Redistribution of Photons and Frequency Mixing with Cross-Polarized Beams in Sodium

G. Grynberg, M. Vallet, and M. Pinard
Laboratoire de Spectroscopie Hertzienne de l'Ecole Normale Supérieure, Université Pierre et Marie Curie, 75252 Paris CEDEX 05, France
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The interaction of two linearly cross-polarized beams having frequencies ω_1 and ω_2 with sodium atoms leads to the generation of new waves having frequencies $(2p+1)\omega_1-2p\omega_2$. The intensities of these waves are well explained by a simple model based on self-induced optical activity. The relation with the atomic cooling in a polarization gradient and with the extra resonances in multiwave mixing is pointed out.

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When two electromagnetic fields of frequencies ω_1 and ω_2 propagate in matter, the nonlinear response of the medium induces the generation of waves having new frequencies.[1] The studies are generally done in situations where the conversion efficiency is small. On the other hand, in the case of high conversion efficiency, there are only a few examples where a complete analytical theory has been obtained.[2] We show here that for two linearly cross-polarized fields nearly resonant with an atomic transition, the intensities of the generated waves can be analytically calculated even for very high values of the conversion efficiency.[3] The method presented here gathers ideas coming from nonlinear optics and from optical pumping and leads to great simplifications in the case of cross-polarized beams. We compare the predictions of this model with experimental results obtained in sodium and we discuss the implication of these results for other topics. In particular, we show that the photons are redistributed between the two cross-polarized beams in a way that is reminiscent of the ideas found in atomic cooling.[4]

Let us consider a sample containing atoms having a $J_g=\frac{1}{2}$ ground state. These atoms interact with two incident linearly polarized fields $E_1\mathbf{e}_x\cos(\omega_1 t-kz)$ and $E_2\mathbf{e}_y\cos(\omega_2 t-kz)$. For the sake of simplicity, we consider a $J_g=\frac{1}{2}\rightarrow J_e=\frac{1}{2}$ transition and we call $\Delta=\omega_1-\omega_{eg}$ the detuning from resonance. This detuning is assumed to be very large compared to the natural width Γ and to the Doppler width of the transition and also compared to the frequency difference $\delta=\omega_1-\omega_2$ ($|\Delta|\gg|\delta|$). The polarization of the field in the cell depends on the value of δt and is periodically σ^+ circularly polarized, linearly polarized, σ^- circularly polarized, etc. When the field is circularly polarized the atoms are optically pumped from one Zeeman sublevel to the other. This transfer of population creates a huge nonlinearity that has been used in several experiments.[5] The difference of population between the Zeeman sublevels creates an orientation in the medium which induces an optical activity for the linearly polarized incident beams.[6] At the exit of the cell, the components E_x and E_y of the field are (apart from a common phase factor)

$$\begin{pmatrix}E_x\\E_y\end{pmatrix}=\begin{pmatrix}\cos\theta & -\sin\theta\\ \sin\theta & \cos\theta\end{pmatrix}\begin{pmatrix}E_1\cos\omega_1 t\\E_2\cos\omega_2 t\end{pmatrix}, \tag{1}$$

where θ is the rotation angle equal to $k\chi_0 l\langle J_z\rangle$ (where k is the light wave vector, χ_0 is the linear susceptibility equal to $-Nd_{+}^2/\epsilon_0\hbar\Delta$, l is the length of the sample, and $\langle J_z\rangle$ is the mean value of the ground-state electronic angular momentum[7]). The evolution of $\langle J_z\rangle$ is governed by an equilibrium between the time-modulated optical pumping and the relaxation:

$$\frac{d}{dt}\langle J_z\rangle=-\gamma_2\langle J_z\rangle+\gamma_1\sin\delta t\,, \tag{2}$$

where $\gamma_1=\Gamma(d_{+}^2E_1E_2/6\hbar^2\Delta^2)$ and $\gamma_2=\gamma+\Gamma[d_{+}^2(E_1^2+E_2^2)/6\hbar^2\Delta^2]$. γ_2 is the total relaxation rate of the ground state. It is the sum of the intrinsic relaxation rate γ and of the relaxation rate due to the pumping mechanism. Equation (2) is valid if $\gamma_2\ll\Gamma$, a condition which is assumed to be fulfilled in the following. By solving (2), we find that the rotation angle θ is

$$\theta=\frac{b}{1+u^2}(\sin\delta t-u\cos\delta t)\,, \tag{3}$$

where $b=k\chi_0 l(\gamma_1/\gamma_2)$ and $u=\delta/\gamma_2$. The rotation angle thus follows the variation of the optical pumping with a time lag associated with the finite response time of the ground state.

If we insert solution (3) into Eq. (1), we find that the component of the field polarized along the x direction is a sum of exponentials having angular frequencies $(2p+1)\omega_1-2p\omega_2$, where p is an integer which can be positive, negative, or zero. The intensity I_p of the harmonic $(2p+1)\omega_1-2p\omega_2$ is

$$I_p=E_1^2J_{2p}^2\left(\frac{b}{(1+u^2)^{1/2}}\right)+E_2^2J_{2p+1}^2\left(\frac{b}{(1+u^2)^{1/2}}\right)+2E_1E_2\frac{u}{(1+u^2)^{1/2}}J_{2p}\left(\frac{b}{(1+u^2)^{1/2}}\right)J_{2p+1}\left(\frac{b}{(1+u^2)^{1/2}}\right), \tag{4}$$

where $J_p(x)$ is the Bessel function of order p. This formula is valid for any conversion efficiency and can be used as long as the absorption is negligible. It is a nonperturbative formula which includes the possible saturation of the optical pumping. The perturbative limit ($E_1, E_2 \to 0$) and the limit of a thin medium ($l \to 0$) both correspond to $b \to 0$. In this limit, we have the following inequality $I_0 > I_{-1} > I_1 > I_{-2} > I_2$, etc. Let us note that I_0 corresponds to the intensity of the transmitted beam of frequency ω_1 and I_{-1} corresponds to the four-wave-mixing generation which has already been studied for small values of b.[8]

In order to verify the predictions of this model, we have performed an experiment in sodium vapor. Two beams coming from a cw dye laser of frequency ω have their frequencies shifted by two acousto-optic modulators (Fig. 1). The first shift is constant and gives a beam of frequency $\omega_1 = \omega + \omega_s$. The second shift can be scanned and gives a beam of frequency $\omega_2 = \omega + (\omega_s - \delta)$. These two beams which are cross polarized are recombined in a Glan prism and sent into a Corning 1720 glass cell containing sodium. The temperature of the cell is 215 °C. The cell is placed between three pairs of Helmholtz coils which are used to cancel any stray magnetic field. The beam at the exit of the sodium cell is divided into its two polarization components using a second Glan prism. With a spectrum analyzer we study the beat frequency between the component polarized along the x direction and an auxiliary beam of intensity I and frequency ω coming directly from the laser (Fig. 1).

An example of a spectrum obtained for $\delta = -100$ kHz is shown in Fig. 2(a). One clearly sees waves having frequencies up to $9\omega_1 - 8\omega_2$ (H_4) on one side and up to $8\omega_2 - 7\omega_1$ (H_{-4}) on the other side. Using an expanded scale, we have observed all the harmonics up to $20\omega_1 - 19\omega_2$. The high efficiency of the multiwave-mixing process is clearly apparent here since the amplitude of the $4\omega_2 - 3\omega_1$ harmonic (H_{-2}) is larger than the amplitude of the four-wave-mixing generation at $2\omega_2 - \omega_1$ (H_{-1}). We note also that the beam of frequency $3\omega_1 - 2\omega_2$ (H_1) is larger than the transmitted beam of frequency ω_1 since the value of H_0 recorded in Fig. 2(a) corresponds to the addition of the signal and of the parasitic electronic noise at the acousto-optic modulator frequency. Note that because of our detection scheme, the peak of H_p is proportional to $\sqrt{I I_p}$. The theoretical amplitudes deduced from formula (4) are shown in Fig. 2(b). The agreement is qualitatively good and the hierarchy of the various harmonics are the same ($H_1 > H_{-2} > H_{-1} > H_2 > H_{-3}$). We have also shown in Fig. 2(c) the variation of $\sqrt{I_{-1}}$ and $\sqrt{I_{-2}}$ vs δ. Here again, the agreement between the experimental points and the theoretical curve deduced from formula (4) is satisfactory. Note that a very good quantitative agreement is not expected since some important effects such as the transverse distribution of intensity, the velocity distribution, and the hyperfine interaction in the ground state have not been included in the theory. It can also be noticed that all these experiments have been done in zero magnetic field. The addition of a transverse magnetic field of the order of 10 G strongly reduces the efficiency of the frequency-mixing process. The variation of the multiwave-mixing generation versus the magnetic field will be presented in a more complete paper.

Another interesting signal is the total intensity along the x axis. The summation of the intensities of all the

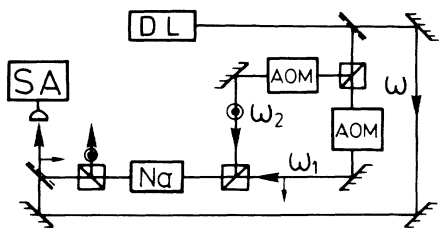

FIG. 1. Scheme of the experimental setup.

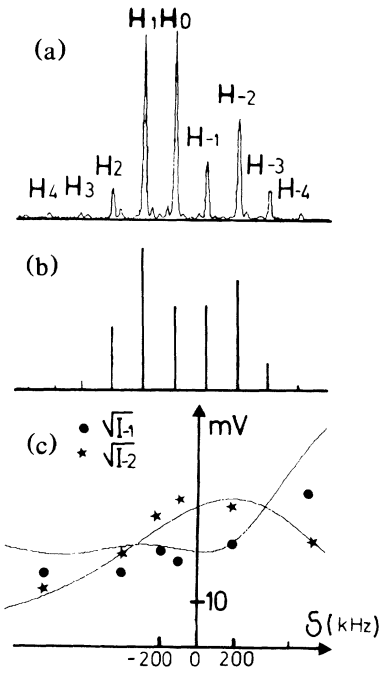

FIG. 2. Amplitudes of the $(2p+1)\omega_1 - 2p\omega_2$ harmonics (H_p). (a) Waves generated by the nonlinear interaction. The picture of the spectrum analyzer has been recorded with a linear scale. It has been obtained for a detuning $\Delta = 3(\pm 0.3)$ GHz from the center of the D_1 resonance line. The value of $\delta = \omega_1 - \omega_2$ is -100 kHz. The experiment has been done with two beams of intensity $P_1 = 50(\pm 10)$ mW and $P_2 = 70(\pm 10)$ mW focused on a spot of radius $w = 130(\pm 20)$ μm. (b) Theoretical values of the amplitudes calculated using formula (4) with $b = 3.9$ and $E_2^2/E_1^2 = 1.4$. (c) Variation of the amplitudes of the waves of frequency $(2\omega_2 - \omega_1)$ and $(4\omega_2 - 3\omega_1)$ vs δ. The theoretical curves are deduced from formula (4).

harmonics gives

$$I_x = \sum_{p=-\infty}^{+\infty} I_p = \frac{E_1^2+E_2^2}{2} + \frac{E_1^2-E_2^2}{2} J_0\left(\frac{2b}{(1+u^2)^{1/2}}\right) + E_1E_2 \frac{u}{(1+u^2)^{1/2}} J_1\left(\frac{2b}{(1+u^2)^{1/2}}\right). \quad (5)$$

In particular, in the limit of small values of b, this leads to the dispersive shape of two-wave mixing[9]

$$I_x = E_1^2 + bE_1E_2 \frac{u}{1+u^2}. \quad (6)$$

The second term of this formula is associated with a third-order nonlinearity since b in proportional to E_1E_2 at low-field intensity. An experimental recording of I_x and I_y vs δ is shown in Fig. 3. The curve is well described by formula (6) and, in particular, its extrema occur at $|\delta| \simeq 0.3$ MHz which is a typical value for the width of the ground state.[10] This behavior is very different from the one observed earlier with beams of the same polarization where the extrema occur for larger values of $|\delta| \sim \Gamma = 10$ MHz.[9]

One of the interests of this experiment is its connection with the results obtained for atomic cooling in a polarization gradient.[11] In these experiments, the atoms move in a field distribution created by two counterpropagating linearly cross-polarized waves of equal frequency. During its motion the atom interacts with a field whose polarization is periodically σ^+, linear, σ^-, etc. In its own frame, the atom thus samples exactly the same field that is applied to the atoms in our experiment (the Doppler shift replaces the applied frequency difference δ of our experiment). The cooling mechanism is generally described by a redistribution of photons between the two counterpropagating waves. Indeed, the experimental recording of Fig. 3 shows this redistribution of photons.[12]

Another interest of this experiment is that it verifies the possibility of observing extra resonances in multiwave mixing without collisional relaxation for an atom having a level degeneracy in the ground state.[8,13] This is exactly what is observed in Fig. 3 since the experiment is done without a buffer gas and at a low sodium density where self-broadening is negligible.

In conclusion, we have presented a method for studying the propagation of two linearly cross-polarized beams nearly resonant with an atomic transition. The preceding analysis has been done for a $J_g = \frac{1}{2} \rightarrow J_e = \frac{1}{2}$ transition but the extension to a $J_g = \frac{1}{2} \rightarrow J_e = \frac{3}{2}$ transition is straightforward. We have also shown that this problem has several important connections with subjects like atomic cooling or extra resonances. Finally, it is important to note that the simple approach presented here can be applied to many problems in nonlinear optics involving the propagation of cross-polarized beams or the apparition of polarization instabilities.[5]

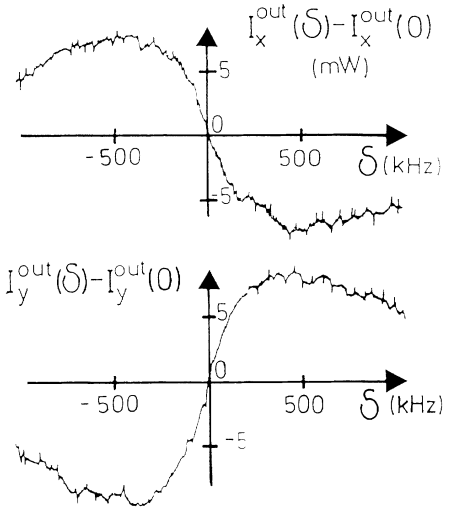

FIG. 3. Total intensity along the x polarization and the y polarization vs δ. The redistribution of photons between the two beams is apparent on these recordings. These curves have been obtained for a detuning $\Delta = 4.5(\pm 0.3)$ GHz from the center of the D_1 resonance line. The experiment has been done with two beams of intensity $P_1 = P_2 = 110(\pm 10)$ mW focused on a spot of radius $w = 400(\pm 20)$ μm. Because of the absorption in the cell, the maximum value of $I_x^{out}(\delta)$ is smaller than E_1^2.

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[2]A classical example is the second-harmonic generation (see Ref. 1, Sec. 7.1). Other examples deal with the competition between third-harmonic generation and multiphoton ionization [J. C. Miller, R. N. Compton, M. G. Payne, and W. W. Garrett, Phys. Rev. Lett. **45**, 114 (1980); D. J. Jackson and J. J. Wynne, Phys. Rev. Lett. **49**, 543 (1982)] or between four-wave mixing and amplified spontaneous emission [M. S. Malcuit, D. J. Gauthier, and R. W. Boyd, Phys. Rev. Lett. **55**, 1086 (1985)].

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