Spontaneous Generation of a Longitudinal Atomic Density Grating in Sodium Vapor

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(Received 18 July 1995; revised manuscript received 6 November 1995)

We present measurements of probe beam amplification in Na vapor at intermediate density values, in both the counterpropagating and copropagating directions (with respect to the pump beam) in the case of degenerate pump and probe frequencies. Strong gain is observed only in the counterpropagating direction for parameters that preclude the possibility of Raman or Rayleigh gain. We show that the experimental results can be interpreted in terms of the spontaneous formation of a density grating due to the nonresonant interaction between the electromagnetic field and the atoms.

PACS numbers: 34.50.–s, 42.25.Fx

The interaction between a coherently pumped collection of atoms and light has been studied extensively both theoretically and experimentally [1]. Gain coming from a variety of different physical mechanisms has been predicted and observed in systems that can be modeled by two-level or three-level atoms. Several experiments have confirmed the existence of the Rayleigh gain and Raman gain [2], as predicted by the theory of Mollow. Recently, experiments performed in sodium vapor have used these types of gain to obtain lasing action [3–5]. Gain has also been predicted, and observed, for a three-level atom in the absence of population difference [6–8]. Atomic recoil, coupled to the optical field, has also been considered as a mechanism for optical gain [9]. Recent measurements have confirmed the exploitability of mechanical energy as a source for the electromagnetic field [10]. We remark that these gain mechanisms can be described in terms of a single-atom interaction, averaged over an ensemble. Gain in the direction opposite to that of a pump beam has been recently predicted [11], as stemming from a collective instability of the medium's spatial distribution.

Here we present observations of gain for a probe beam that is frequency degenerate and counterpropagating with respect to the pump beam. We find that gain does not exist for a probe beam in the copropagating direction for our experimental conditions. We attribute this gain to a new phenomenon which is based on a collective rather than a single-atom effect: The counterpropagating gain is produced by an atomic density grating, formed via the interaction between the coherent pump beam and the moving atoms, which backscatters the pump beam into the probe beam.

An Ar^+ laser-pumped Coherent 899 dye laser, operating in single longitudinal and single transverse (TEM_{00}) mode with a frequency stability of 20 MHz is used to pump a sodium vapor cell (Fig. 1). This pump beam is focused to an $\approx 30\ \mu$m radius in the center of the oven. Part of the beam is split away before reaching the oven and is then separated with a 50% beam splitter into two seed beams. Each of these beams is individually mode matched to the fundamental mode of a ring cavity built around the Na-filled oven, at a small angle ($\approx 5°$) with respect to the pump beam. The beam waist of the cavity has a 370 μm radius and is located about 14 cm away from the center of the oven towards the pump laser. We use high reflectivity (99.6%) mirrors, thereby obtaining a very high theoretical cavity finesse estimated at about 88, considering the losses at the windows, while the measured experimental value is about half the theoretical one. The free spectral range (FSR) of the cavity is about $2 \times 10^8\ s^{-1}$, and the cavity resonance frequency is continuously scanned at about 400 Hz.

We pump and probe the vapor with circularly polarized light of the *same* frequency for all beams. The laser power is constantly monitored by detector D_3 (cf. Fig. 1). The oven is made out of stainless steel [12] and is heated by a resistor which is double coiled in order to compensate for the magnetic field generated by the heating current. The oven aperture is 1.5 mm in diameter and the length of the interaction region is 6 mm. Outside the oven, a flow of argon at low pressure (≈ 0.7 mbar) is

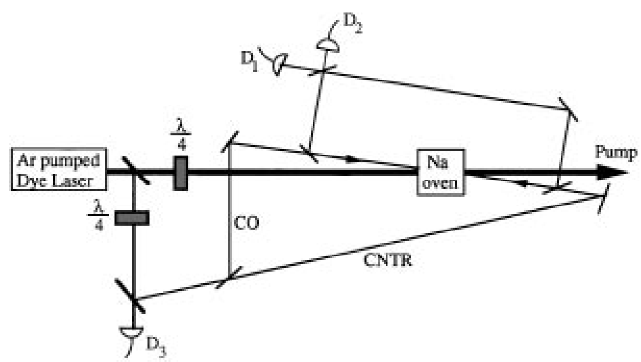

FIG. 1. Schematic of the experimental setup. The light of the cw dye laser is circularly polarized with quarter-wave plates ($\lambda/4$) for the pump beam (Pump), the counterpropagating probe beam (CNTR), and the copropagating probe beam (CO). Detector D_3 monitors the total power of the laser, used for normalization. Detector D_1 monitors the power of the probe beam in the copropagating direction and D_2 in the counterpropagating one. Probe beam power is measured by synchronous detection and only one probe is injected into the cavity at a time.

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used to prevent Na atoms from reaching the antireflection-coated windows. Changing Ar pressure, measured before the gas inlet, from 0.5 to 0.9 mbar, does not qualitatively affect the observations reported below. The buffer gas pressure is low enough to ensure an inhomogeneously broadened linewidth in the absence of the pump field. A lock-in amplifier averages in time the power of either probe beam transmitted by the cavity. Its resonance frequency is scanned over four FSRs at about 400 Hz and for each beam the output is normalized to the laser power. Measurements of the power of each seed beam are taken, in the presence and in the absence of the pump beam.

The time averaged, normalized power of the counterpropagating probe beam is shown in Fig. 2(a) as a function of detuning, in the presence and in the absence of the pump beam. Near the line center (Fig. 2) we observe the usual absorption of the probe beam corresponding to the sodium line. When the detuning is increased towards higher frequencies, an amplification of up to 30% is observed. We remark that we observe actual gain for this counterpropagating seed beam, as proven by the fact that at large detuning the probe beam's transmitted power decreases towards the value it takes in the absence of sodium vapor (not shown in the figure). In the copropagating case instead [Fig. 2(b)], the probe signal in the presence and in the absence of the pump beam has the same form for all frequencies. The maximum amplification of the counterpropagating probe beam is obtained for a detuning of about 3.5 GHz, as seen in Fig. 2(a). The Rabi frequency for the pump power density of Fig. 2 is about 16 GHz.

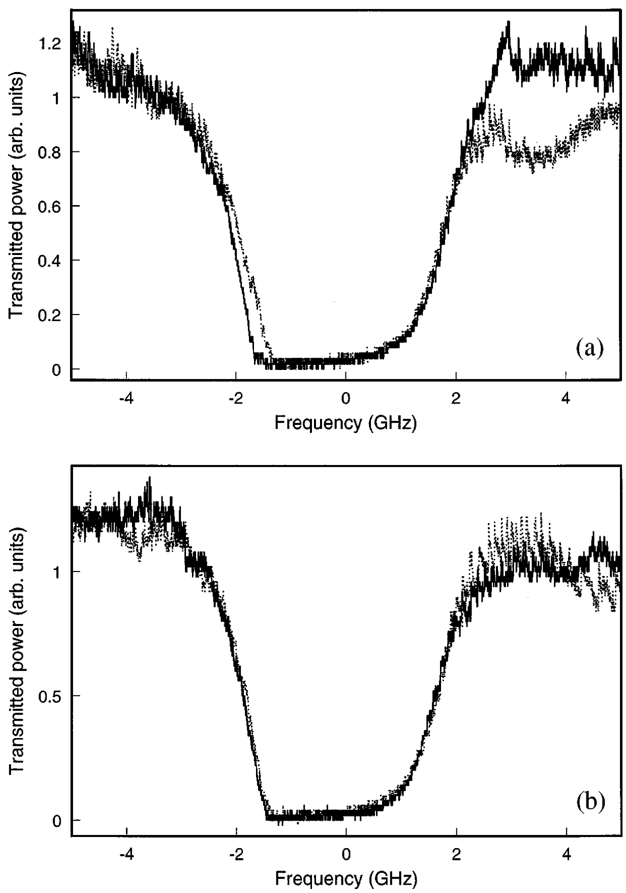

FIG. 2. (a) Amplification of the probe in the counterpropagating direction with respect to the pump beam. The two curves represent the power of the probe signal synchronously detected at the output of the ring cavity in the presence (solid line) and in the absence (dotted line) of the pump beam for sodium density $\approx 1.6 \times 10^{13}$, and pump power ≈ 400 mW. (b) Transmitted power for the probe beam in the copropagating direction under the same experimental conditions as in (a). In this case, the two curves are practically indistinguishable; the small discrepancies change from one measurement to the next and are to be attributed to deviations from linearity in the frequency scan. Note that the frequency scale in this figure (and in Fig. 3) is obtained indirectly from a separate reference and thus its readings should be taken as qualitative.

This gain, which depends strongly on the direction of propagation of the probe, also changes with the pump power and the atomic density. For low pump, there is no net gain in either direction. When the pump power exceeds a well-defined critical value amplification is observed. For increasing pump power, the gain increases until a saturation value is reached. This saturation effect depends on the atomic density: Saturation occurs sooner (i.e., for lower pump powers) at lower vapor densities. In our experiment, we observe gain for pump values greater than 200 mW at a vapor density estimated at 5×10^{12} atoms/cm^3. Because we focus the pump beam down to a spot size of about 30 μm, at the critical pump value for the appearance of gain the power density in the center of the oven is 7×10^3 W/cm^2. Amplification of the counterpropagating probe beam is observable until the maximum power density experimentally available (1.6×10^4 W/cm^2) is reached, although there are indications that a saturation may exist.

Gain is observed for temperatures in the range of 180–240 °C, which correspond to atomic densities of 2.5×10^{12} to 1.6×10^{13} atoms/cm^3 [13], for detuning values that change with density. At 180 °C, the maximum gain is observed at a detuning around 1.5 GHz and at 240 °C the maximum shifts towards 3.5 GHz. Since the frequency at which maximum gain occurs shifts towards resonance with decreasing atomic density, at temperatures lower than 180 °C absorption becomes the predominant effect. At temperatures greater than 240 °C, strong nonlinear effects such as radiation trapping [14] make it difficult to interpret experimental results. We also measured the depletion of the pump as a function of frequency in the presence and in the absence of a counterpropagating seed. A typical result is shown in Fig. 3, where it is obvious that the pump power drops nonlinearly whenever there is a net gain for the counterpropagating probe. Finally, we have also explored the influence of the position of the pump beam waist with respect to the center of the oven. Depending on the position of the waist (before or after the interaction region) the gain appears for either sign of the detuning value.

Our interpretation of these results is based on the following empirical facts: (i) that gain exists in the

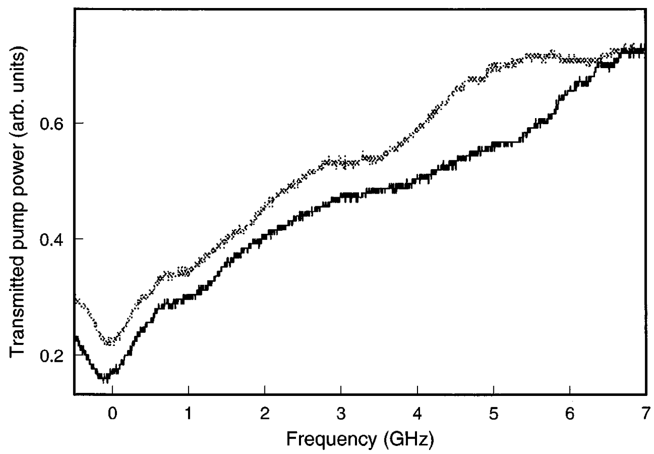

FIG. 3. Transmitted pump power through the cell. Solid line: Transmitted pump power in the presence of a counterpropagating seed beam. Dotted line: Transmitted pump power in the absence of a probe beam. The experimental conditions are similar to those of Fig. 2.

counterpropagating direction only, for a probe frequency identical to the pump frequency; (ii) that gain reaches a maximum for a nonzero detuning that is different from the Rabi frequency; (iii) that gain and the detuning value where it occurs depend on the atomic density; (iv) that there is a clear critical value of the pump power for gain to occur, and that it is independent of the probe power; (v) that the pump is depleted whenever the counterpropagating seed shows net gain; and (vi) that the position of the beam waist affects the sign of the detuning for which we observe gain.

From these observations, we can safely eliminate the possibility that the amplification we see is due to an elastic scattering process, such as Rayleigh or Raman gain, processes well described in [2]. In both those cases, the sodium vapor is pumped by a coherent field whose frequency is detuned from the atomic resonance. Raman gain is seen only for probes detuned from the pump frequency by approximately the Rabi frequency, while in the case of Rayleigh gain the necessary pump-probe detuning is between 20 and 300 MHz [5]. In addition, since Rayleigh gain changes sign at resonance, it should vanish for degenerate probe and pump frequencies. Both of these effects are stronger in the copropagating direction than in the counterpropagating direction [3,5]. As explained in Ref. [5], this is due to the averaging over the velocity distribution which reduces the gain strongly for the counterpropagating beam. Rayleigh lasing action has been almost exclusively observed in the presence of homogeneous broadening (which is not the case in our experiment) because it would require a much stronger field without a buffer gas. Finally, other nonlinear effects, such as self-focusing and self-defocusing, are frequency selective but not sensitive to the direction of propagation and may also be eliminated as possible explanations, even if a small amount of focusing or defocusing can be expected.

In any case, scattering processes do not predict the existence of a critical pump value for probe amplification because they are simply proportional to the amount of pump incident on a single atom, integrated over the ensemble. Hence, a macroscopic amplification of the counterpropagating probe, coming from a single-atom scattering process (integrated over many atoms), would grow continuously with pump strength, regardless of its magnitude. The existence of a critical value, instead, leads us to consider a collective phenomenon as the source of our observations.

What we propose is an interpretation based on the self-generation of an inhomogeneous distribution of atoms in space [11], which implies the existence of a spatial instability, breaking the continuous translational symmetry for the atomic distribution. Thus the critical pump and density values for the occurrence of gain are real thresholds for the system. Recall that the interaction occurs far from resonance, with negligible absorption, so that we have a collection of unexcited, polarized atoms. We are thus proposing the existence of a *density grating* which is responsible for the appearance of gain in the counterpropagating direction through partial backscattering of the pump beam. This interpretation is supported by an estimate of the efficiency of the dispersive relative to the absorptive contribution to the grating [15]. For our experimental parameters, including Rabi nutation, the efficiency of the phase grating turns out to be 50 times stronger than that of the amplitude grating.

The physical mechanism that gives rise to the instability can be illustrated as follows. The presence of a strong pump beam polarizes the atoms. Thus, the source for the gain of the probe is provided by a polarization term which is induced by the pump field on the atoms. The application of a probe beam in the counterpropagating direction forms a very weak periodic potential, which implies a force that acts on the atoms. Through momentum transfer by elastic scattering of pump and probe photons, the atoms are moved towards the minima of the potential, thus creating an inhomogenous spatial distribution. Once begun, this grating "amplifies" the probe beam through backscattering of the pump, which in turn implies a stronger potential, and this positive feedback ensures the growth of an atomic grating.

The existence of a threshold in pump power for the formation of a grating makes sense as the strength of the force spatially localizing the atoms has to be sufficiently large to overcome all other effects which tend to reestablish a uniform atomic distribution (such as atomic motion, velocity spread, and short interaction time). At high densities collisions tend to wash out the grating formed and the effect is harder to achieve. Obviously we do not need to have stationary atoms for the grating to appear; it suffices to regroup them in velocity classes that have a longitudinal component consistent with the formation of a dynamical periodic structure.

It is important to note that the density grating is a dynamical entity, changing in amplitude and phase relative to the total electromagnetic field, and thus affecting the dynamics of other variables, such as the atomic polarization, which in turn affects the atomic density and the backscattered field. In our case the spatial phase shift between atomic density and field is π, as is the case for population inversion gratings in bidirectional ring lasers [16]. A shift of $\pi/2$, while more effective, as in photorefractive media, is not necessary for net gain.

As the system must conserve momentum, this physical process is direction selective and can only take place in the counterpropagating direction. In the copropagating direction, the atoms will only be accelerated, a longitudinal grating cannot be formed, and there is therefore no amplification of the probe. The behavior of the gain as a function of detuning between the laser frequency and the atomic resonance may be explained by the fact that the source of gain in our experiment is an atomic polarization term induced by the pump. Close to resonance, absorption is predominant for both directions of propagation of the probe beam. As detuning is increased, absorption vanishes and dispersion becomes predominant, giving rise to a nonzero polarization. The maximum gain is therefore to be expected at values for which the dispersion reaches a maximum (and absorption is negligible). The need for momentum conservation and the importance of atomic polarization also explain the experimental dependence on the atomic density.

The dependence of the *sign* of the detuning for which gain is observed on the beam waist position can be interpreted as follows. In addition to the collective effect that redistributes the atoms in a periodic structure, there are two forces that act on each single atom: one coming from radiation pressure and the other from the field gradient. In the parameter region for which these two forces have the same sign, their compounded effect "blows away" the atoms in the direction of propagation of the pump beam, and the collective process is not strong enough to initiate the spatial structuring. On the other hand, for detuning values for which the dipole force has opposite sign to the radiation pressure, there are regions in space where the two forces can compensate each other to a sufficient degree that the collective process can initiate. From the functional form of the dipole force [17],

$$F_{\mathrm{dip}} = -\frac{1}{4}\,\frac{\hbar\Delta}{\Delta^2 + (\Gamma/2)^2 + (\Omega^2/2)}\,\nabla\Omega^2, \qquad (1)$$

one sees that its sign changes both with the sign of the detuning and with the relative position of the pump beam waist with respect to the interaction region. Thus, the compensation of the effects of radiation pressure can be achieved on the blue side of the resonance when the beam waist is placed *after* the oven center and on the red side in the opposite case. This prediction is in agreement with our observations.

In conclusion, we have presented experimental evidence of amplification of a frequency degenerate counterpropagating probe beam, in the presence of a strong pump, in a "hot" atomic vapor in the dispersive regime. No amplification is seen in the case of a copropagating probe, under the same experimental conditions. We propose an interpretation based on the spontaneous formation of a spatial density grating. A more detailed model explaining our observations will be presented elsewhere. However, our interpretation of the origin of gain in the counterpropagating direction and its dependence with frequency is well supported by the model of Ref. [11], which predicts and explains the spontaneous formation of a density grating.

We thank N. B. Abraham, R. Bonifacio, L. De Salvo, G. Giusfredi, G. Grynberg, L. M. Hoffer, L. M. Narducci, and P. Verkerk for stimulating discussions and G. Giusfredi, L. M. Hoffer, and H. Grassi for valuable technical consultations. All technical support has been provided by A. Roux.

[1] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions* (Wiley, New York, 1992).

[2] B. R. Mollow, Phys. Rev. A **5**, 2217 (1972); F. Y. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow, Phys. Rev. Lett. **38**, 1077 (1977).

[3] D. Grandclément, G. Grynberg, and M. Pinard, Phys. Rev. Lett. **59**, 40 (1987).

[4] G. Giusfredi (private communication).

[5] G. Khitrova, J. F. Valley, and H. M. Gibbs, Phys. Rev. Lett. **60**, 1126 (1988).

[6] L. M. Narducci, H. M. Doss, P. Ru, M. O. Scully, S. Y. Zhu, and C. Keitel, Opt. Commun. **81**, 379 (1991).

[7] J. Gao, C. Guo, X. Guo, G. Jin, P. Wang, J. Zhao, H. Zhang, Y. Jiang, D. Wang, and D. Jiang, Opt. Commun. **93**, 323 (1992).

[8] A. Nottelmann, C. Peters, and W. Lange, Phys. Rev. Lett. **70**, 1783 (1993).

[9] D. Marcuse, Proc. IEEE **51**, 849 (1963); L. A. Rivlin, Kvant. Electron. (Moscow) **19**, 513 (1992) [Sov. J. Quantum Electron. **22**, 471 (1992)]; J. Guo and P. R. Berman, Phys. Rev. A **47**, 4128 (1993); K. M. Gheri, H. Ritsch, D. F. Walls, and V. I. Balykin, Phys. Rev. Lett. **74**, 678 (1995).

[10] J.-Y. Courtois, G. Grynberg, B. Lounis, and P. Verkerk, Phys. Rev. Lett. **72**, 3017 (1994).

[11] R. Bonifacio, L. De Salvo, L. M. Narducci, and E. J. D'Angelo, Phys. Rev. A **50**, 1716 (1994).

[12] Cell and oven design courtesy of G. Giusfredi.

[13] N. Ioli, F. Strumia, and A. Moretti, J. Opt. Soc. Am. **61**, 1251 (1971).

[14] G. Ankerhold, M. Schiffer, D. Mutschall, T. Scholz, and W. Lange, Phys. Rev. A **48**, R4031 (1993).

[15] H. J. Eichler, P. Guenther, and D. W. Pohl, *Laser Induced Dynamic Gratings* (Springer, Berlin, 1985).

[16] H. Zeghlache, P. Mandel, N. B. Abraham, L. M. Hoffer, G. L. Lippi, and T. Mello, Phys. Rev. A **37**, 470 (1988).

[17] C. Cohen-Tannoudji, in *Systèmes Fondamentaux en Optique Quantique,* Proceedings of the Les Houches Summer School, Session LII, edited by J. Dalibard, J. M. Raymond, and J. Zinn-Justin (North-Holland, Amsterdam, 1992).