Zeitschrift: Helvetica Physica Acta

Band: 45 (1972)

Heft: 5

Artikel: Coherence resonances in the alignment-orientation coupling process induced by an electric field

Autor: Faist, A.

DOI: https://doi.org/10.5169/seals-114412

Helvetica Physica Acta
Vol. 45, 1972.
Birkhäuser Verlag Basel

Coherence Resonances in the Alignment-Orientation Coupling Process Induced by an Electric Field

by **A. Faist**

Laboratoire d'optique physique, EPF, Lausanne

(6. III. 72)

Summary. The alignment-orientation coupling process is studied in the case of atomic excited states in the presence of an electric field. The basic properties are obtained by applying a static electric field. The phenomenon is more complex in the case of an oscillating electric field and new resonances of the coherence type appear. The general properties of these new resonances are studied with the aid of computational methods.

We consider an atomic excited state of angular momentum $J = 1$ ($I = 0$) subject, in the presence of an electric field E, to a Stark effect giving rise to an energy separation [1].

$$\Delta W = -m^2 \beta_s = -m^2 \cdot a \cdot E^2$$

These sublevels are optically pumped from a unique ground state ($m = 0$) and one observes the light re-emitted by fluorescence.

A. Static Field E

Let us first consider the case of a static electric field. If $\vec{E} = E \cdot [\vec{z}]$, Oz being chosen as the axis of quantification, then the Hamiltonian is for the excited state:

$$\mathscr{H} = \begin{pmatrix} -\beta_s & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -\beta_s \end{pmatrix} \quad \begin{array}{r} m = 1 \\ 0 \\ -1 \end{array}$$

As this matrix is diagonal and time independent, the stationary solution of the matrix density equation [2]

$$\dot{\rho}_{mm'} = \Gamma \rho^0_{mm'} - \Gamma \rho_{mm'} - i[\mathscr{H}, \rho]_{mm'}$$

is given by ($\hbar = 1$)

$$\rho^{\text{stat}}_{mm'} = \rho^0_{mm'} \frac{\Gamma}{\Gamma + i(W_m - W_{m'})}$$

where Γ is $1/\tau$, τ being the lifetime of the excited sublevel $1m\rangle$.

The vector of polarization of the pumping light is chosen so as to give rise to a coherent excitation of the three sublevels

$$\vec{e}_\lambda = \frac{1}{\sqrt{2}} ([\vec{x}] + [\vec{z}])$$

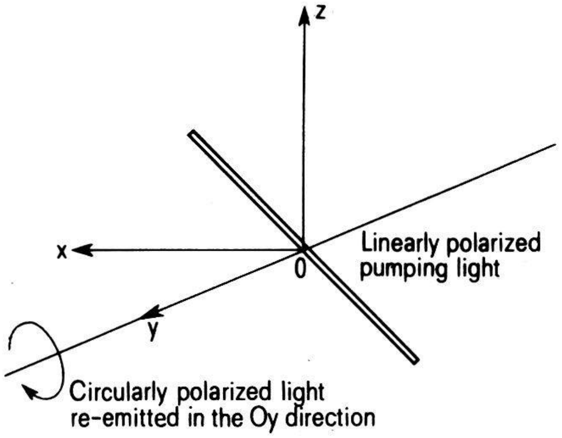

Figure 1
Geometric disposition of the polarizations of the pumping and fluorescence lights.

This gives the initial condition

$$\rho^0_{mm'} = \frac{1}{2} \begin{pmatrix} \frac{1}{2} & -\frac{1}{\sqrt{2}} & -\frac{1}{2} \\ -\frac{1}{\sqrt{2}} & 1 & \frac{1}{\sqrt{2}} \\ -\frac{1}{2} & \frac{1}{\sqrt{2}} & \frac{1}{2} \end{pmatrix}.$$

The stationary solution can now be written down

$$\rho^{\text{stat}}_{mm'} = \frac{1}{2} \begin{pmatrix} \frac{1}{2} & -\frac{1}{\sqrt{2}}\frac{\Gamma}{\Gamma - i\beta_s} & -\frac{1}{2} \\ -\frac{1}{\sqrt{2}}\frac{\Gamma}{\Gamma + i\beta_s} & 1 & \frac{1}{\sqrt{2}}\frac{\Gamma}{\Gamma + i\beta_s} \\ -\frac{1}{2} & \frac{1}{\sqrt{2}}\frac{\Gamma}{\Gamma - i\beta_s} & \frac{1}{2} \end{pmatrix}.$$

The intensity of the re-emitted light can be calculated using the relation

$$I_{\text{fl}} = \sum_m \sum_{m'} < 0|\vec{e}_\lambda \vec{D}|m\rangle \langle m|\rho|m'\rangle \langle m'|\vec{e}_\lambda \vec{D}|0 \rangle.$$

We shall first consider linearly polarized light with the vector of polarization parallel (+) or perpendicular (–) to the polarization of the pumping light:

$$\vec{e}_\lambda = \frac{1}{\sqrt{2}} \quad ([\vec{x}] \pm [\vec{z}])$$

$$I_{fl}^{lin} \propto \tfrac{1}{2}(1 \pm \operatorname{Re}\rho_{10}) = \tfrac{1}{2}\left(1 \pm \frac{\Gamma^2}{\Gamma^2 + \beta_s^2}\right) = \tfrac{1}{2}\left(1 \pm \frac{\Gamma^2}{\Gamma^2 + a^2 E^4}\right).$$

These are absorption curves given on Figure 2:

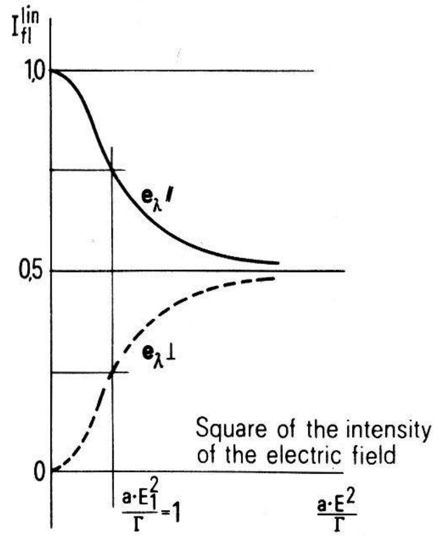

Figure 2
Linearly polarized re-emitted light.

As E^2 is always positive, these curves have no left part. The shape is similar to the right-hand part of a Hanle curve with

$$\Gamma = aE_1^2$$

where Γ is the inverse of the lifetime and a the Stark constant of the considered level.

This particular aspect of the phenomenon had already been observed previously [3].

The linearly polarized light with $\vec{e}_\lambda = [\vec{x}]$ or $\vec{e}_\lambda = [\vec{z}]$ has a constant intensity $I = \frac{1}{2}$. Let us now consider circularly polarized light re-emitted in the Oy direction [4]. We find:

$$I_{fl}^{circ\pm} \propto \tfrac{1}{2}(1 \pm \operatorname{Im}\rho_{10}) = \tfrac{1}{2}\left(1 \pm \frac{\beta_s \Gamma}{\Gamma^2 + \beta_s^2}\right) = \tfrac{1}{2}\left(1 \pm \frac{aE^2 \cdot \Gamma}{\Gamma^2 + a^2 E^4}\right)$$

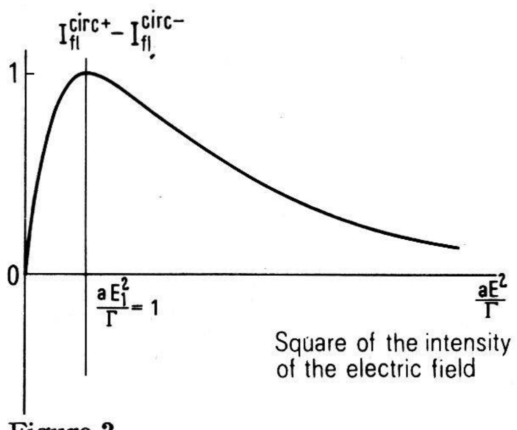

Figure 3
Circularly polarized re-emitted light.

Again $I_{fl}^{circ\pm}$ does not depend on the sign of E.

One should notice that the 'creation' of orientation is maximum for $E_1 = \sqrt{\Gamma/a}$ and vanishes for higher values of E [5].

B. Oscillating Electric Field

Let us now consider the action of an oscillating electric field $\vec{E} = [\vec{z}]\, E \cos \omega t$. The (time-dependent) Stark separation is now

$$\begin{aligned} \Delta W &= -m^2\, aE^2 \cos^2 \omega t \\ &= -m^2 \frac{a}{2} E^2 (1 + \cos 2\omega t) \\ &= -\beta(1 + \cos 2\omega t) \end{aligned}$$

Owing to the fact that the Stark effect is quadratic, the Hamiltonian is now the sum of a time-independent part $(-\beta)$ and a time-dependent part $(-\beta \cos 2\omega t)$.

As before the only field strength dependent terms are the $\rho_{m,m\pm1}$ terms which are solution of the equation

$$\dot{\rho} = \Gamma \rho^0 - \Gamma \rho + i(\beta + \beta \cos 2\omega t)\, \rho$$

where

$$\rho = \rho_{1,0} = \overset{\times}{\rho}_{0,1} = \rho_{-1,0} = \overset{\times}{\rho}_{0,-1}.$$

This is a typical coherence–resonance equation with the solution [6]

$$\frac{\rho}{\rho_0} = A_0 + \sum_{p=1}^{\infty} (A_p\, e^{i2p\omega t} + B_p\, e^{-i2p\omega t})$$

with

$$A_0 = \sum_{n=-\infty}^{+\infty} \frac{\Gamma}{\Gamma - i(\beta + 2n\omega)}\, \mathfrak{J}_n^2\left(\frac{\beta}{2\omega}\right)$$

$$A_p = \sum_{n=-\infty}^{+\infty} \frac{\Gamma}{\Gamma - i(\beta + 2n\omega)}\, \mathfrak{J}_n\left(\frac{\beta}{2\omega}\right) \mathfrak{J}_{n+p}\left(\frac{\beta}{2\omega}\right)$$

$$B_p = \sum_{n=-\infty}^{+\infty} \frac{\Gamma}{\Gamma - i(\beta + 2n\omega)}\, \mathfrak{J}_n\left(\frac{\beta}{2\omega}\right) \mathfrak{J}_{n-p}\left(\frac{\beta}{2\omega}\right).$$

In the case of ordinary coherence–resonances, the intensity of the oscillating magnetic field appears only in the argument of the Bessel functions. Here the intensity of the oscillating electric field appears not only in the Bessel functions but also in the expression $\Gamma/\Gamma - i\ (\beta + 2n\omega)$. Therefore it is possible to pass a resonance by varying only the intensity of the oscillating field. For the same reason, the different resonances corresponding to different values of n are not separated for ordinary values of the electric field.

When β is positive, only the terms corresponding to $n \leqslant 0$ are resonant.

Let us again calculate the intensity of the re-emitted light:

Circularly polarized light

$$I_{\mathrm{fl}}^{\mathrm{circ}\pm} \propto \tfrac{1}{2}(1 \pm \mathrm{Im}\,\rho)$$
$$= \tfrac{1}{2}\left\{1 \pm \left(I_I + \sum_{p=1}^{\infty} I_c^p + \sum_{p=1}^{\infty} I_s^p\right)\right\}$$

where

$$I_I = \sum_{n=-\infty}^{+\infty} \frac{x+2ny}{1+(x+2ny)^2}\,\mathfrak{J}_n^2\left(\frac{x}{2y}\right)$$

$$I_c^p = \sum_{n=-\infty}^{+\infty} \frac{x+2ny}{1+(x+2ny)^2}\left\{\mathfrak{J}_n\left(\frac{x}{2y}\right)\mathfrak{J}_{n+p}\left(\frac{x}{2y}\right) + \mathfrak{J}_n\left(\frac{x}{2y}\right)\mathfrak{J}_{n-p}\left(\frac{x}{2y}\right)\right\}\cos 2p\omega t$$

$$I_s^p = \sum_{n=-\infty}^{+\infty} \frac{1}{1+(x+2ny)^2}\left\{\mathfrak{J}_n\left(\frac{x}{2y}\right)\mathfrak{J}_{n+p}\left(\frac{x}{2y}\right) - \mathfrak{J}_n\left(\frac{x}{2y}\right)\mathfrak{J}_{n-p}\left(\frac{x}{2y}\right)\right\}\sin 2p\omega t$$

with $x = \beta/\Gamma$ and $y = \omega/\Gamma$.

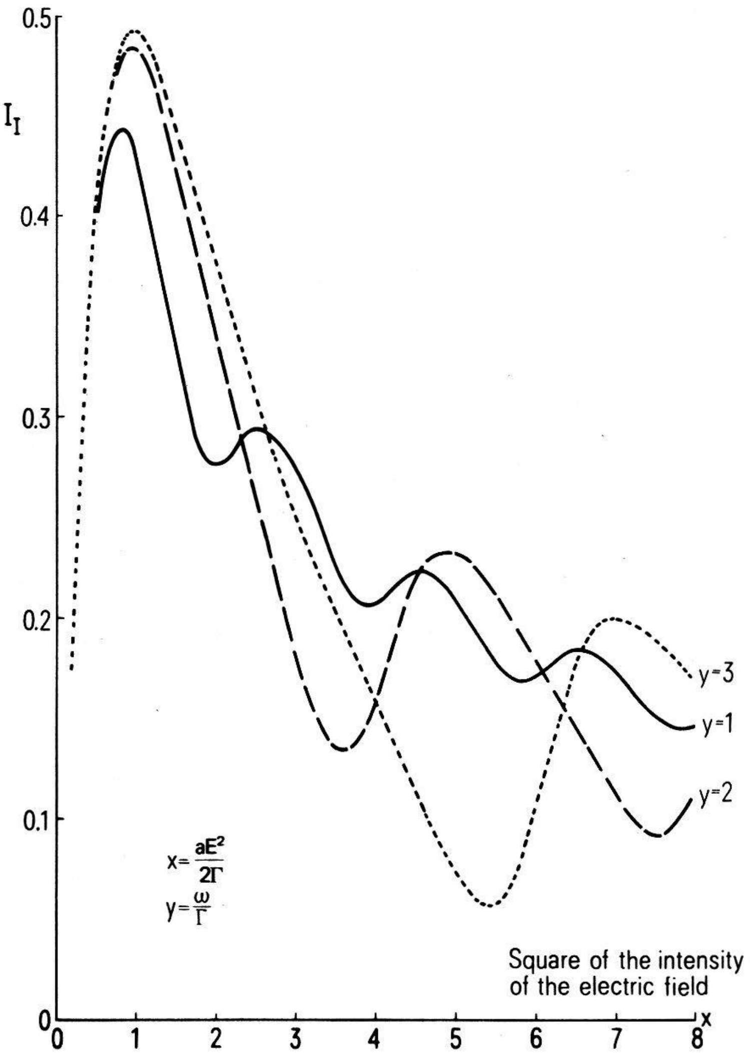

Figure 4

Intensity of the static part of the circularly polarized fluorescence light.

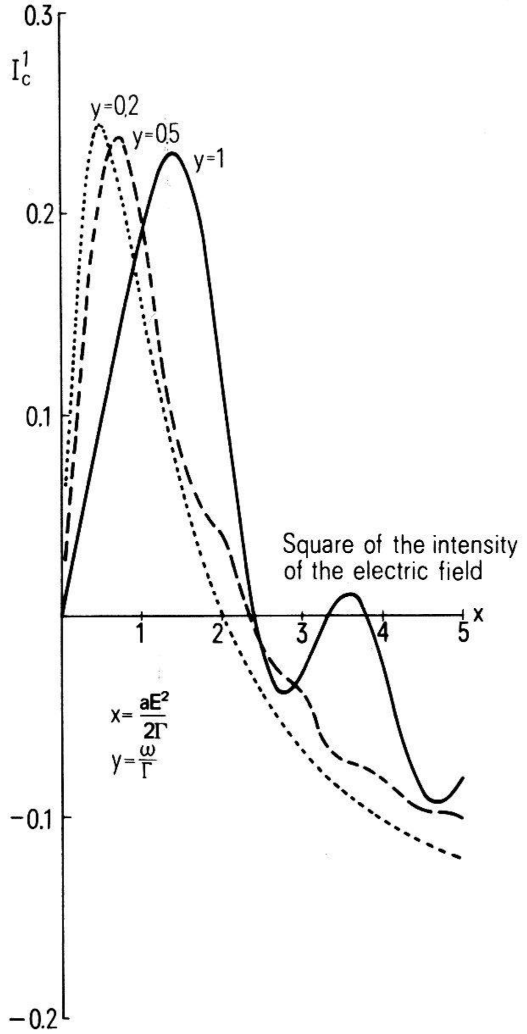

Figure 5
Intensity of the first harmonic ($p = 1$) of the circularly polarized fluorescence light (in phase term).

The re-emitted light consists of a static part I_I and a time-dependent part which can be separated into I_c^p (in phase term) and I_s^p (in quadrature term). With a lock-in type detection, it is possible to detect I_c^p or I_s^p corresponding to a selected harmonic of order p. The intensities I_I, I_c^p and I_s^p have been computed for different values of x, y and p and some of them are given on Figures 4, 5 and 6. For $y = 1$, the re-emitted light I_I (Fig. 4) presents a sharp peak for $x = 0,85$. The position and shape of this peak depends on the value of y. It is interesting to notice that the I_c^1 term ($p = 1$, Fig. 5) presents at a frequency inferior to Γ ($y = \omega/\Gamma = 0,2$) a sharp peak for a Stark separation equal to half of the natural linewidth ($x = \beta/\Gamma = 0{,}5$). The width is of the order of Γ.

Magnetic coherence resonances with the same properties could be obtained by varying the intensity of the oscillating field proportionally to the intensity of the static field.

Linearly polarized light

$$I_{fl}^{lin} \propto \tfrac{1}{2}(1 \pm \operatorname{Re} \rho)$$

$$= \tfrac{1}{2}\left\{1 \pm \left(I_I + \sum_{p=1}^{\infty} \Gamma_c^p + \sum_{p=1}^{\infty} I_s^p\right)\right\}$$

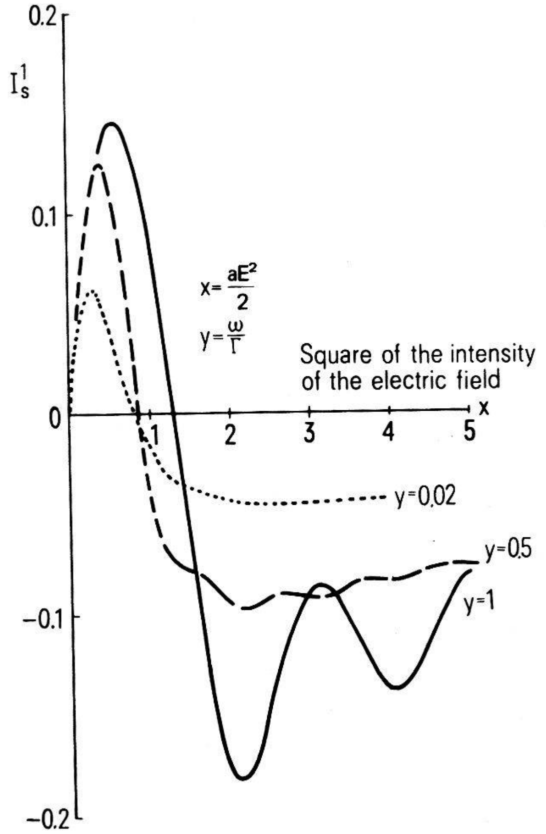

Figure 6
Intensity of the first harmonic ($p = 1$) of the circularly polarized fluorescence light (in quadrature term).

where

$$I_I = \sum_{n=-\infty}^{+\infty} \frac{1}{1+(x+2ny)^2} \mathfrak{J}_n^2\left(\frac{x}{2y}\right)$$

$$I_c^p = \sum_{n=-\infty}^{+\infty} \frac{1}{1+(x+2ny)^2} \left\{\mathfrak{J}_n\left(\frac{x}{2y}\right)\mathfrak{J}_{n+p}\left(\frac{x}{2y}\right) + \mathfrak{J}_n\left(\frac{x}{2y}\right)\mathfrak{J}_{n-p}\left(\frac{x}{2y}\right)\right\} \cos 2p\omega t$$

$$I_s^p = \sum_{n=-\infty}^{+\infty} \frac{x+2ny}{1+(x+2ny)^2} \left\{\mathfrak{J}_n\left(\frac{x}{2y}\right)\mathfrak{J}_{n+p}\left(\frac{x}{2y}\right) - \mathfrak{J}_n\left(\frac{x}{2y}\right)\mathfrak{J}_{n-p}\left(\frac{x}{2y}\right)\right\} \sin 2p\omega t$$

with $x = \beta/\Gamma$ and $y = \omega/\Gamma$

The intensities of I_I, I_c^p and I_s^p have also been computed for different values of x, y and p (Figs. 7, 8 and 9). They do not seem to present very interesting properties.

Interpretation in terms of the 'dressed atom' theory

In this theory [7] we consider the energy levels of the atoms 'dressed' with N photons. Because of the properties of the second order Stark effect, only states with $\Delta N = 2$ have to be taken into account [8].

Coherence resonances of the kind described above occur at $\beta = 0, 2\,\omega, 4\,\omega, \ldots$ when the pumping light puts the atoms in a superposition state $m = 0, \pm 1$. These resonances correspond to

$$n = 0, -1, -2, \ldots$$

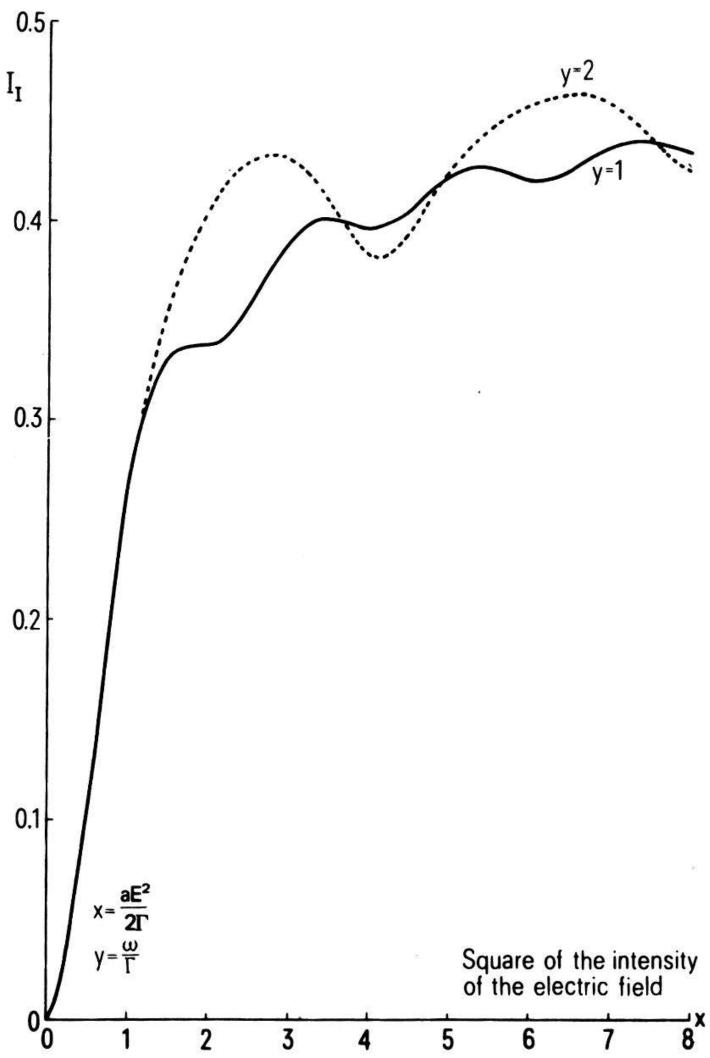

Figure 7
Intensity of the static part of the linearly polarized fluorescence light.

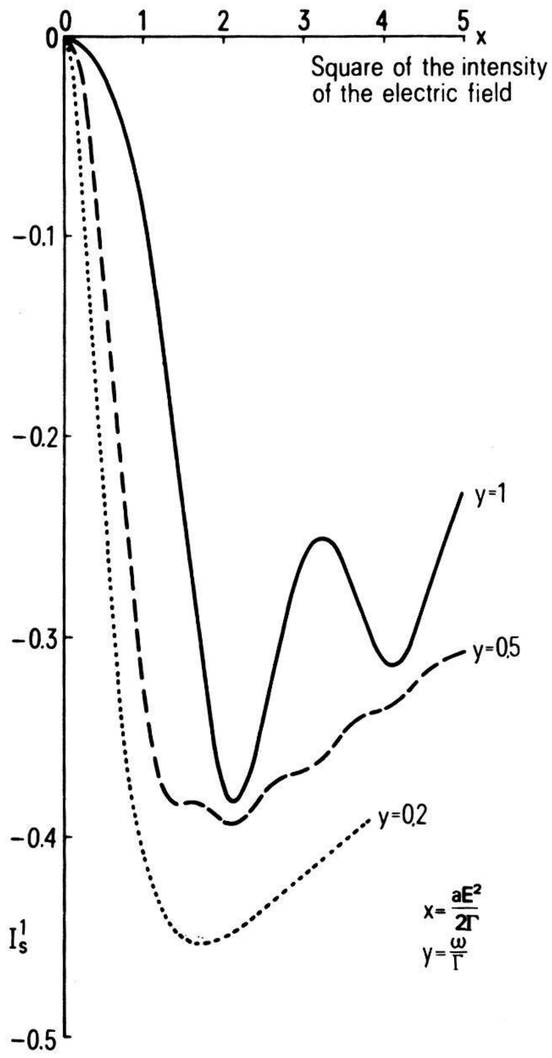

Figure 8
Intensity of the first harmonic ($p = 1$) of the linearly polarized fluorescence light (in phase term).

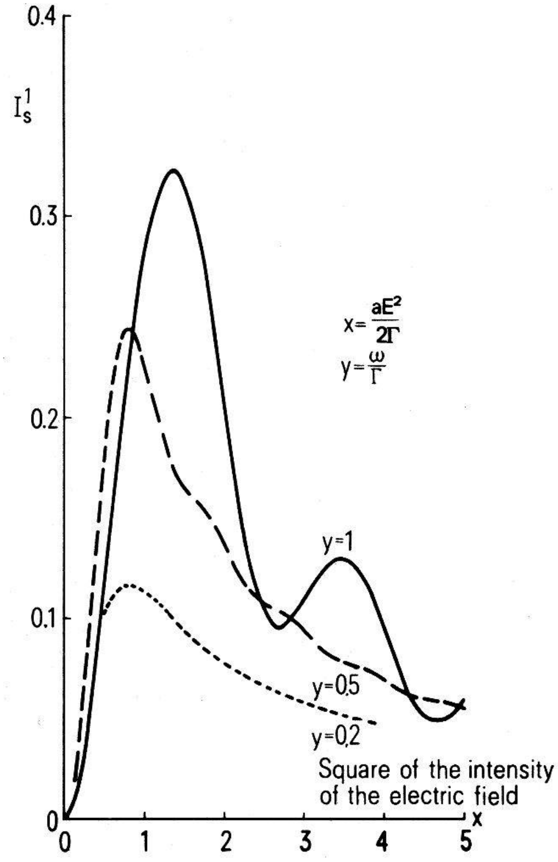

Figure 9
Intensity of the first harmonic ($p = 1$) of the linearly polarized fluorescence light (in quadrature term).

and we see again that if the Stark constant 'a' is positive, it is not possible to observe the resonances corresponding to $n > 0$.

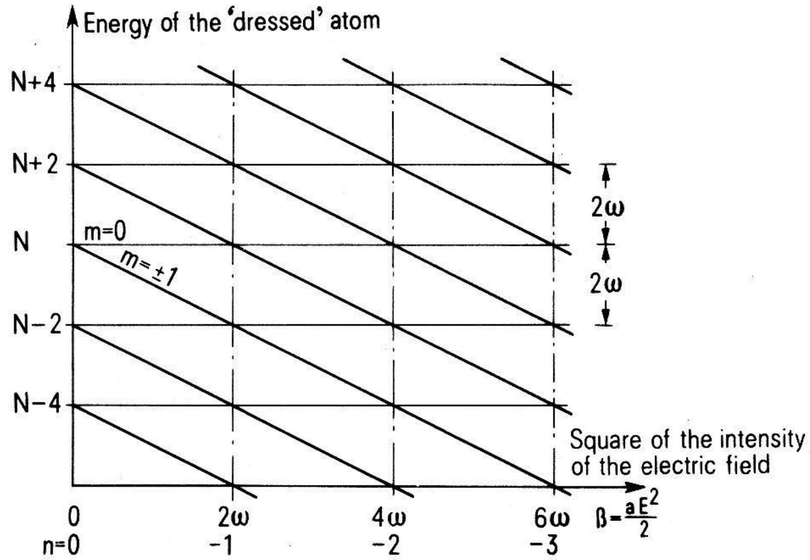

Figure 10
Energy diagram of the 'dressed' atoms.

Conclusion

It has been shown that resonances of the coherence type can be observed in the alignment–orientation coupling process. The principal properties of these new resonances have been described. They could be applied to the measurement of lifetimes, and to the development of an electrometer. If this theory can be applied to states having a sufficiently long lifetime, weak electric fields could be detected and measured.

Acknowledgments

I am indebted to Dr. E. Geneux and Prof. A. Quattropani for many fruitful discussions and to O. D'Auriol for the computation work.

REFERENCES

[1] L. Landau and E. Lifchitz, Mir (1966).
[2] C. Cohen-Tannoudji, Ann. Phys. *7*, 423 (1962).
[3] E. Geneux, private communication (1971).
[4] L. Lombardi, Thesis (Grenoble 1969).
[5] C. Cohen-Tannoudji and J. Dupont-Roc, Opt. Com. *1*, No. 4, 184 (1969).
[6] C. J. Favre and E. Geneux, Phys. Lett. *8*, No. 3, 190 (1964).
[7] C. Cohen-Tannoudji and S. Haroche, C.r. Acad. Sci. Paris *262*, 37 (1966).
[8] A. Faist, Thesis (Geneva 1970).