Nonlinear Electromagnetism
Tutorial n°5
Two-photon Absorption

We study the interaction between a wave at ω and a nonlinear medium with two energy levels $|a\rangle$ (lower energy state) and $|b\rangle$, such as their energy difference is equal to 2ω. The wave at ω is characterized by an electromagnetic mode with a wave vector $\boldsymbol{k}$, a polarization vector $\boldsymbol{e}$, and a number of photons N. In the following, we seek to determine the variation of the number of photons N that occurs during the propagation of the beam through this nonlinear material. Subsequently, we suppose that the population on the level $|b\rangle$ is initially equal to zero and that it does not exist any one-photon transition.

The quantum approach followed in this exercise consists in calculating the transition rate between an initial composite state $|i\rangle$ to a final state $|f\rangle$:

$$\frac{dP_{i\to f}}{dt} = \frac{2\pi}{\hbar}\left|K_{fi}\right|^2 \rho(\Delta E),$$

where $\rho(\Delta E)$ denotes the transition lineshape of the transition $|a\rangle \to |b\rangle$, and K_{fi} is the transition operator. In the case of a two-photon transition, it takes the following form :

$$K_{fi}^{(2)} = \sum_g \frac{\langle f|H'|g\rangle\langle g|H'|i\rangle}{E_f - E_g},$$

where g denotes an intermediate state of the composite system, E_m (with $m = f$ or g) the energy of the composite system in the state $|m\rangle$ and H' the interaction Hamiltonian that is given by (in the electric dipole approximation) :

$$H' = -\boldsymbol{E}\cdot\boldsymbol{P} = -i\sqrt{\frac{\hbar}{2\epsilon_0 V}}\left[\frac{\sqrt{\omega}}{n}\boldsymbol{e}\cdot\boldsymbol{P}\left(ae^{i\boldsymbol{k}\cdot\boldsymbol{r}} - a^+e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}\right)\right].$$

We remind the following relations for the annihilition and creation operators :

$$a^+|N\rangle = \sqrt{N+1}|N+1\rangle \text{ and } a|N\rangle = \sqrt{N}|N-1\rangle,$$

where $|N\rangle$ gives the quantum state of N photons in the electromagnetic mode defined by $(\boldsymbol{k},\boldsymbol{e})$.

1. Describe the initial $|i\rangle$ and the final $|f\rangle$ states of the composite system formed by the material and the electromagnetic mode.
2. Identify the intermediate states $|g\rangle$ with a non-zero contribution to the calculation of the two-photon transition operator.
3. Give an expression for $\left|K_{fi}^{(2)}\right|^2$. Subsequently, we suppose that $N \gg 1$.
4. Derive a relation for the rate of energy W that is transferred from the wave at ω to the material. We remind the relation :

$$\left(\frac{dW}{dt}\right)_\omega = \hbar\omega\frac{dP_{i\to f}}{dt}\mathcal{N}_0 V\rho_{aa}^0,$$

with $\mathcal{N}_0$ the total density of population in the material, ρ_{aa}^0 the population in the lower level $|a\rangle$.

5. Show that the variation of photons follows the relation

$$\frac{dN}{dz} = -\beta N^2, \tag{1}$$

with β a coefficient to be determined.

Derivation of the nonlinear susceptibility

6. Which is the order of nonlinearity under study ?
7. In a degenerate case, derive a relation for the nonlinear polarization generated at ω, assuming that the two-photon absorption is predominant.
8. What can you tell about the real and imaginary parts of the nonlinear susceptibility ? Justify.
9. Derive the nonlinear wave equation for the wave at ω.
10. Give an expression of the quantity dN/dz in terms of the nonlinear susceptibility. We remind the relation between the field amplitude and the number of photons in the mode :

$$I = 2nc\epsilon_0|A|^2 = \hbar\omega\frac{cN}{nV}.$$

11. Identifying this expression with the equation (1), give an expression of the nonlinear susceptibility of the material in the degenerate case.