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Two-wave mixing in sodium vapour

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Résumé. — Nous avons observé une oscillation unidirectionnelle dans une cavité en anneau quand une vapeur de sodium interagit avec un faisceau incident intense et non résonnant. Nous avons vérifié que la fréquence du faisceau oscillant est proche de celle du faisceau pompe. Nous proposons une interprétation théorique de l'origine du mélange à deux ondes dans les vapeurs atomiques. Nous montrons la nécessité d'un processus de relaxation et nous insistons sur l'étroite relation entre cet effet et les résonances induites par pression de Bloembergen.

Abstract. — We have observed a unidirectional oscillation in a ring cavity when a sodium atomic vapour interacts with a non resonant pump beam. We have checked that the frequency of the oscillating beam is almost the same as the frequency of the pump beam. We propose a theoretical interpretation of the origin of a two-wave mixing process in an atomic vapour. We show the necessity of a relaxation process and we emphasize the close relationship between this effect and the Bloembergen PIER 4 resonances.

In the domain of nearly degenerate four-wave mixing and optical phase conjugation, many results first obtained in photorefractive materials have been afterwards extended to other systems. For instance, the study of self oscillation of a cavity bounded by a phase conjugate mirror has been done in photorefractive materials [1-3] before similar studies were developed using atomic vapours [4] ⁽¹⁾. One of the most spectacular effect in photorefractive materials is the observation of two-wave mixing [6-9]. We show in this paper that a similar effect can be observed in an atomic sodium vapour. We first describe our experimental results and show that an oscillation due to two-wave mixing is observed. Afterwards we present a theoretical model and show that a relaxation process is necessary to obtain such an effect in an atomic vapour when the incident beam is not resonant. This is a new type of relaxation-assisted process in non-linear optics. There is a close relationship with the Bloembergen PIER 4 resonances [10-14] and many theoretical papers written on this subject [15-23] can also be extended to the process described in the present paper.

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⁽¹⁾ It can however be noticed that the first evidence of self oscillation has been obtained by Bloom *et al.* [5] using sodium vapour.

1. Experimental observation.

We first describe the observation of a laser oscillation in a two-wave mixing experiment in sodium vapour. The experimental set-up is shown in figure 1. The sodium cell is put inside an oven and its temperature is about 300 °C. The cell is put in the arm of a ring cavity which consists of two totally reflective mirrors M_1 and M_2 and a partially reflective mirror M_3 ($R = 0.92$). The atoms interact with an incident light beam E_1 whose direction makes a very small angle ($\theta \sim 10^{-2}$ rad) with the direction of the two mirrors M_1 and M_2 . The incident beam comes from a flashlamp-pump dye laser injection locked to a monomode c.w. dye laser [24]. The characteristics of this laser are : output (1 kW), pulse duration (1 μ s), linewidth (~ 10 MHz). The beam E_1 has a linear polarization and its frequency ω_1 is close to an atomic sodium resonance frequency ω_0 (we have done the experiment on the two transitions D_1 and D_2). The frequency detuning from the resonance $\omega_0 - \omega_1$ is of the order of 5 ± 1 GHz (the detuning is larger than the Doppler width and the experiment is done on the self-defocusing side of the resonance). For both transitions, a unidirectional oscillation is observed in the ring cavity (Fig. 2). The direction of propagation of the oscillating beam and of the pump beam are almost parallel in the cell. The oscillation lasts 100 ns, is located at the maximum of the pump beam intensity,

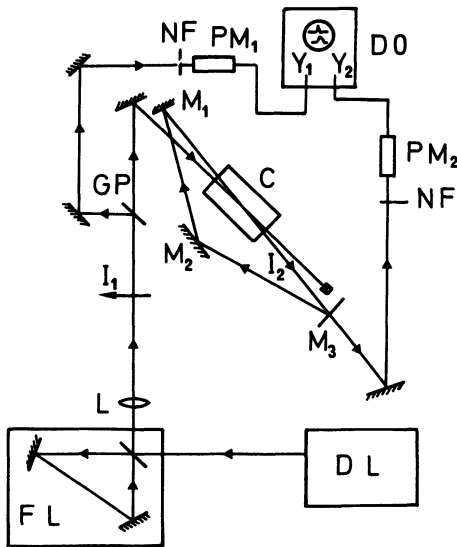


Fig. 1. — Schematic drawing of the experimental set-up. I_1 and I_2 are the intensities of the pump beam and of the clockwise propagating field in the cavity DL — single mode dye laser, FL — flashlamp pumped dye laser; L — lens; GP — glass plate; C — steel cell with sapphire windows containing sodium vapour at the temperature of 300 °C; M_1 — M_2 — totally reflecting mirrors; M_3 — partially reflecting mirror; NF — neutral filters; PM_1 — PM_2 — photomultipliers; DO — Digital oscilloscope.

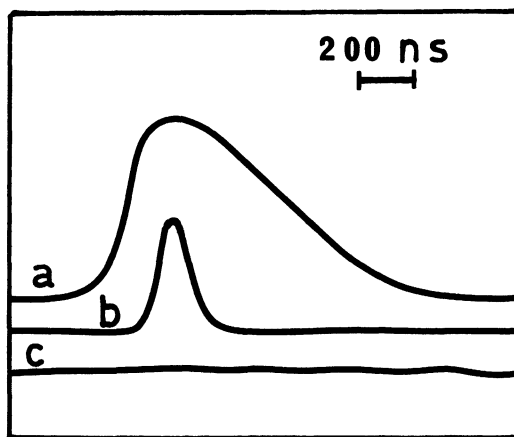


Fig. 2. — Experimental recording of the intensities. The upper trace (a) corresponds to the pump beam intensity I_1 ; the middle trace (b) corresponds to the oscillating beam intensity I_2 ; the lower trace (c) shows that no signal is observed in the counterpropagating direction.

and the power output transmitted through the mirror M_3 is 0.1 watt (similar intensities are observed on the D_1 and D_2 lines). The oscillation is observed on a frequency range of the order of 2 GHz. Using a 5 GHz free spectral range interferometer, we have checked that the frequency ω_2 of the oscillating beam is close to the frequency ω_1 of the pump beam (Fig. 3). In particular, the present experiment differs from the obser-

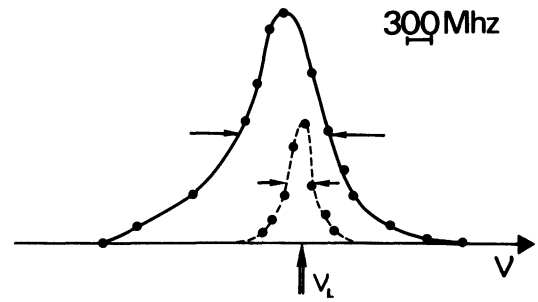


Fig. 3. — Spectral analysis of the oscillating beam. We have observed both the incident pump beam (dashed line) and the oscillating beam (solid line) through a 5 GHz F.S.R. interferometer which is pressure scanned. The instrumental bandwidth is 0.3 GHz and corresponds to the width of the dashed curve because the frequency dispersion of our flashlamp pump dye laser is of the order of 10 MHz.

vation of Kumar and Shapiro [25] who have observed stimulated Raman scattering on the same transition (in that experiment $\omega_1 - \omega_2 = 1.7$ GHz, which is the hyperfine splitting of the ground level). A possible explanation for the difference is that, in our pulse experiment, optical pumping does not last a sufficiently long time to obtain a population difference between the two hyperfine sublevels $F = 1$ and $F = 2$. The relatively large spectral width (~ 1 GHz) of the oscillating beam (Fig. 3) is probably related to phase-modulation [26] and will be discussed later on.

We now consider the origin of the two-wave mixing process. In the case of photorefractive crystals, the coupling between the two waves is related to the dephasing between the light grating and the photo-induced refractive index grating. We show thereafter that a similar interpretation can be developed in the case of an atomic vapour. However, in the non-resonant excitation case, a relaxation process is required to create the grating of excited atoms. We present here a steady-state theory and we shall mainly consider the case of collisional damping. As we mention it below, a steady-state theory cannot explain all the features of our pulse experiment. In a similar way, other relaxation mechanisms such as radiative relaxation [20-22] or phase fluctuation [23] could be considered in connection with our experiment. However, we feel that the simple model presented here gives some basic ideas on the physical mechanisms involved in a two-wave mixing process in an atomic vapour.

2. Collision-induced two-wave mixing in steady-state regime.

We consider a set of two-level atoms (ground level $|g\rangle$, excited level $|e\rangle$, energy difference $E_e - E_g = \hbar\omega_0$). The matrix element of the electric dipole moment between these two levels is d . The relaxation rate of the excited level and of the optical coherence are Γ_e and Γ_{eg} . In the case of collisional relaxation, we make the

impact approximation. Γ_{eg} is equal to

$$\Gamma_{eg} = \frac{\Gamma_e}{2} + (\beta + i\beta')p \quad (1)$$

where β and β' describe the broadening and shift of the optical transition and p is the gas pressure. This set of atoms interact with two electromagnetic waves polarized along the x axis. The wave \mathbf{E}_1 has a frequency ω_1 and propagates along the z direction. The wave \mathbf{E}_2 has a frequency ω_2 and propagates along a direction \mathbf{k}_2 which makes a small angle θ with Oz . The total electromagnetic field $\mathbf{E}(\mathbf{r}, t)$ is thus equal to :

$$\mathbf{E}(\mathbf{r}, t) = \text{Re} \{ \varepsilon_1 \exp - i(\omega_1 t - k_1 z) + \varepsilon_2 \exp - i(\omega_2 t - \mathbf{k}_2 \cdot \mathbf{r}) \} \mathbf{e}_x. \quad (2)$$

We assume that the frequency detunings $\delta_1 = \omega_0 - \omega_1$ and $\delta_2 = \omega_0 - \omega_2$ are much larger than the Doppler width ku and the homogeneous width Γ_{eg} . On the other hand, $|\delta_1 - \delta_2|$ is assumed to be a small quantity compared to $|\delta_1|$. The atomic polarization \mathbf{P} can be written as a function of the non-linear susceptibilities χ_1 and χ_2 :

$$\mathbf{P} = \text{Re} \{ \varepsilon_0 \chi_1 \varepsilon_1 \exp - i(\omega_1 t - k_1 z) + \varepsilon_0 \chi_2 \varepsilon_0 \exp - i(\omega_2 t - \mathbf{k}_2 \cdot \mathbf{r}) \} \mathbf{e}_x. \quad (3)$$

In the following, we shall always assume that the beam E_2 is much weaker than the beam E_1 . We thus calculate \mathbf{P} at first order in ε_2 . We consider the case where \mathbf{E}_1 does not saturate the atomic medium. The Rabi frequency $\Omega_1 = d\varepsilon_1/\hbar$ has then to be small compared to the frequency detuning $|\delta_1|$. The assumptions of our calculation are thus :

$$\Omega_2 \ll \Omega_1 \ll |\delta_1|, |\delta_2|. \quad (4)$$

We calculate χ_2 by solving the atomic density matrix equation in third order in electric field. We find

$$\chi_2 = \left(\frac{Nd^2}{\varepsilon_0 \hbar} \right) \frac{1}{\delta_2 - i\Gamma_{eg}} \left[1 - \frac{1}{2} \frac{\Omega_1^2}{\delta_1^2} \left(1 + \frac{2\beta p}{\Gamma_e} \right) - \frac{1}{2} \frac{\Omega_1^2}{\delta_1 \delta_2} \left(1 + \frac{2\beta p}{\Gamma_e + i(\omega_1 - \omega_2)} \right) \right]. \quad (5)$$

When performing this calculation, we have assumed that $|k_1 v_z - \mathbf{k}_2 \cdot \mathbf{v}| \ll \Gamma_e$. Let us consider the imaginary part χ_2'' of χ_2 . We find

$$\chi_2'' \simeq \frac{Nd^2}{\varepsilon_0 \hbar \delta_2^2} \left[\left(\frac{\Gamma_e}{2} + \beta p \right) + \frac{\Omega_1^2}{\delta_1} \frac{\beta p (\omega_1 - \omega_2)}{\Gamma_e^2 + (\omega_1 - \omega_2)^2} \right]. \quad (6)$$

The first term $\left(\frac{\Gamma_e}{2} + \beta p \right)$ corresponds to the linear absorption of the beam E_2 . The following term can be

either positive or negative depending on the sign of $\omega_1 - \omega_2$. However, this term is only present in the case of collisional damping ($\beta p \neq 0$). Let us now discuss the possibility of observing amplification of beam E_2 . This process can occur if the second term is negative, i.e. if $(\omega_1 - \omega_2)$ and δ_1 have opposite signs. Furthermore the second term must be larger than $(\Gamma_e/2 + \beta p)$. We must have

$$\frac{\Omega_1^2}{|\delta_1|} \frac{\beta p}{2\Gamma_e} > \frac{\Gamma_e}{2} + \beta p. \quad (7)$$

For a sufficiently high pressure ($\beta p > \Gamma_e$), we can neglect $\Gamma_e/2$ and (7) becomes :

$$(\Omega_1/|\delta_1|)^2 > 2(\Gamma_e/|\delta_1|). \quad (8)$$

This condition can be fulfilled even when $\Omega_1 < \delta_1$ because Γ_e is very small compared to $|\delta_1|$. A collision-induced two-wave mixing is thus possible in an atomic vapour. Let us note that a similar result has been mentioned by Berman *et al.* [27].

Let us now make some comments about this result. First, we notice that there is a strong connection with the pressure-induced extra resonances observed by Bloembergen *et al.* in nearly degenerate four-wave mixing [11]. The only difference comes from the fact that we have here two incident beams instead of three and that we consider the modification of one beam instead of looking at the generation of a fourth beam. Secondly we notice that this effect depends on the relative direction of the two beams. In particular a similar amplification would not occur if \mathbf{E}_1 and \mathbf{E}_2 have opposite directions because in that case the energy denominator of (5) would be $\Gamma_e + i(\omega_1 - \omega_2 + 2kv_z)$ and, after averaging on the velocity distribution, the second term is reduced by a factor of the order of Γ_e/ku . Thirdly, we can understand the necessity of collisions. The amplification of the beam \mathbf{E}_2 comes from the diffraction of the beam \mathbf{E}_1 on the population grating, the dephasing between the light grating and the population grating being adjusted to a value such as \mathbf{E}_2 and the diffracted beam are in phase. In the absence of collisions, the population grating and the light grating are in phase because the atom spends a very short time in the excited level (of the order of $1/|\delta_1|$). In the presence of collisions, the atom is really excited [19] and the population grating lasts a time of the order of Γ_e^{-1} . When $\omega_1 = \omega_2$, the two gratings are also in phase and neither amplification nor absorption due to the non-linear term occurs. On the other hand, when $(\omega_1 - \omega_2) \neq 0$, the two beams can exchange energy [27, 28], the maximum of absorption or amplification being obtained for $|\omega_1 - \omega_2| \sim \Gamma_e$. At last, we can note that the dependence of formula (6) upon $(\omega_1 - \omega_2)$ is the same as the one usually obtained in stimulated Rayleigh scattering [26, 29]. We can thus also describe the present effect as a collision-induced stimulated Rayleigh scattering.

3. Discussion.

It is well known that, in four-wave mixing, extra-resonances can also be generated by other relaxation processes such as spontaneous emission [20-22] or phase fluctuation [23]. For example, if we calculate the non-linear susceptibilities at fifth order, we obtain a similar dependence of $\chi_2^{(5)}$ upon $(\omega_1 - \omega_2)$ even without collisions [27] (the population grating is created by a three-photon scattering process [30, 31]). In a pure radiative case $\Gamma_{eg} = \Gamma_e/2$, we obtain

$$\chi_2^{(5)} = \frac{Nd^2}{\epsilon_0} \frac{\Omega_1^4}{\delta_2} \frac{\Gamma_e(\omega_1 - \omega_2)}{4\delta_1^4 \Gamma_e^2 + (\omega_1 - \omega_2)^2}. \quad (9)$$

It shows that $\chi_2^{(5)}$ can be either positive or negative depending on the sign of $(\omega_1 - \omega_2)$. An amplification process is possible if the non-linear amplification is larger than the linear losses, i.e. when :

$$\frac{\Omega_1^4}{4|\delta_1|^4} > \frac{\Gamma_e}{|\delta_1|}. \quad (10)$$

As a matter of fact, a calculation exact to all orders in $\Omega_1/|\delta_1|$ can be analytically performed and shows that a radiative-relaxation induced amplification is possible when $\Gamma_e/|\delta_1| \ll 1$. In fact, this point corresponds to an earlier observation of Wu *et al.* [32]. In an experiment done on a sodium atomic beam, they observe (see Fig. 4a of this reference) a very small amplification for $|\omega_1 - \omega_2| \sim \Gamma_e$ in the non resonant case. This amplification also appears in their theoretical curves (Fig. 4b) and is implicitly present in a preceding paper [33]. However this amplification and its origin were not discussed in these papers.

In our experiment, we have $\Omega_1/|\delta_1| \sim 0.5$ at the maximum of the pulse intensity (while $\Gamma_e/|\delta_1| \sim 2 \times 10^{-3}$). We have also $\beta p/\Gamma_e \gtrsim 1$ if we only take into account the sodium-sodium collisions [34]. Both conditions (7) and (10) are thus fulfilled in our experimental situation and it is not possible to precisely specify a single relaxation process in our case. Furthermore, the steady state analysis presented here neglects the effect of phase modulation induced by the intense pump beam. Indeed, as other preceding authors [35],

we have observed an important self-phase modulation on the pump beams. We feel that the width of the oscillating beam (Fig. 3) is related to this effect ⁽²⁾. A complete theoretical analysis of our experiment would require to take into account those transient effects. We feel that it should be easier to precisely check the present theory on a c.w. experiment.

4. Conclusion.

In conclusion, we have observed a two-wave mixing process in a sodium vapour. We have theoretically studied two-wave mixing in the non-resonant case in steady-state operation and we have shown that this process should be relaxation-assisted. We have considered here the case of a two-level atom to explain our nearly degenerate two-wave mixing experiment. However, similar processes also occur in three-level systems. For instance, if we consider two excited level e and e' , a collision assisted energy transfer can be predicted for a probe beam of frequency ω' if the medium interacts with a pump beam of frequency ω when $\hbar(\omega' - \omega) = (E_{e'} - E_e) \pm \hbar\Gamma$ (the sign + or - depending on the sign of the detuning from the resonance). There is thus a profound relationship between these effects and the Bloembergen PIER 4 resonances since they occur on the same systems in similar conditions.

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⁽²⁾ A first analysis of the shift of the curve of figure 3 seems to be in disagreement with the prediction of (6) and (9). The maximum of the signal appears at a frequency lower than ω_L while we expect that amplification occurs for $\omega_2 - \omega_1 > 0$. In fact, the analysis of the frequency spectrum of the pump wave after transmission through the sodium cell shows that its frequency shift towards lower frequencies is larger than the frequency shift of the signal. The condition $(\omega_2 - \omega_1) > 0$ is fulfilled but ω_1 differs from the incident laser frequency ω_L .

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