Observation of Continuous-Wave Self-Oscillation Due to Pressure-Induced Two-Wave Mixing in Sodium

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Collisional damping can induce a coupling between two nonresonant light beams propagating in an atomic vapor. This coupling is sufficiently important to induce a gain process for a weak probe beam interacting with an intense pump beam. We present here the first observation of a cw oscillation generated by this process using a cell which contains sodium and helium, the pump beam being slightly detuned from the D_2 resonance line. We also show how the self-oscillation due to four-wave mixing is modified by the presence of a buffer gas.

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The fact that collisional relaxation can strongly modify the nonlinear response of an atomic vapor and can induce new resonances in four-wave mixing has been theoretically predicted by Bloembergen at the very beginning of nonlinear optics.[1] However, the subject had not been studied experimentally before 1981 when Prior, Bogdan, Dagenais, and Bloembergen made the first experimental observation of these new resonances.[2] The puzzling aspect of a resonance created by a collision process has stimulated new theoretical developments to understand their origin.[3] New experimental schemes and new relaxation processes[4] have also been suggested and a large number of experimental results has now been obtained on these new resonances.[5] However, it is clear from the formula giving the third-order nonlinear susceptibility[1,6] that other nonlinear processes should be magnified in the presence of collisional damping.[7] For example, in the case of a single traveling beam, collision-aided self-focusing and self-defocusing has recently been observed.[8]

A more subtle and spectacular effect occurs when the interaction of two beams is considered. It can be shown theoretically that a relaxation phenomenon can induce a new type of coupling between two nonresonant light beams.[9,10] When relaxation is neglected, each beam propagates independently in the vapor, but when a buffer gas is introduced, a coupling between the two beams becomes possible and an energy transfer from one beam to the other can be predicted. This transfer can even be sufficiently important to induce a gain mechanism. This result may look strange since the first effect of collisions is to increase absorption. However, the pressure-induced coupling can overcome the absorption losses and a net gain is then expected. It is the aim of this paper to present the first experimental observation of this effect.[11] In fact, rather than looking at the pressure-induced gain, we have directly observed the self-oscillation induced by this gain process. The experiment is performed with use of a cell containing sodium and helium which is placed inside a ring cavity and which interacts with a nonresonant pump beam.

A second interest of this experiment is that it makes a link between the different behaviors preceedingly observed when a phase-conjugate medium is used as an amplifier in a cavity. In the case of photorefractive materials[12] it has been shown that the frequency of the oscillating beam is slightly different from the frequency of the pump beam, while in the case of pure sodium vapor the two frequencies exactly coincide.[13] We show here that a behavior similar to the one obtained with photorefractive materials is observed with a cell which contains sodium and buffer gas. The point is that in the absence of collisions, the upper level is only virtually excited, while in presence of collisional damping a real grating of excited-state atoms,[14] similar to the grating obtained in photorefractive materials, is created.

Let us consider two beams $\mathbf{E}_1$ and $\mathbf{E}_+$ of frequencies ω_1 and ω_+ which interact with a set of motionless two-level atoms of resonance frequency ω_0. We assume that $\mathbf{E}_1$ is the pump beam and that $\mathbf{E}_+$ is a probe beam which has a much weaker intensity. If we calculate the imaginary part χ'' of the susceptibility of the beam $\mathbf{E}_+$, we find at second order in the pump field,[10]

$$\chi''=\frac{Nd^2}{\epsilon_0\hbar\delta^2}\left[\left(\frac{\Gamma_e}{2}+\beta p\right)+\frac{\Omega_1^2}{\delta}\frac{\beta p(\omega_1-\omega_+)}{\Gamma_e^2+(\omega_1-\omega_+)^2}\right]. \tag{1}$$

N is the number of active atoms per unit of volume, d is the matrix element of the electric dipole moment between the ground and excited levels, $\delta=\omega_0-\omega_1$ is the frequency detuning from the resonance, Γ_e^{-1} is the lifetime of the excited level, p is the pressure of buffer gas, and β is the pressure broadening coefficient. Ω_1 is the resonance Rabi frequency ($\Omega_1=dE_1/\hbar$). We have assumed that $|\delta|\gg\Gamma_e$ and $|\delta|\gg|\omega_1-\omega_+|$. The first term corresponds to linear absorption. The following term depends on the pump-beam intensity and only ex-

ists in the presence of collisional damping ($\beta p \neq 0$). This term can be either positive or negative depending on the sign of $\omega_1 - \omega_+$. Let us point out that this second term can be larger than the linear absorption if $\Omega_1^2 > \Gamma_e |\delta|$.[15] If $(\omega_1 - \omega_+)/\delta$ is negative and if the second term of (1) is larger than the first one, a pressure-induced amplification of the probe beam is possible.

The amplification of $\mathbf{E}_+$ comes from the diffraction of $\mathbf{E}_1$ on the grating of excited-state atoms created by the beams $\mathbf{E}_1$ and $\mathbf{E}_+$.[10] The maximum amplification is achieved when the diffracted beam and $\mathbf{E}_+$ are in phase. This requires that the atomic grating is dephased with respect to the light grating. This occurs neither in the case $\omega_1 = \omega_+$ because the two gratings are then stationary nor in absence of collisions ($\beta p = 0$) because an excited atom spends a very short time in the excited state (virtual transition) and the atomic grating follows adiabatically the light grating. On the other hand, in the presence of collisions, the atom can be really excited through a collision-aided process and the atomic grating lasts a time of the order of Γ_e^{-1}.[14] In this case (and when $\omega_1 \neq \omega_+$) the atomic grating follows the light grating with a delay and amplification [when $(\omega_1 - \omega_+)/\delta < 0$] or extra absorption [when $(\omega_1 - \omega_+)/\delta > 0$] is possible.

The experimental setup is shown in Fig. 1. A cw dye laser pumped by an Ar^+-ion laser delivers about 300 mW at 5890 Å. The main part of this beam is focused into a quartz cell with Brewster angles containing sodium and a variable amount of helium. The sodium cell is placed inside a ring cavity which consists of four plane mirrors M_1, M_2, M_3, and M_4. A lens L with an antireflection coating on one side is placed between M_3 and M_4. The 4% reflection on the other side of the lens is used to obtain the output beam. The mirrors M_2, M_3, and M_4 are totally reflective, while M_1 has a transmission coefficient equal to 0.5%. When an oscillation occurs, the output through M_1 is used to stabilize the length of the cavity using a servoloop and a reference level which can be tuned. To determine the frequency spectrum of the output beam, we analyze with a Hewlett-Packard 8588B spectrum analyzer the beat frequency between the output beam coming from L and a reference beam taken from the pump beam with a plate P.

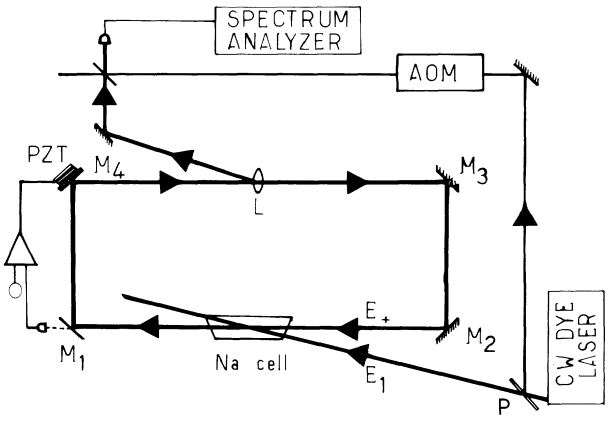

FIG. 1. Experimental setup for the two-wave mixing experiment. The pump beam $\mathbf{E}_1$ is a traveling wave. We use an acousto-optic modulator (AOM) to shift the frequency of the reference beam.

In the case of a pure sodium cell we have never observed any oscillation when the pump beam is a traveling wave. On the other hand, with cells containing 1.5 and 3 Torr of helium an unidirectional oscillation in the ring cavity is observed for temperatures of the cell larger than 160 °C. This oscillation is observed on both sides of the D_2 resonance: on the low-frequency side, at 2 GHz from the center of the absorption line and over a range of 1 GHz; on the high-frequency side, over a narrower range of frequency. The striking observation is that the frequency ω_+ of the oscillating beam differs from the frequency ω_1 of the pump beam. The difference $|\omega_+ - \omega_1|$ varies between 8 and 50 MHz, depending on the length of the cavity. We have determined the sign of $\omega_+ - \omega_1$ by shifting by 80 MHz the reference-beam frequency with an acousto-optic modulator and we have observed that the sign of $\omega_+ - \omega_1$ is reversed when the sign of the frequency detuning δ is changed. When the length of the ring cavity varies, both the output intensity I_{out} and the frequency difference $\omega_+ - \omega_1$ vary. We show in Fig. 2 the variation of I_{out} versus $\omega_+ - \omega_1$ for a cell containing 3 Torr of helium. We see that I_{out} is maximum for $\omega_+ - \omega_1 \cong 19$ MHz and that the value of I_{out} at the maximum is 1 mW.

All these results are in agreement with the predictions of formula (1). First, we note that the new gain mechanism only occurs in the presence of collisional damping. Second, we have verified that the sign of $\omega_+ - \omega_1$ changes when the sign of δ changes. Furthermore,

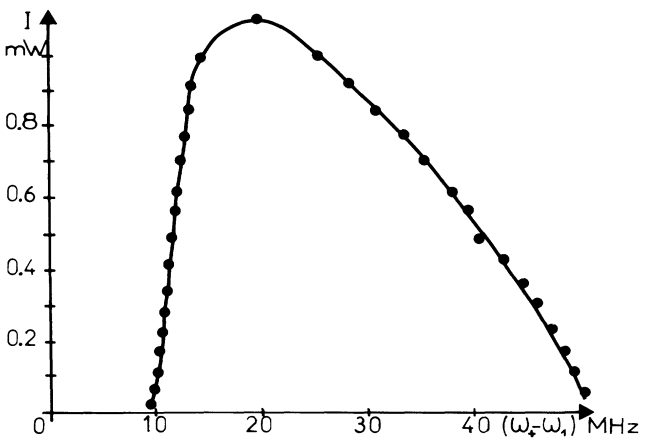

FIG. 2. Output intensity vs frequency of oscillation.

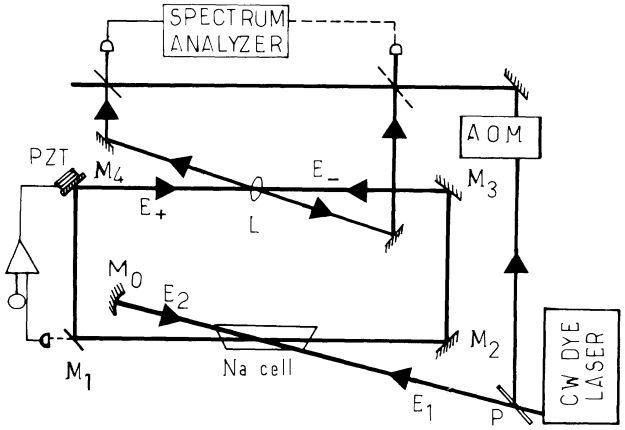

FIG. 3. Experimental setup for the four-wave mixing experiment. The pump beam $\mathbf{E}_1$ is now reflected on itself with use of the mirror M_0.

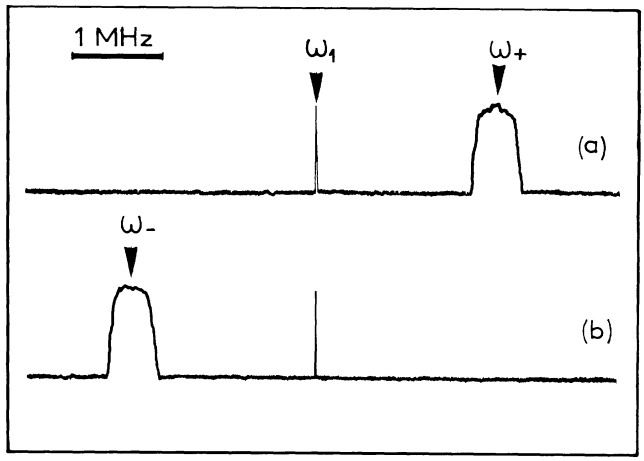

FIG. 4. Signal obtained on the spectrum analyzer for the two counterpropagating beams. The beat frequency between $\mathbf{E}_+$ and a reference beam whose frequency is shifted with use of an acousto-optic modulator is shown on curve (a). The result for $\mathbf{E}_-$ is shown on curve (b). The position of the pump-beam frequency ω_1 is determined by the narrow peak of noise.

amplification occurs when $(\omega_1-\omega_+)/\delta$ is negative in agreement with formula (1). Third, we remark that the dependence of I_{out} versus $\omega_+-\omega_1$ closely follows the variation of the gain with $\omega_+-\omega_1$. Formula (1) predicts that the gain should be maximum for $\omega_+-\omega_1=\Gamma_e$ whose value is 10 MHz in the sodium case. The fact that we obtain a slightly larger value for the maximum of I_{out} is mainly due to the residual Doppler effect. When the atoms are not stationary ω_1 and ω_+ are Doppler shifted. Since the oscillating beam makes an angle of about 10^{-2} rad with the pump beam, we expect to find a residual Doppler effect of about 10 MHz on $\omega_1-\omega_+$. The Doppler effect also explains why the oscillation is unidirectional. In the case of a probe beam propagating in a direction opposite to the pump beam, the averaging over the velocity distribution reduces the maximum value of the energy transfer between the two beams by a factor of the order of 100.[16]

In a second set of experiments, the pump beam is reflected on itself with use of a curve mirror M_0 (Fig. 3). In this case, we have two pump beams $\mathbf{E}_1$ and $\mathbf{E}_2$ propagating in opposite directions, and a new gain mechanism associated with the pair production of photons in four-wave mixing is possible.[13] Indeed, in the case of a pure sodium cell, an oscillation is observed when the temperature of the sodium cell is larger than 150 °C. The oscillation consists of two counterpropagating beams $\mathbf{E}_+$ and $\mathbf{E}_-$ of the same intensity and frequency. In particular, a fringe interference pattern is observed between any one of these beams and the reference beam, and we can assert that $|\omega_+-\omega_1|$ and $|\omega_--\omega_1|$ are less than 0.1 Hz. We have thus a perfectly degenerate oscillation.

If we repeat the experiment with the cell containing 3 Torr of helium, a completely different behavior is observed. The two beams $\mathbf{E}_+$ and $\mathbf{E}_-$ have different intensity and frequency. The intensity of $\mathbf{E}_+$ is twice the intensity of $\mathbf{E}_-$ and $\omega_+-\omega_1$ and $\omega_--\omega_1$ have opposite values. This point has been verified with use of a reference beam whose frequency is shifted by an acousto-optic modulator (Fig. 4). The value of $|\omega_+-\omega_1|$ varies between 2 and 5 MHz, depending on the length of the cavity. The asymmetry between the two counterpropagating waves comes from the fact that $I_2=|E_2|^2$ is smaller than $I_1=|E_1|^2$ by about 30% because of absorption losses in sodium and on the mirrors. A non-degenerate oscillation is thus observed because the pressure-induced gain on $\mathbf{E}_+$ is larger than the extra losses on $\mathbf{E}_-$.

In conclusion we have shown that self-oscillation in vapors can be generated by two types of processes. In the absence of collisional damping, the gain comes from a pair production of photon[13] and the device is similar to a doubly resonant parametric oscillator.[6] When collisional damping is important, a new type of gain mechanism becomes possible, which involves the creation of a real grating of excited-state atoms and the diffraction of the pump beam on this grating.

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[11]A two-wave mixing oscillation in sodium has been briefly reported in Ref. 10. This experiment was made with a pulsed laser and the experimental results were difficult to interpret. Several explanations for the oscillation have been suggested but it was not possible to choose among them nor to assert that the oscillation was due to a single cause.

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[15]This condition can be fulfilled in the perturbative limit ($\Omega_1 \ll |\delta|$) considered here, see Ref. 10.

[16]One must have in mind that a two-level model is a very crude approximation for sodium atoms. It is well known that the degeneracy in the ground state can play an important role in pressure-induced extra resonances in four-wave mixing (see Ref. 5). A similar result is also obtained here when one does a more precise calculation of the value of χ'' for the probe beam. The threshold condition is then modified. However, a pressure-induced contribution identical to the one of formula (1) is still present and permits one to explain the observations presented here.