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Heterodyne detection of phase-conjugate emission in an Ar discharge with a low-power c.w. laser (*)

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Résumé. — Nous décrivons une expérience de mélange à quatre ondes (quasi dégénéré), réalisée dans une décharge d'argon avec un laser continu de faible puissance, à 867 nm (transition $1s_3-2p_7$). L'émission conjuguée, de faible intensité, a été détectée par une technique de battement hétérodyne à 30 MHz. Ce genre de montage permet en principe d'atteindre la sensibilité ultime liée au bruit de photons et se révèle donc particulièrement adapté aux études sur la conjugaison de phase requérant une grande sensibilité.

Abstract. — We report the observation of nearly degenerate four-wave mixing in Ar gas discharges at 867 nm (transition $1s_3-2p_7$) using a low power c.w. laser. The weak phase-conjugate emission has been observed by means of a heterodyne detection technique at 30 MHz. This experimental scheme should be able to yield ultimate shot-noise limited signals, and thus could be used as a sensitive tool for phase-conjugation studies.

The emission of a phase-conjugate (PC) field by four-wave mixing (FWM) in a nonlinear medium has been recently the subject of many investigations, especially because of the optical properties of the emitted beam [1]. The possibility of enhancement of the nonlinearity in a gas medium by tuning the laser frequency to an atomic resonance was demonstrated early [2]. The theory of FWM in gases is complicated by the atomic motion and because the saturation is easily reached for the most efficient systems. A simple way to check these theories — suppression of Doppler-broadening [3-5], angular dependence of the reflection coefficient [3-5], asymmetry of the lineshape for a two-photon transition [6] — is to use a c.w. single mode laser, rather than a pulsed one. However, the high-order nonlinear process in FWM gives generally weak emissions with low power lasers, and so, very few experiments have been performed. Until now, the only reported experiments have been carried out on the resonance lines of the Na vapour [3, 7]. Liao *et al.* [3] observed the PC emission for the D_2 hyperfine components in which the population of the ground state was not depleted by hyperfine optical pumping. In a subsequent experiment, they increased the sensitivity by a modulation of the PC

emission, but still on the most favourable component.

This letter reports on some preliminary experiments performed in Ar discharges with a c.w. laser. The basic scheme is the — nearly degenerate — FWM with two incident frequencies, ω for the pump, $\omega + \delta$ for the *object* — The *object* beam is sent back on itself and the PC emission at $\omega - \delta$, which propagates opposite to the incident object wave, is detected as an heterodyne beat signal at frequency 2δ as shown in figure 1. The features of the PC emission (without wave 4) have already been discussed in several papers [3-5]. It is now well known that the most intense emission is obtained for a small angle between pump

Fig. 1. — Basic scheme for heterodyne detection of four-wave mixing. Wave at ω is the pump, wave at $\omega + \delta$ is the object.

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and object beams, with a frequency-dependence limited by the homogeneous linewidth. If the intensity of the return *object* beam is not strong enough to saturate the atomic transition, the amplitude of the beat at 2δ just depends on the three *usual* waves — 1, 2, 3 — : because of momentum and energy conservation, emission at $\omega - \delta$ cannot be generated in direction $-\mathbf{k}$ as a result of a three-wave interaction between 1, 2 and 4. Hence, the observed beat at 2δ cannot be interpreted as a modulated absorption effect, and the beat intensity $I(2\delta)$ is the product :

$$I(2\delta) \propto A_4 A_{PC}^* e^{i2\delta t} + \text{c.c.}$$

where A_n is the complex amplitude for the wave n , and where

$$A_{PC} = \chi^{(3)} A_1 A_2 A_3^*$$

is the amplitude of the PC beam. Wave 4 simply acts as a local oscillator, and the signal is increased by heterodyne amplification to the detection level required to be beyond the background noise — stray light, electronics noise... —

We have performed our experiment on the $1s_3$ ($J=0$)- $2p_7$ ($J=1$) line of argon, at $\lambda = 866.8$ nm. The $1s_3$ level is metastable, with lifetime of about 10^{-4} s in the experimental conditions; the level is populated by a d.c. discharge (10-30 mA) in a 20 cm long, 2 cm diameter cell. The laser source was a single mode dye (HITC) laser — commercial type CR 599-21 — able to deliver up to 30 mW at $\lambda = 867$ nm, and pumped by a Kr^+ laser. The $1s_3$ - $2p_7$ transition is rather intense, and a 90 % absorption was observed with a few hundred of millitorrs and low laser intensities. The saturation was reached around 1 mW/mm^2 . It is particularly worth noticing that once excited in the $2p_7$ level, the atom will generally not decay to the metastable $1s_3$, as it is shown by the following transition probabilities [8] :

$2p_7 \rightarrow 1s_5$	$\lambda = 772.4 \text{ nm}$	$\gamma = 0.9 \text{ MHz}$
$\rightarrow 1s_4$	$\lambda = 810.4 \text{ nm}$	$\gamma = 4.4 \text{ MHz}$
$\rightarrow 1s_3$	$\lambda = 866.8 \text{ nm}$	$\gamma = 0.4 \text{ MHz}$
$\rightarrow 1s_2$	$\lambda = 935.4 \text{ nm}$	$\gamma = 0.2 \text{ MHz}$

A scheme of the experimental set-up is presented in figure 2. The two frequencies ω and $\omega + \delta$ are generated by an acousto-optic modulator (AOM) — commercial device, SORO IM 20, crystal of PbMoO_4 —. This modulator is excited by two RF frequencies $\Delta \pm \frac{\delta}{2}$ with $\Delta = 70 \text{ MHz}$ and $\delta = 15 \text{ MHz}$, generated by a double side band technique. The theory of two-frequency-excited AOM has been studied by Hécht [9]. The lens L_1 reduces the beam divergence within the AOM. The pair of deflected beams, which

Fig. 2. — Schematic of the experimental configuration. Angular deflections are strongly exaggerated.

are shifted 62.5 MHz and 77.5 MHz from the laser frequency, are selected by diaphragm D_1 and focussed into the argon cell by lens L_2 . The reflection of the incident beams by the phase-matched mirror M creates a pair of standing wave fields within the argon cell crossing at an angle determined by L_2 .

— Practically, we used a magnification $\Gamma \simeq 1$, and then a $3 \sim 4$ mrad. crossing angle in the cell —. Due to the quarter-wave plate, the two return beams are reflected by the Glan prism G [10]. The desired *object* beam is then selected by the diaphragm D_2 and sent onto a fast photodiode PD followed by a 30 MHz heterodyne receiver. Since the output signal is proportional to the electric power modulated at 30 MHz, the detection system is sensitive to $|I(2\delta)|^2$.

Figure 3 shows the signal as a function of the laser frequency. As predicted, the resonance linewidth is much smaller than the Doppler-broadening (see Fig. 3 : linewidth $\sim 70 \text{ MHz}$, estimated Doppler width $\sim 400 \text{ MHz}$). However, one observes an important line asymmetry which could be a serious limita-

Fig. 3. — Beat intensity when the laser frequency is tuned to the $1s_3$ - $2p_7$ transition of argon, with 300 mtorrs of Ar and $P_{\text{laser}} \simeq 1 \text{ mW}$, a) detection on the object beam shifted by $\Delta + \frac{\delta}{2} = 77.5 \text{ MHz}$, with pump shifted by $\Delta - \frac{\delta}{2} = 62.5 \text{ MHz}$; b) detection on the object beam at $\Delta - \frac{\delta}{2} = 62.5 \text{ MHz}$, with pump shifted by $\Delta + \frac{\delta}{2} = 77.5 \text{ MHz}$. This situation is the one represented on figure 2.

tion for spectroscopic purposes. Its origin lies in a residual overlap of the two deflected beams which are not entirely separated at the AOM output. Since the distinction between *pump* and *object* in figure 1 is quite arbitrary, a 2δ beat is also observed on the beam at frequency ω , and the theory [5] shows that the centre of this resonance is shifted by $2\delta = 30$ MHz. A residual overlap of *pump* and *object* beams in the detector will then introduce a second small but shifted resonance, and so induces an asymmetry in the lineshape. Our interpretation is justified by the comparison between figure 3a and figure 3b, where the detected beam has been changed: the resonance is shifted by 30 MHz, while the asymmetry changes its side; the relative position of the *stray* resonance which is responsible for the asymmetry, is modified by exchanging pump and object [11].

Since our receiver was not calibrated on an absolute scale, it was difficult to assess the sensitivity of this method of detecting PC emission [12]. However, it was surely very high compared to the effective signal-to-noise ratio generally obtained in FWM on *zero background* — note that the ultimate (shot-noise limited) sensitivity for both methods is the same —.

There are several reasons for the reduced emission efficiency in comparison with the Na resonance line [3]:

- (i) Weak metastable population (absorption measurements give about 10^{10} atoms/cm³).
- (ii) Depletion of the metastable state by optical pumping (as discussed above).
- (iii) Weaker transition probability.
- (iv) High value of the detuning δ , compared to the inverse lifetime γ_M of the metastable state $1s_3$: in the limit of quasi-collinear configuration, the emitted field amplitude is proportional to $1/\delta$ for $\delta \gg \gamma_M$ [5, 13, 14].
- (v) Linear absorption in the medium, and possible effects of saturation.

In another paper [15], we have demonstrated that for a sufficiently high beat frequency (typically 2 MHz for a laser power of 1 mW), the main noise source becomes the shot-noise (SN) of the detected beam, and it is then feasible to reach the ultimate SN-limited sensitivity for the detection of the PC signal. With some improvements [11], our heterodyne scheme should be able to provide this ultimate sensitivity. One should also note that the SN-limit could also be obtained by an attenuation of the local oscillator (L.O.) as in polarization spectroscopy [16]. The noise contribution of the L.O. decreases faster with attenuation than the heterodyne signal. This is one of the main differences with the collinear FWM (heterodyne saturation spectroscopy [14]) where the emitted field itself is affected by a reduction of the L.O. (because $|A_{L.O.}| = A_2$). Such a technique of reduction of the L.O. level without degrading the signal-to-noise ratio could also be of interest when one needs to use laser powers larger than the saturation thresholds of photodiodes (e.g. in the case of two-photon transitions): it is then an alternative to polarization spectroscopy, which permits optical isolation.

The collinear configuration is preferable for spectroscopic purposes, because there is no residual Doppler-broadening, no variation of the resonance centre with the angle, and more complete calculations are possible [14-15, 17]. But much fundamental research on the nonlinear processes involved in FWM in gases is yet to be done, as indicated in the introduction. Our heterodyne scheme should turn out to be valuable for such studies, owing to the potentially shot-noise-limited sensitivity. An important improvement can be brought by the use of a phase-sensitive detector: as the detection system is sensitive to the amplitude of the PC wave, interference effects between signal and background are suppressed and it becomes also possible to discriminate between real and imaginary parts of the nonlinear susceptibility.

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- [11] That asymmetry should be eliminated by increasing δ and thus the angular separation between the two output beams of the AO modulator. Another possibility would be to use two single-frequency-excited AOM's, to provide two independent beams.
Another cause of asymmetry is connected to the presence of a small coherent background at frequency 2δ . Thus our power receiver yields a dispersion-shaped interference term which contributes to the signal asymmetry. This contribution should be eliminated by the use of a phase-sensitive detector.
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