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Dispersive Character and Directional Anisotropy of Saturated Susceptibilities in Resonant Backward Four-Wave Mixing

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Heterodyne detection of backward four-wave mixing in resonant gas media allows one to explore both real and imaginary parts of the reemitted electromagnetic fields. It provides the first experimental proof of the dispersive character of nonlinear susceptibilities in the saturated regime. It also allows one to demonstrate the strong directional anisotropy of the saturation induced by either forward or backward pump wave.

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Recent work in phase-conjugate (PC) optics[1] has been partly stimulated by the quest of large PC mirror reflectivities. In comparison with solid materials which can exhibit high reflectivities,[2] one advantage of gas media is to accept very intense irradiations without irreversible damage. Recently, Lind and Steel[3] got a cw efficiency exceeding unity by resonant backward degenerate four-wave mixing (DFWM) in Na vapor. Up to now, current experimental information has been limited to the *intensity* of PC emission. In this Letter, we report on the results obtained via a powerful technique based on an optical heterodyne detection scheme[4] which allows one to monitor simultaneously the real and imaginary parts of the nonlinear susceptibility (χ_{NL}) responsible for PC emission, and we give the first experimental evidence for the dispersive character of *saturated* DFWM in resonant gas media. We also demonstrate the strong directional anisotropy of the saturation induced by either forward or backward pump wave, and present the general outline of a strong-field theoretical analysis supporting all these observations.

From the very first experiments of cw DFWM conjugation in resonant gas media (Liao, Bloom, and Economou[5]), one knows that the PC line shape exhibits two main characteristics: (i) At low intensities, it is Doppler free, at least for small crossing angles between pump and probe, and (ii) for increasing (and equal) pump intensities, the emission line shape broadens and splits, tending towards a fully resolved double-peak structure. As an example, such a frequency behavior is shown in Fig. 1(a), in the case of resonant DFWM at $\lambda = 640$ nm in a neon discharge [transition $1s_5$ $(J=2) \rightarrow 2p_9$ $(J=3)$]. Up to now, two interpretations have been suggested. (i) One is based upon ac Stark splitting[6]; however, one should expect a substructure in both peaks, reflecting the different Rabi frequencies from each transition between Zeeman sublevels. (ii) Another one is based upon a faster saturation of the "absorption" component (imaginary part) of the nonlinear susceptibility χ_{NL}, as compared to the "dispersion" component: At saturation, the total intensity would behave like the square of a dispersion curve. This behavior has been inferred by Woerdman and Schuurmans[7] from a simplified model based on the nonlinear response of *stationary* two-level atoms to a cw plane wave. However, the validity of such a model is questionable, since it is well known[8] that, in the absence of propagation effects, an exact, nonperturbative treatment of DFWM in stationary absorbing media always pre-

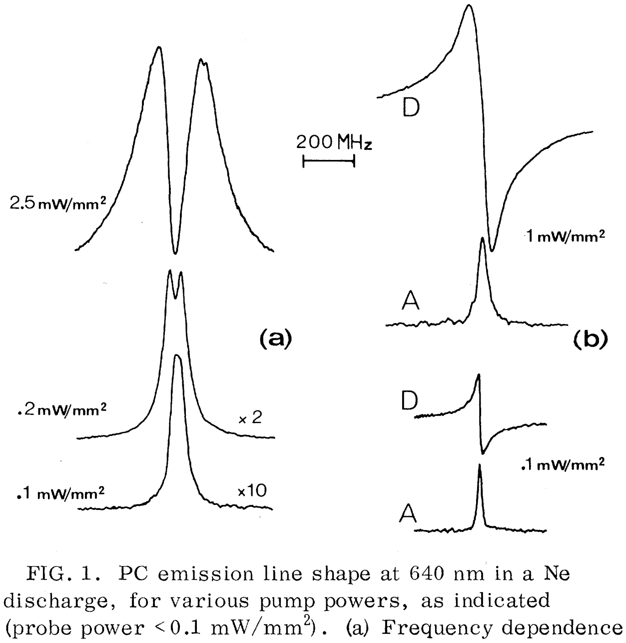

FIG. 1. PC emission line shape at 640 nm in a Ne discharge, for various pump powers, as indicated (probe power < 0.1 mW/mm²). (a) Frequency dependence of the PC *intensity*. (b) Heterodyne detection (at frequency $2\delta = 44$ MHz) of the PC field *amplitude*. A, absorption. D, dispersion.

dicts a single-peak line shape: A comprehensive treatment should take into account the decisive influence of the inhomogeneous broadening coming from the velocity distribution.

To get a deeper understanding of these nonlinear processes, we have used a heterodyne detection technique (Fig. 2) in which the amplitude of the conjugate field is monitored. In principle, the optical setup, similar to that described in Ref. 4, consists in two standing waves (pump wave, frequency ω; probe, $\omega+\delta$) crossing in a Ne discharge cell at an angle $\theta \simeq 1°$. These two beams are generated by an acousto-optic deflector (AOD) operated with two rf frequencies, and are kept collimated along their path by means of convenient optics (Fig. 2). This allows us to produce a constant saturation over the interaction volume. Via nearly degenerate four-wave mixing (FWM) processes, two conjugate fields at frequency $\omega-\delta$ are reemitted counter-directionally to either probe, and are observed through their heterodyne beating with the other probe, at frequency 2δ. These beat signals, I_1 or I_2, are monitored by a high-frequency phase-sensitive (lock-in) detector, yielding phase and quadrature components simultaneously. With an adequate choice of the reference phase, one gets signals proportional to the real and imaginary parts of χ_{NL}.

Figure 1(b) shows the beat-signal line shape for various (and equal) pump intensities, at $\lambda = 640$ nm. At low pump intensities, χ_{NL}, as predicted by third-order perturbation theory,[9,10] is Doppler free and satisfies a Kramers-Kronig-type relationship, $\chi_{\mathrm{NL}} \simeq \chi^{(3)} \propto (\nu - i\gamma)^{-1}$ where $\nu = \omega - \omega_0$ is the frequency detuning and γ the homogeneous linewidth. Absorption (A) and dispersion (D) contributions have the same amplitude. With increasing pump intensities, Kramers-Kronig relations break down, and a striking difference appears between A and D amplitudes. In the fully saturated regime, D/A *amplitude* ratios as large as 7 were observed on several neon transitions (involving different angular momenta). This result fully justifies the conclusion that the emission line shape follows the *square* of a dispersion curve.

To further discriminate between the optical saturation induced by *forward* (f) and *backward* (b) pump wave, a series of experiments has been performed in which the return pump wave of Fig. 2 was strongly attenuated, in order to isolate the saturation produced by a single, intense running pump wave. Now the beat signals I_1 and I_2 behave differently. Figures 3(a) and 3(b) show the signal line shapes respectively observed on I_1 and I_2, at $\lambda = 607$ nm [Ne transition $1s_4$ ($J=1$) $\rightarrow 2p_3$ ($J=0$)]. In these experiments, the two return beams (pump and probe) have parallel linear polarizations, orthogonal with the incident ones (the cell being placed between cross polarizers). The directional anisotropy of the pump saturation appears in the following remarkable features: (i) When the saturating beam is the *forward* pump (pump nearly copropagating with the probe), A and D components keep equal amplitudes, but the optical saturation affects their line shapes [I_1, Fig. 3(a)]: At the onset of the saturating regime, a narrow structure appears at line center, on both components. At full saturation, this structure has grown to a full-scale amplitude, dramatically altering the original A and D line shapes. Also the overall A-D amplitudes, after reaching a maximum value, decrease with increasing f pump intensity and go back to zero. Note that, at saturation, the double-peak structure of the *intensity* line shape is no longer of dispersive origin, but is mainly caused by the characteristic absorption line shape which exhibits a minimum at line center. (ii) For a *backward* saturating pump [I_2, Fig. 3(b)], the main feature is the change in the A/D ratio, producing a strongly dispersive character similar to the one observed for equal pump intensities. At saturation, the signal amplitude reaches a finite (nonzero) value, which is much larger (by at least one order of magnitude) than the one obtained for a forward saturating pump.

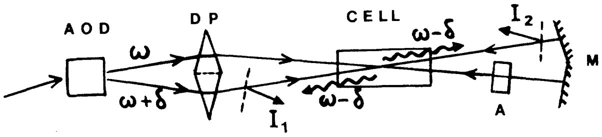

FIG. 2. Basic scheme for heterodyne detection of FWM emission (AOD, acousto-optic deflector; DP, double prism; A, attenuator, used in experiments of Fig. 3).

All these observations can be interpreted with a density-matrix calculation of the nonlinear susceptibility, limited to first perturbation order in the probe and weak pump fields, but taking into account through a nonperturbative treatment the effect of arbitrary intensities of the strong pump.[11] In this calculation, the $J=1 \rightarrow 0$ transition interacting with cross-polarized fields is described as a three-level system (with degenerate frequencies). Probe and pump f interact with one transition, while cross-polarized pump b interacts

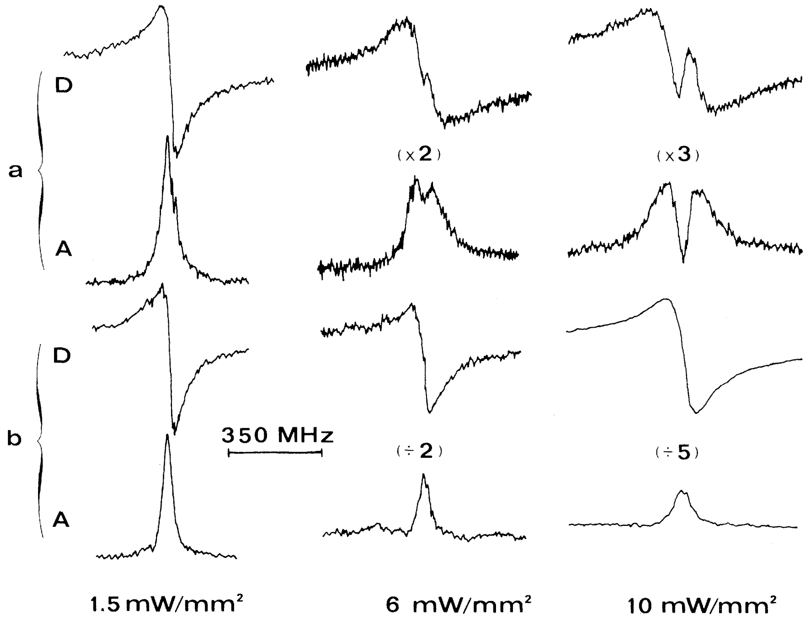

FIG. 3. PC emission line shape at 607 nm, in Ne with a single saturating pump (powers are as indicated; $2\delta = 44$ MHz). Probe and return pump intensities are kept below a nonsaturating level, $\lesssim 0.1$ mW/mm^2. (a) Forward saturation (I_1, Fig. 2); (b) backward saturation (I_2).

with the coupled transition. The final result is averaged over the velocity distribution. In the Doppler limit, for small crossing angles, one gets analytical expressions for $\chi_{\rm NL}$. As an example, an intense *forward* pump produces a saturated $\chi_{\rm NL}$ of the form

$$\chi_{\rm NL} \propto \frac{2+S}{2(1+S)^{3/2}}\,\frac{\nu - i\Gamma'}{(\nu - i\Gamma)^2}$$

with $\Gamma = [1 + 3(1+S)^{1/2}]\gamma/4$ and $\Gamma' = [1 + 6(1+S)^{1/2}/(2+S)]\gamma/4$. One assumes a single relaxation rate γ, and $S = \Omega^2/\gamma^2$ is the normalized f-pump intensity (Ω, Rabi frequency). For a *backward* saturating pump, the expressions are more complicated.[11] Figures 4(a) and 4(b) show that the DFWM line shapes predicted by such calculations accurately reproduce the main experimental features.[12]

A number of experimental results may be apprehended if one recalls that, in presence of the thermal motion, the contribution of the population grating induced by probe (p) and forward pump (f) is predominant, because of its large spacing. Thus, if pump f is the intense one, it saturates the transition on which the population grating is created. At full saturation, the grating contrast tends to vanish, as does the light interference pattern produced by probe and pump f: The overall PC intensity goes to zero [Fig. 4(a)].

Such a behavior does not exist for a *backward* (b) saturating pump. The p-f grating contrast is not directly affected by pump b saturation. However, as pump b saturates the atomic system more and more strongly on line center, it explores the atomic response further in the line wings, where the *dispersive* character becomes predominant. The emission saturation may be

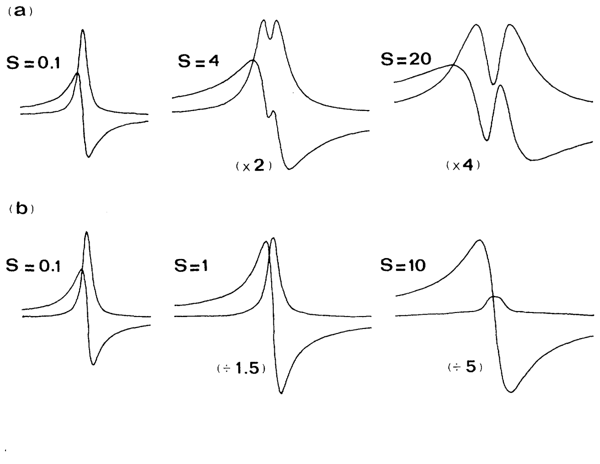

FIG. 4. Theoretically predicted DFWM line shapes ($S^{1/2}\chi_{\rm NL}$ vs ω) in a single-lifetime model. (a) f saturating pump, (b) b saturating pump.

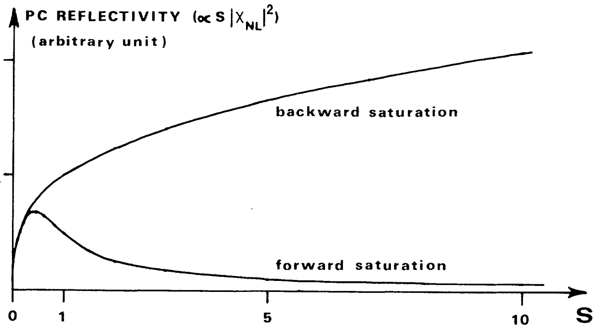

FIG. 5. Theoretical peak reflectivity vs (f or b) saturating pump intensity.

understood by realizing that the conjugate field is radiated at the expense of an equal number of photons from the *two* pumps. If pump f is kept at a finite, nonsaturating intensity level, the emission intensity cannot increase indefinitely. Figure 5 shows the PC reflectivity as a function of the saturating pump intensity, and exemplifies the anisotropic behavior of the DFWM efficiency. As regards D/A ratio, this directional anisotropy presents some analogy with saturated absorption: One knows[13] that a strong traveling wave saturates its own absorption but not its refraction index, while it saturates equally the absorption rate and the index of a weak counter-propagating field. This feature is not specific to a three-level system: Indeed, a fifth-order perturbation treatment for two-level systems predicts a similar behavior.

This set of results demonstrates that heterodyne detection is an ideal tool for an accurate knowledge of high-order nonlinear susceptibilities. This knowledge should bring a substantial help for improving the reflectivity of PC mirrors, and more generally for the operation of PC resonators.[3] The directional anisotropy, which is a typical effect of inhomogeneous broadening, suggests that an optimum ratio of the two pump intensities must exist. Work in this direction is under progress.

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[1] See, e.g., A. Yariv, IEEE J. Quantum Electron. 14, 650 (1978); M. Ducloy, in *Festkörperprobleme—Advances in Solid State Physics* (Vieweg, Braunschweig, 1982), Vol. XXII.

[2] J. Feinberg and R. W. Hellwarth, Opt. Lett. 5, 319 (1980).

[3] R. C. Lind and D. G. Steel, Opt. Lett. 6, 554 (1981).

[4] D. Bloch, R. K. Raj, J. J. Snyder, and M. Ducloy, J. Phys. (Paris), Lett. 42, L31 (1981).

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

[6] D. J. Harter and R. W. Boyd, IEEE J. Quantum Electron. 16, 1126 (1980).

[7] J. P. Woerdman and M. F. H. Schuurmans, Opt. Lett. 6, 239 (1981).

[8] R. L. Abrams and R. C. Lind, Opt. Lett. 2, 94 (1978), and 3, 235 (1978).

[9] M. Ducloy and D. Bloch, J. Phys. (Paris) 42, 711 (1981), and 43, 57 (1982).

[10] Note that a singlet transition yields a *single* resonance, contrary to heterodyne spectroscopy [*collinear* nearly degenerate FWM; see R. K. Raj, D. Bloch, J. J. Snyder, G. Camy, and M. Ducloy, Phys. Rev. Lett. 44, 1251 (1980), and Ref. 9], in which one gets *doublets*, because pump and probe are geometrically degenerate.

[11] D. Bloch and M. Ducloy, to be published.

[12] A quantitative comparison between theoretical and experimental curves would need a more sophisticated theory taking into account collisional relaxation, Doppler motional broadening (due to nonzero θ), and cascade effects induced by spontaneous emission (Ref. 11). Another delicate point lies in the propagation effects (pump self-focusing and defocusing affecting the beam overlap, etc.).

[13] See, e.g., B. Couillaud and A. Ducasse, thesis, Université de Bordeaux, 1978 (unpublished).