Cross-Relaxation Effects in the Saturation of the 6328-Å Neon-Laser Line

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We report experiments which show that saturation of the inhomogeneously broadened 6328-Å neon-laser line is strongly affected by cross relaxation which tends to redistribute the excited atoms over the Maxwellian velocity distribution and thus leads to partially homogeneous line saturation. Our results for Ne^{20} discharges agree well with theoretical calculations based on a "strong-collision" model. Results for He^3:Ne^{20} mixtures show deviations from this model that we attribute to the "weak" cross relaxation caused by He-Ne collisions.

In this Letter we report experimental results which demonstrate that saturation of the inhomogeneously broadened 6328-Å neon laser occurs not just for atoms with resonance frequencies in the vicinity of the saturating frequency, but also for atoms with resonance frequencies far removed from this frequency. As well as the usual "hole-burning" in the vicinity of the frequency of the saturating radiation,[1] there is a reduction of gain over a large part of the Doppler-broadened gain curve. These results are in fairly good agreement with a simple theory based on a "strong-collision" model[2] which assumes that cross-relaxation effects redistribute excited atoms uniformly over the Maxwellian velocity distribution. One important implication of this result is that previous measurements of the homogeneous linewidth γ, based on Lamb-dip measurements[3] or nonlinear wave interactions,[4] overestimate the linewidth at high pressures, especially for a pure neon discharge. More accurate values for this parameter are given in this paper.

The experiments reported here are performed by passing two beams of light of the same frequency in opposite directions through a sample discharge. One of these beams is amplitude modulated at an audio rate, and the audio signal imposed on the other beam due to their nonlinear interaction in the sample discharge is detected. By tuning the frequency of these beams away from the atomic resonance frequency, each beam can be made to interact with different velocity groups of atoms, and the gain saturation can be probed over a wide frequency range.

An appropriate theoretical description of this experiment can be made using a semiclassical model, describing the quantum state of atoms with velocity v located at position r at time t statistically by an ensemble-averaged density matrix $\rho(r,v,t)$. The interaction with the two light fields in the dipole approximation can be introduced as a perturbation into the Hamiltonian. Cross-relaxation effects which redistribute atomic excitation in velocity space can be taken into account phenomenologically in the equation of motion obeyed by the matrix ρ. Some physical processes which could contribute to cross relaxation are elastic atom-atom collisions, trapping of resonance radiation,[5] and collisional atomic excitation exchange. Once the matrix ρ is known for atoms of a given velocity v, the corresponding macroscopic polarization of the amplifying medium, and hence its gain and dispersion, can be obtained by averaging the expectation value of the electric dipole moment over the atomic velocity distribution. If we restrict our analysis to small light intensities, use a simple "strong-collision" model, and assume that the Doppler width $\Delta\nu_D$ is large compared with the pressure-broadened linewidth[6] γ, the final result, as pointed out by Szöke,[7] is the same as that expected from a simple "hole-burning" picture.

For the sake of simplicity we restrict our attention to this case. We start from the familiar rate equations for the population densities of the upper and lower laser levels $n_a(v)$ and $n_b(v)$, considered as functions of the axial atomic velocity v. In the absence of velocity-changing collisions, these equations may be written

$$dn_a(v)/dt = \lambda_a(v) - \gamma_a n_a(v) - [I_1\sigma_1(v) + I_2\sigma_2(v)][n_a(v) - n_b(v)], \quad (1)$$

$$dn_b(v)/dt = \lambda_b(v) - \gamma_b n_b(v) + [I_1\sigma_1(v) + I_2\sigma_2(v)][n_a(v) - n_b(v)], \quad (2)$$

where $I_1 = (c/8\pi)|E_1|^2$ is the intensity of laser field 1, and

$$\sigma_1(v) = \frac{4\pi}{\hbar c} \frac{\gamma|\mu_{ab}|^2}{[\gamma^2 + (\omega_{ab} - \omega_1 - k_1 v)^2]} . \tag{3}$$

Here μ_{ab} is the dipole moment of the transition,[8] ω_{ab} is $2\pi\nu_{ab}$ where ν_{ab} is the resonant frequency of a stationary atom, and $\omega_1 = 2\pi\nu_1$ where ν_1 is the frequency of the incident radiation field 1. Corresponding definitions hold for I_2 and σ_2. For the experiments reported here $k_1 = -k_2$ and $\omega_1 = \omega_2$. For the collisional excitation rates λ we assume a Maxwellian velocity distribution, i.e.,

$$\lambda_a(v) = \Lambda_a f(v), \quad \lambda_b(v) = \Lambda_b f(v); \quad f(v) = (\pi\bar{v})^{1/2} \exp(-v^2/\bar{v}^2). \tag{4}$$

Spontaneous and collisional decay processes for the upper and lower laser levels are described by the decay rates γ_a and γ_b, respectively.

To allow for cross-relaxation effects we can modify the rate equations (1) and (2) by adding to the right-hand side a term

$$[dn_\alpha(v)/dt]_{\text{x-relax}} = -n_\alpha(v)\int \Gamma_\alpha(v', v)dv' + \int n_\alpha(v')\Gamma_\alpha(v, v')dv', \quad \alpha = a \text{ or } b. \tag{5}$$

The "collision kernel" $\Gamma(v, v')$ has to obey the principle of detailed balancing. We adopt a "strong-collision" model[2] by which we mean that the probability of finding the atom with velocity v' after a cross-relaxation event is independent of the initial velocity v; i.e., we set

$$\Gamma_\alpha(v', v) = \Gamma_\alpha f(v'), \quad \alpha = a \text{ or } b, \tag{6}$$

where Γ_α is a basic cross-relaxation rate characteristic of level α. Thus, the cross-relaxation terms are simplified to

$$[dn_\alpha(v)/dt]_{\text{x-relax}} = -\Gamma_\alpha n_\alpha(v) + \Gamma_\alpha f(v)\int n_\alpha(v')dv'. \tag{7}$$

We are interested in the perturbations $\Delta n_\alpha(v) = n_\alpha(v) - [n_\alpha(v)]_{I_1=0}$ of the population densities n_α, $\alpha = a$ or b, caused by the saturating field 1. Assuming small light intensities and considering only steady-state solutions, we obtain

$$\Delta n_a(v) = -\frac{I_1(N_a{}^0 - N_b{}^0)f(v)}{\Gamma_a + \gamma_a}\left[\sigma_1(v) + \frac{\Gamma_a}{\gamma_a}\int \sigma_1(v')f(v')dv'\right], \tag{8}$$

where $N_\alpha{}^0 = \Lambda_\alpha/\gamma_\alpha$; $\alpha = a$ or b. Starting from Eq. (2) we obtain corresponding results for Δn_b with a and b interchanged. Obviously the first term in the square brackets gives the Lorentzian "hole," i.e., the inhomogeneous saturation of the population density, whereas the second term describes a Gaussian background due to the cross relaxation.

The corresponding change of the probe-field transition rate, i.e., the measurable signal S, can now be calculated:

$$S \equiv \Delta\int I_2\sigma_2(v)[n_a(v) - n_b(v)]dv = -I_2 I_1(N_a{}^0 - N_b{}^0)\left\{\left[\frac{1}{\Gamma_a + \gamma_a} + \frac{1}{\Gamma_b + \gamma_b}\right]\int f(v)\sigma_1(v)\sigma_2(v)dv + \left[\frac{\Gamma_a/\gamma_a}{\Gamma_a + \gamma_a} + \frac{\Gamma_b/\gamma_b}{\Gamma_b + \gamma_b}\right]\int f(v)\sigma_1(v)dv\int f(v)\sigma_2(v)dv\right\} . \tag{9}$$

The integrals can be evaluated by means of the tabulated plasma dispersion function. In the limit $\gamma/\Delta\omega_D \ll 1$, where $\Delta\omega_D = |k|\bar{v} = 2\pi\Delta\nu_D$, and with the definition $|\nu_1 - \nu_{ab}| = \Delta\nu$, we obtain

$$S = -I_1 I_2(N_a{}^0 - N_b{}^0)\left(\frac{\pi}{c\hbar}\right)^2|\mu_{ab}|^4\frac{8}{\pi^{1/2}\Delta\nu_D}\left\{\left[\frac{1}{\Gamma_a + \gamma_a} + \frac{1}{\Gamma_b + \gamma_b}\right]\frac{\gamma}{\gamma^2 + (2\pi\Delta\nu)^2}\exp\left(\frac{-\Delta\nu^2}{\Delta\nu_D{}^2}\right) + \left[\frac{\Gamma_a/\gamma_a}{\Gamma_a + \gamma_a} + \frac{\Gamma_b/\gamma_b}{\Gamma_b + \gamma_b}\right]\frac{1}{2\pi^{1/2}\Delta\nu_D}\exp\left[\frac{-2(\Delta\nu)^2}{\Delta\nu_D{}^2}\right]\right\}. \tag{10}$$

We see, then, that with these approximations the measured signal consists of a peak of width $\sim\gamma/2\pi$ and a wide Gaussian background.

Experiments were performed with the apparatus shown in Fig. 1. The single-frequency 6328-Å He-Ne laser beam from laser 1 is split into two components. One beam passes directly through the gas discharge tube; the other is chopped and passed through the discharge tube in the opposite direction.

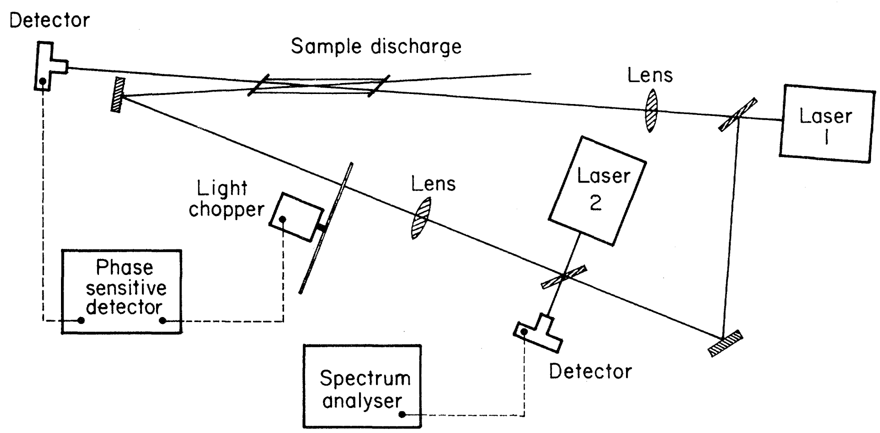

FIG. 1. Experimental setup. When measuring the interaction signal, the beam from laser 2 is blocked off.

A similar setup using beams traveling in the same direction was described by Shank and Schwarz.[4] The gas pressure in the discharge tube is monitored with a thermocouple vacuum gauge. Maximum interaction is obtained by focusing the two beams into a short length of active discharge. A second laser is stabilized to line center. Its output is then beat with the output from laser 1 in a square-law detector and the beat frequency observed with a spectrum analyzer. In this way the frequency detuning of laser 1 from line center can be measured accurately. After the frequency detuning is measured, light from laser 2 is blocked off so that no light from laser 2 enters the sample discharge during a measurement of an interaction signal.

Experiments were performed with various pressures of a 7:1 mixture of He^3:Ne^{20} or pure Ne^{20} in the discharge tube. Figure 2 shows typical sets of data obtained with 1.1 Torr of 7:1 He^3:Ne^{20} and 2.2 Torr of Ne^{20}. The plotted normalized signal has been corrected for the variation in intensity of the probing laser as it is tuned across the gain curve. The results are fitted, using a least-squares curve-fitting computer program, by the expression

$$S=A\left\{\frac{\gamma^2}{\gamma^2+4\pi^2(\Delta\nu+\delta)^2}+C\exp\left[\frac{(\Delta\nu+\delta)^2}{\Delta\nu_D{}^2}\right]\right\}\exp\left[\frac{(\Delta\nu+\delta)^2}{\Delta\nu_D{}^2}\right], \qquad (11)$$

where A, γ, C, and δ are adjustable parameters. The parameter A is a scale factor. The parameter δ is included to allow for the fact that the frequency-stabilized reference laser may not be stabilized to exactly line center. C is the cross-relaxation parameter. The values of γ and C are the useful data and these results are presented in Fig. 3. The value $\Delta\nu_D$ was taken to be 850 MHz, corresponding to a gas temperature of 350°K. A typical fit of the theoretical curve to the experimental data is shown in Fig. 2. The dashed line is the curve using Eq. (11) and the solid line is a curve for no cross relaxation ($C=0$) fitted to the experimental data at the peak and $\frac{1}{2}$-signal points. It is clear that the in-

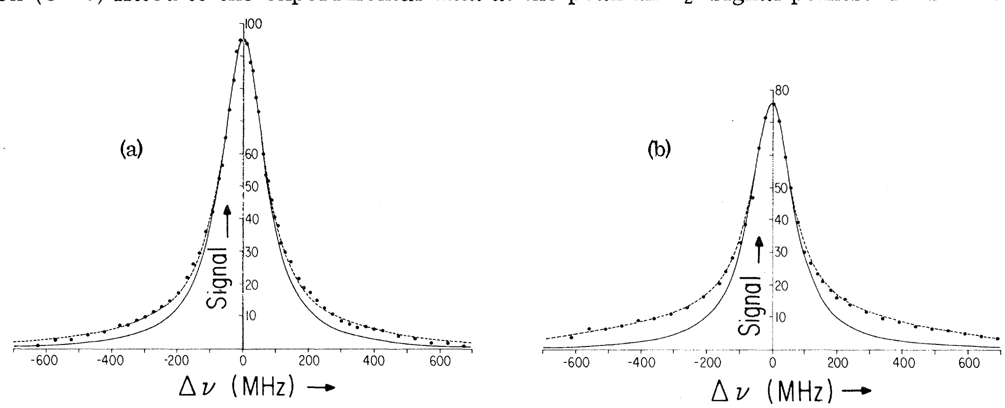

FIG. 2. Typical data for (a) 1.1 Torr 7:1 He^3:Ne^{20} and (b) 2.2 Torr Ne^{20}. In both cases the dashed line is the curve for no cross relaxation.

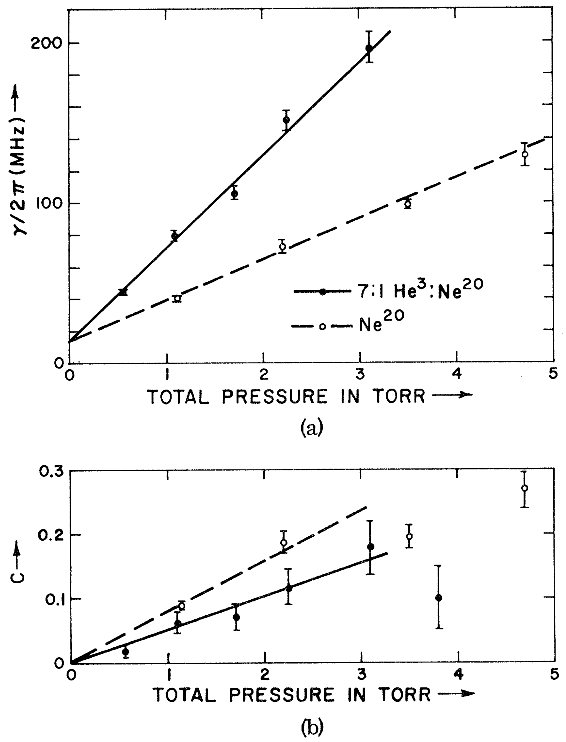

FIG. 3. Experimental results for (a) linewidth γ and (b) cross-relaxation parameter C versus pressure for a 7:1 mixture of He^3:Ne^{20} and for pure Ne^{20} discharges. The error bars denote two standard deviations in the parameters determined by a least-squares curve-fitting computer program.

clusion of the Gaussian background greatly improves the fit with the experimental data. Note that although the "strong-collision" model gives a good fit to the Ne^{20} data, the 7:1 He^3:Ne^{20} data do not fit well in the wings of the curve. We believe that this is due to the fact that collisions between a He and a Ne atom cause velocity shifts of the Ne atom that are much less than in the case of Ne-Ne collisions because of the smaller mass of the He atom. The "strong-collision" model does not take account of small velocity shifts characteristic of the relatively weak He-Ne collisions.[2,9]

Figure 3 shows the "best-fit" values of γ and C for Ne^{20} and 7:1 He^3:Ne^{20} mixtures as a function of total gas pressure. Note that although very large cross-saturation effects are observed in the case of Ne^{20}, the cross-saturation effects in the He^3:Ne^{20} mixtures are still appreciable. For example, the data for 1.1 Torr 7:1 He^3:Ne^{20} indicate that approximately 20% of the atoms contributing to the gain come from outside the region of the "hole" to be expected in the absence of cross saturation.

We have shown that the saturation behavior of the 6328-A Ne-laser transition is strongly affected by cross-relaxation effects. A more complete theory including "weak" cross-relaxation effects characteristic of He-Ne collisions, and a discussion of some of the implications of these cross-relaxation effects, will appear in a subsequent publication now in preparation.

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[6]The relaxation rate γ has units of sec^{-1}. Linewidth measurements of γ usually quote values of $\gamma/2\pi$ in units of ordinary frequency.

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[8]In order to properly take account of the m degeneracy of the 6328-Å Ne-laser levels, different μ's should be used for the different allowed transitions. The final result is not significantly altered, however, if a single μ is used as we have done here.

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