv_r where N is the density of the inert gas atoms, Q is the total collisions cross section, $v_r=(8k_BT/\pi\mu_m)^{1/2}$ is the relative He-Na velocity, and μ_m is the He-Na reduced mass. Γ_{aa} and Γ_{bb} are the spontaneous emission rates from levels $|a\rangle$ and $|b\rangle$, respectively. γ_{ab}, γ_{ac}, and γ_{bc} are the transverse damping terms. Using the usual perturbative approach to third order, and if we assume that the population in levels $|a\rangle$ and $|b\rangle$ vary much more slowly than the inverse of the detuning of our lasers or their own reciprocal linewidths, we could solve these equations and obtain an expression for the third-order susceptibility tensor $\chi^{(3)}$:

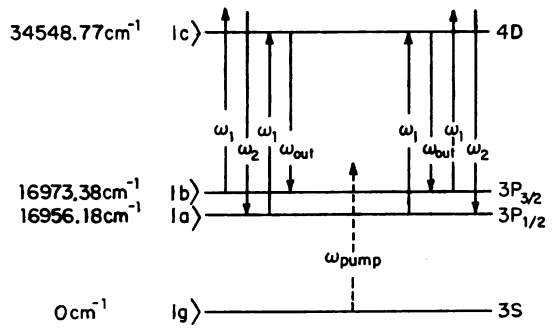

FIG. 1. Energy level diagram of sodium showing the tuning of lasers 1 and 2 and the quantum interference between two coherent pathways. The pump laser is detuned by 24 cm^{-1} from the doublet center.

$$\chi^{(3)}(\omega_{out}, \omega_1, \omega_1, -\omega_2) \propto \frac{1}{(\omega_{ab} - \omega_1 + \omega_2 - i\gamma_{ab})(\omega_{cb} - \omega_{out} - i\gamma_{cb})} \times \left(\frac{[-\rho_{aa}^{(0)}(\omega_{cb} - \omega_1) + \frac{1}{2}\rho_{bb}^{(0)}(\omega_{ca} - \omega_2)] + i(\rho_{aa}^{(0)}\gamma_{cb} + \frac{1}{2}\rho_{bb}^{(0)}\gamma_{ca})}{(\omega_{ca} - \omega_2 + i\gamma_{ca})(\omega_{cb} - \omega_1 - i\gamma_{cb})} \right), \quad (2)$$

where $\rho_{aa}^{(0)}$ and $\rho_{bb}^{(0)}$ are obtained by solving the following two equations:

$$\frac{d\rho_{aa}^{(0)}}{dt} = \frac{KpI_{pump}(t)}{(\omega_{pump} - \omega_{ag})^2} + bp(\rho_{bb}^{(0)} - 2\rho_{aa}^{(0)}) - \Gamma_{aa}\rho_{aa}^{(0)}, \quad (3a)$$

$$\frac{d\rho_{bb}^{(0)}}{dt} = \frac{2KpI_{pump}(t)}{(\omega_{pump} - \omega_{bg})^2} + bp(2\rho_{aa}^{(0)} - \rho_{bb}^{(0)}) - \Gamma_{bb}\rho_{bb}^{(0)}. \quad (3b)$$

In the presence of collisions and within the impact approximation limit, the transverse damping elements γ_{ij} could be rewritten in the following way:

$$\gamma_{ij} = \tfrac{1}{2}(\Gamma_{ii} + \Gamma_{jj}) + \eta_{ij}p, \quad i \neq j \quad (4)$$

where p designates the buffer gas pressure and η_{ij} is the dephasing coefficient for the $i \rightarrow j$ transition. Assuming that lasers 1 and 2 could be tuned with an accuracy $\delta\omega$, which is a fraction of the laser linewidth, on the $\omega_{ab} = \omega_1 - \omega_2$ and $\omega_{cb} = 2\omega_1 - \omega_2$ resonances, and if the pressure broadening effect is introduced according to Eq. (4), Eq. (2) becomes

$$\chi^{(3)}(\omega_{out}, \omega_1, \omega_1, -\omega_2) \propto \frac{1}{(\delta\omega - i\eta_{ab}p)(\delta\omega - i\eta_{cb}p)} \left((\rho_{aa}^{(0)} - \tfrac{1}{2}\rho_{bb}^{(0)}) + i\frac{\eta_{cb}p}{\omega_{ab}}(\rho_{aa}^{(0)} + \tfrac{1}{2}\rho_{bb}^{(0)}) \right). \quad (5)$$

Equation (5) clearly shows the dynamics of the interference between two coherent pathways. As the pressure increases, the population in each of the sublevels of the P states increases but also starts to equalize due to the inelastic transfer of population during collisions. This means that the real part of the term in parentheses in Eq. (5) becomes progressively less important relative to the imaginary part. At very high pressure, the imaginary part predominates, but the generated signal does not keep on increasing with pressure because the denominator of Eq. (5) grows at an even faster rate. The fast growth of the denominator is attributed to a pressure broadening of the transition linewidths. Thus, the generated signal will rise as the buffer gas is initially increased, go through a maximum, and finally disappear at high pressure. Equation (2) could be made very insensitive to pressure variations at high buffer gas pressure by just tuning lasers 1 and 2 in the following way: $\omega_{ab} \approx \omega_1 - \omega_2$ and $\omega_{cb} \neq 2\omega_1 - \omega_2$. This was checked in our experiment. Up to now, we have neglected the velocity distribution of the atoms and the finite-laser linewidth of our lasers. In a more rigorous treatment of this problem, we would have to integrate $\chi^{(3)}$ over the velocity distribution of atoms[14] and we would have to convolve $|\chi^{(3)}|^2$ with the spectrum of our lasers.[15] The accuracy of our measurements does not justify such a detailed treatment.

IV. RESULTS AND DISCUSSION

Since we are detecting with a boxcar integrator the integrated intensity and not the instantaneous intensity, the generated signal S will be given by

$$S \propto \int_{-\infty}^{\infty} |\chi^{(3)}|^2 I_1^2(t) I_2(t)\, dt. \quad (6)$$

Equations (3a), (3b), (5), and (6) were iteratively solved and a fit to the experimental results is shown in Fig. (2) using the following parameters: $\eta_{ca} = \eta_{cb} = 21$ MHz/Torr,[16] $\eta_{ab} = 6$ MHz/Torr,[17] $\Gamma_{aa} = \Gamma_{bb} = 16$ nsec, $b = 1.2 \times 10^{-2}$/nsec Torr,[13] and $\delta\omega = 2.2$ GHz. The time dependence of the laser pulse was assumed to be a Gaussian of 5 nsec duration. All the parameters used in the fit were directly measured by us or were taken from the published

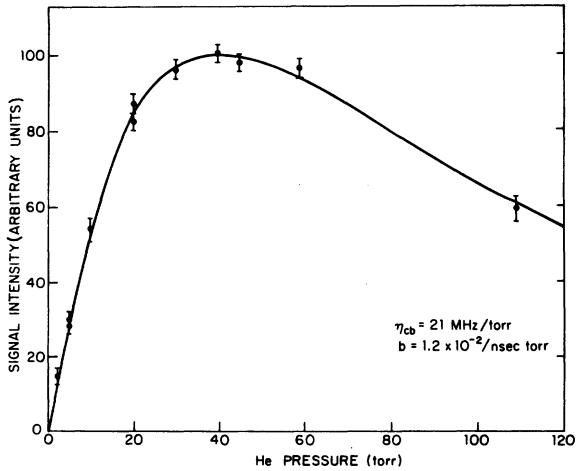

FIG. 2. Dependence of the generated four-wave mixing signal on the buffer gas pressure. The solid line is a theoretical fit to our experimental results using Eqs. (5) and (6).

literature. Only one arbitrary scaling factor and the tuning accuracy $\delta\omega$ were left to be chosen. We get a good fit to the experimental data points. By changing the value of the inelastic-collision cross section via the b parameter and the value of the dephasing coefficient η_{cb}, it is found that the low pressure of the fit is very sensitive to the value of the b parameter and the high-pressure part is very sensitive to the chosen value of η_{cb}. In our experiments, we were not able to see the part of the signal that goes as the square of the sum of the populations in the two P states because of the degradation of our signal-to-noise ratio at very high buffer gas pressure. Measurements to demonstrate this effect would be better done on the ground-state hyperfine levels of Na with a cw single-mode laser since the splitting of the levels is much smaller.[18] Figure 3 shows the intensity dependence of the signal for a definite pressure as we attenuate sequentially both lasers 1 and 2 and the pump laser. The intensity dependence on the pump laser is quadratic, as should be expected since the P states are populated by one-photon absorption and since the generated coherent signal should be proportional to the square of the number of atoms in the excited state. As we attenuate both lasers 1 and 2, we expect an I^3 dependence at low intensity and the onset of saturation at higher intensity. This is in agreement with our measurements. A perturbative approach like the one we have used only applies in the former case. Measurements of the type just described could be used to avoid the problem of light trapping faced in the usual collisional redistribution experiments. In fact, if lasers 1 and 2 are left fixed and if the pump laser frequency is varied, the variation of the population in the P state could be measured directly by monitoring the intensity of the generated signal. Useful information about interatomic Na–rare-gas-atom potential could then be obtained.

In conclusion, we have demonstrated that an incoherent process can be used to trigger the generation of a coherent signal. Reasonable agreement between theory and experiment was obtained. As in the case of previously reported pressure-induced effects in four-wave mixing experiments, collisional redistribution plays a dominant role. Our technique could be used to extract important spectroscopic information about dephasing times and inelastic-collision cross sections in the excited state. We have also predicted in the high-pressure region the appearance of a four-wave mixing signal that goes as the square of the sum of the populations in two levels. This suggests that in the presence of collision a Raman-type resonance should be observable between equally populated states.

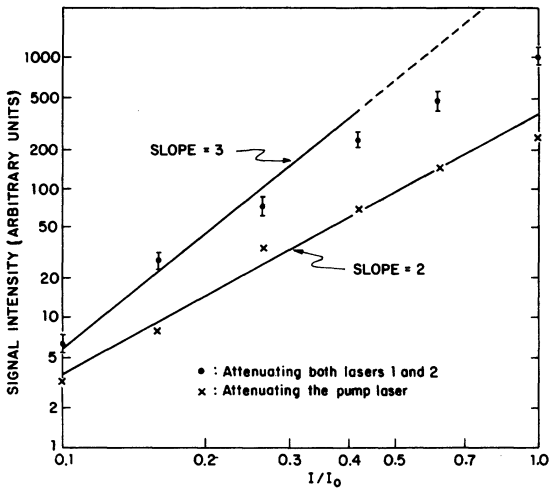

FIG. 3. Intensity dependence of the four-wave mixing signal on the pump laser and on both lasers 1 and 2.

V. ACKNOWLEDGMENTS

I am grateful to Professor Bloembergen for fruitful discussions and for critical reading of the manuscript. I also want to thank the National Research Council of Canada for giving me a NATO fellowship during the time of my stay at Harvard University.

---

*A preliminary report on this work was presented at the 1980 Optical Society of America Meeting in Chicago, Illinois.

†Present address.

[1]N. Bloembergen, H. Lotem, and R. T. Lynch, Jr., Indian J. Pure Appl. Phys. 16, 151 (1978).

[2]N. Bloembergen, in *Laser Spectroscopy IV*, edited by H. Walther and K. W. Rothe (Springer, Berlin, 1979), p. 340.

[3]L. Ducuing, *Nonlinear Optics* (Academic, London, 1977), p. 11.

[4]J. L. Carlsten, A. Szoke, and A. G. Raymer, Phys. Rev. A 15, 1029 (1977).

[5]M. G. Raymer, J. L. Charlsten, and G. Pichler, J. Phys. B 12, L119 (1979).

[6]G. Grynberg, J. Phys. B (in press).

[7]Y. Prior, A. R. Bogdan, M. Dagenais, and N. Bloembergen, Phys. Rev. Lett. 46, 111 (1981).

[8]A. R. Bogdan, M. Downer, and N. Bloembergen, Phys. Rev. A (in press).

[9]C. Cohen-Tannoudji, B. Diu, and F. Laloe, *Quantum Mechanics* (Wiley, New York, 1977), Complement EIII.

[10]A. R. Bogdan, Y. Prior, and N. Bloembergen, Opt. Lett. 6, 82 (1981).
[11]N. Bloembergen, *Nonlinear Optics* (Benjamin, New York, 1965).
[12]See Appendix 4 in A. Yariv, *Quantum Electronics* (Wiley, New York, 1975).
[13]J. Pitre and L. Krause, Can. J. Phys. 45, 2671 (1967).
[14]S. A. J. Druet and J.-P. E. Taran, J. Phys. (Paris) 40, 819 (1979).
[15]M. A. Yuratich, Mol. Phys. 38, 625 (1979).
[16]P. F. Liao, J. E. Bjorkholm, and P. R. Berman, Phys. Rev. A 20, 1489 (1979).
[17]T. W. Mossberg, E. Whittaker, R. Kachru, and S. R. Hartmann, Phys. Rev. A 22, 1962 (1980).
[18]This effect has recently been observed by A. R. Bogdan, M. Downer, and N. Bloembergen, Opt. Lett. 6, 348 (1981).