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Optical nutation in a gaussian beam resonator

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Résumé. — Nous examinons la réponse transitoire d'un résonateur plan-sphérique excité continûment et rempli par un gaz optiquement fin qui est brutalement amené en résonance exacte avec le champ excitateur. Contrairement aux traitements antérieurs, notre approche prend en compte la distribution du champ dans le résonateur — aussi bien axiale que transversale — et les mouvements moléculaires à travers l'onde stationnaire. Ceci est obtenu en projetant la polarisation induite dans le gaz sur le mode gaussien fondamental du résonateur et les résultats sont donnés sous une forme quasi analytique. Une réponse oscillante n'est obtenue que lorsque la fréquence de Rabi est grande devant l'élargissement Doppler et le résonateur de mauvaise qualité. Certaines de nos prévisions théoriques ont été confirmées par une expérience à une longueur d'onde de 6 mm.

Abstract. — We examine the transient behaviour of a plane-spherical resonator driven by a continuous wave and filled by an optically thin gas which is suddenly switched on exact resonance with the driving field. Contrary to previous treatments, the axial and transversal field distributions and the molecular motions through the standing wave are taken into account. This is achieved by projecting the induced gas polarization on the fundamental gaussian mode of the resonator and the results are given in a quasi analytic form. An oscillatory response is only obtained when the Rabi frequency is large with respect to the Doppler line width and for a bad quality resonator. Some of the theoretical predictions have been supported by an experiment at a 6 mm-wavelength.

1. Introduction. — In analogy with the well known transient nutation effects in nuclear magnetic resonance [1], the term of optical nutation is currently used to describe the transient response of an optically thin gas of 2-level systems suddenly excited by a strong resonant optical field [2]. Oscillations at the Rabi frequency have been predicted [2] and observed [3] when the optical nutation is excited by a travelling wave. We discuss in this paper the corresponding phenomenon occurring inside a cavity resonator. This study is motivated by the frequent use of intracavity arrangements in order to achieve strongly saturating fields in laser spectroscopy and by the recent prediction of Rabi oscillations in transient absorptive optical bistability [4, 5, 6] when the driving field is switched from the lower to the upper branch of the bistability curve [4].

Since we are interested in the Rabi oscillations, the calculations are made in the infinite saturation limit : the Rabi period is assumed to be very short with respect to the radiative lifetime, the collisional lifetime and the transit time across the optical beam. For the strong optical fields required by this condition, we can assume without loss of generality that the gas is optically thin. On the other hand, we will take a full account of the field distribution (axial and transversal) and of the molecular motions through the standing wave [7] which are usually neglected in spite of their importance (1).

The arrangement of the paper is the following. Our basic model is presented in section 2 which is devoted to a general calculation of the gas polarization projection on the fundamental gaussian mode of the resonator. Theoretical shapes of the time dependence of this polarization and of the resulting transient response of the resonator are discussed in section 3. We present in section 4 some experimental results obtained at a 6 mm-wavelength which are in good agreement with the theoretical ones. We conclude in section 5 by summarizing the main features of this study and by evoking some applications.

2. Transient gas polarization. — 2.1 Basic theoretical model. — We consider a commonly used...
laser resonator of large aperture, which consists of a flat mirror facing a spherical one of radius $R_0$ larger than the length $l$ (see Fig. 1). This open resonator is driven at the frequency $\omega$ of one of its fundamental gaussian mode by an external c.w. optical field, and is filled by a gas of two level systems initially off resonance. At time $t = 0$, the transition frequency $\omega_0$ of the two level systems is suddenly switched on exact resonance with the driving field and we are interested in the resulting time dependence of the induced gas electric polarization $P$ and of the optical field $E$ inside the resonator. These evolutions are governed by the well-known Bloch-Maxwell coupled equations. By a slight adjustment of the Lamb’s laser theory [9], the equation governing the electric field is conveniently written down as:

$$\left( A - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \frac{\omega}{Qc^2} \frac{\partial}{\partial t} \right) [E - E^{(0)}] = - \mu_0 \omega^2 P$$

where $Q$ is the resonator quality factor and $E^{(0)}$ the c.w. optical field in the empty resonator case.

Since the losses of the empty resonator are exactly compensated by the external driving field, $E^{(0)}$ is a solution of the equation:

$$\left( A - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E^{(0)} = 0$$

with suitable limit conditions ($E^{(0)} = 0$ on the mirrors).

As indicated before, we are interested in the optically thin resonator case (the so-called one atom solution or non cooperative state in optical bistability [4]). The absorbing molecules are not coupled with one another but interact individually with the optical field which remains nearly unaffected by the gas ($E \simeq E^{(0)}$). From equation (1) and equation (2), this condition is easily derived as:

$$Q \mid P \mid \ll \varepsilon_0 \mid E^{(0)} \mid.$$

The maximum available gas polarization is generally $n\mu$ where $n$ is the population difference per unit volume and $\mu$ the transition dipole moment element. In the optical domain, the Doppler width $k\tau_0 (k = \omega/c; \tau_0$ most probable molecular velocity) is generally large with respect to the Rabi frequency ($\omega_1 = \mu E^{(0)}/\hbar$) and we have to take account of the hole burnt by the optical field in the velocity distribution. Its width is about $\pi^{1/2} \omega_1$ and the actually available gas polarization is then $n\mu \pi^{1/2} (\omega_1/k\tau_0)$. Introducing the absorption coefficient $\beta = \pi^{1/2} n\mu^2/\hbar \omega_1 \tau_0$ proportional to the pressure, the condition (Eq. (3)) becomes:

$$\beta F/\pi \ll 1$$

where $F$ is the resonator finesse [10] ($F = \pi Q/kl$, $l$: resonator length, see Fig. 1). Equation (4) means that the resonator must be optically thin in the linear regime.

On the other hand, when the whole Doppler profile is excited ($\omega_1 \gg k\tau_0$), the condition given by equation (3) may be conveniently expressed by introducing the linear absorption coefficient at infinitely large pressure $\alpha_\infty = \sigma_\mu^2 \tau/\hbar \epsilon_0 c$, where $\tau$ is the molecular mean free path duration. Equation (3) then becomes:

$$\omega_1 \tau \gg \alpha_\infty (F/\pi).$$

Hence, in so far as the saturation parameter $\alpha_\infty \tau$ is strong, the resonator must not necessarily be optically thin in the linear regime. In the bistability terminology [4], $\alpha_\infty (F/2 \pi)$ is nothing else that the bistability parameter $\alpha_\infty F/2 T = C$ where $T \ll 1$ is the transmittivity of the two mirrors. If the resonator is actually bistable ($C > 4$), equation (5) expresses that the applied field has to be sufficient to put it in the non cooperative region as indicated before. To come back to the crude estimations of equation (4) and equation (5) (corroborated by more exact forthcoming equations (10) and (11)), the condition of optical thinness in the linear regime appears then to be sufficient in all cases but not always necessary.

Besides, we are in this paper interested not in the relaxation processes but in the nutation signal and in the influence of the field inhomogeneity and of the molecular motions on this signal. We shall then assume that the mean Rabi frequency is much larger than collision and radiative broadenings (infinite saturation regime). This is achieved at low sample pressures for which the resonator is actually optically thin.

2.2 DERIVATION OF THE INDUCED GAS POLARIZATION. — Within the previous assumptions, the Bloch-Maxwell equations are uncoupled and the gas polarization is simply calculated from the unperturbed field $E^{(0)}$ (fundamental gaussian mode). This field is quasi-linearly polarized, i.e. tangent to the spherical wave front ($\Sigma$) and parallel to a fixed meridian plane $XOz$ (see Fig. 1). We characterize a point $M$ inside
the resonator by \( z \) being the abscisse of the \( z \)-axis equiphase point and by \( x \) and \( y \) being its curvilinear coordinates measured along the wave front (\( \Sigma \)). This change of coordinates involves some simplification to the expression of the field describing the fundamental gaussian mode (approximate solution of equation (2)) [11]. In complex notation, omitting the \( e^{i\omega t} \) time dependence, the component \( \hat{E}^{(0)} \) of \( E^{(0)} \) according the \( x \)-coordinate becomes:

\[
\hat{E}^{(0)} = E_a \sin \left\{ k z - \Phi(z) \right\} \left\{ w_0/w(z) \right\} \times \\
\times \exp \left\{ - (x^2 + y^2)/w^2(z) \right\} \quad (6a)
\]

\( w_0 \) is the beam waist on the plane mirror (minimum beam waist):

\[
w_0 = (2/\kappa)^{1/2} \left\{ l (R_0 - l) \right\}^{1/4} \quad (6b)
\]

\( w(z) \) is the beam waist in \( z \):

\[
w(z) = w_0 \left[ 1 + (2 z/k w_0^2)^2 \right]^{1/2} \quad (6c)
\]

\( \Phi(z) \) is a phase shift given by:

\[
\Phi(z) = \arctg \left( 2 z/k w_0^2 \right) \quad (6d)
\]

and the \( n \)th gaussian fundamental mode is defined by the condition of resonance:

\[
k l - \Phi(l) = n \pi \quad (6e)
\]

which means that the optical field is zero on the plane and spherical mirrors.

In each point \( M \) defined by \( r = (x, y, z) \) we can characterize the interaction between the optical field \( E^{(0)} \) and a molecule by a Rabi frequency \( \omega_1 (r) \) which depends on \( r \) and has a maximum value \( \omega_1 = \mu E_a/\hbar \). However the beam waist \( w \) is always much larger than the wavelength \( 2 \pi/\kappa \) and the gradient of \( \omega_1 (r) \) is nearly periodic in each point to the corresponding wave front. Then, only molecular motions perpendicular to the wave fronts have to be considered.

This approximation is obviously related to the infinite saturation limit considered here:

\[
\omega_1 \gg v_0/w_0 \quad (7)
\]

In this limit, we can calculate the induced gas polarization by using the result obtained by one of us [7] in the case of a plane standing wave with a full account of the molecular motions. Taking account of the actual distribution of the optical field in the gaussian mode, the polarization \( P(r, t, v) \) in \( r \) due to molecules having a \( v \)-velocity along the wavefront normal is easily written down from equations (13), (14) and (19) of the quoted paper. In complex notation omitting the rapid time dependence in \( e^{i\omega t} \), the polarization component according the \( x \)-coordinate becomes:

\[
\tilde{P}(r, t, v) = -i \mu \sin F \quad (7a)
\]

\[
F = \frac{2 \omega_1}{k \nu} \sin \left\{ \frac{k v t}{2} \right\} \sin \left\{ k z - \Phi(z) - \frac{k v t}{2} \right\} \times \\
\times \left\{ w_0/w(z) \right\} \exp \left\{ - (x^2 + y^2)/w^2(z) \right\} \quad (7b)
\]

To obtain this result the rotating wave approximation has been made and \( \tilde{P}(r, t, v) \) is assumed not to change during a time of the order of \( \omega^{-1} \) (slowly varying amplitude approximation).

2.3 PROJECTION OF THE POLARIZATION ON THE FUNDAMENTAL MODE. — In the case of a plane standing wave, the polarization was developed in spatial harmonics and only the first (fundamental) spatial harmonics in \( \exp[\pm ikz] \) were shown to contribute to the detected signal [7]. In the present problem, we can also reasonably presume that the induced gas polarization has the same axial spatial quasi-periodicity as the optical field \( E^{(0)} \) and then that the resonator eigenmodes with a principal number differing from \( n \) are not excited. On the contrary, it is obvious that the corresponding transverse (higher order) modes of suitable symmetry [9, 11] are actually excited. However, two physical arguments may be given to limit our calculations to a monomode approximation [14]:

(i) The transverse modes are only weakly excited in so far as their distance \( \Delta \omega \) apart the driven fundamental mode is generally larger than the width of the Fourier spectrum of \( \tilde{P}(r, t, v) \) around the zero-frequency (extended slowly varying amplitude approximation). Moreover even excited, these modes, owing to their poor quality factor, will contribute weakly to the transient response of the resonator.

(ii) The contribution of the driven fundamental eigenmode can be easily picked out with a suitable detection scheme, namely with a low-pass filter eliminating the \( \Delta \omega \) — beats corresponding to the excited transverse modes (heterodyne detection, see section 3.2).

For these reasons and also in order to keep tractable equations, we limit our calculations to the projection \( \tilde{P}(t, v) \) of \( \tilde{P}(r, t, v) \) on the driven fundamental mode:

\[
\tilde{P}(t, v) = \int_{(v)} u_\nu (r) \tilde{P}(r, t, v) \frac{dV}{V_0} \quad (8a)
\]

where the integral is extended to the resonator volume \( (V) \), \( u_\nu \) describes the fundamental mode [11] \( (u_\nu = \hat{E}^{(0)}/E_a \) see equation (6a)) and \( V_0 \) is a normalization volume such as:

\[
V_0 = \int_{(v)} u_\nu^2 (r) \, dV = \frac{\pi w_0^2 l}{4} \quad . \quad (8b)
\]
The (x, y) integrations of equation (8a) are easily performed but the z-integration is more difficult. However, since beam parameters, particularly $\Phi(z)$ and $w(z)$ are slowly varying vs. $z$ at a wavelength scale, this latter integration can be split into a sum of $n$ integrals ($n$ : mode number), each of them being calculated between two optical field nodes. Using the Fourier expansion of $\exp(iz \sin \theta)$ in Bessel functions [15] and approximating the previous summation to a z-integral ($n \gg 1$), we finally get:

$$\tilde{P}(t, v) = -i n \Pi(t, v)$$

(9a)

where $\Pi(t, v)$ is a reduced polarization given by:

$$\Pi(t, v) = 4 \int_0^1 \left[ 1 - \frac{2 \omega_1}{kv} \sin \left( \frac{kvt}{2} \right) \frac{w_0}{w(z)} \right] \times$$

$$\times \cos \left( \frac{kvt}{2} \right) \frac{w(z)}{w_0} \frac{dz}{T}.$$  

(9b)

This result is somewhat general since it describes the case of a variable beam waist. The integral can be dropped in the case of a nearly constant beam waist (i.e. for $R_0 \gg l$). In this case, the averaging on the velocity distribution gives:

$$\Pi(t) = \frac{4}{\pi^{1/2}} \int_{-\infty}^{\infty} \left[ 1 - \frac{2 \omega_1}{kv} \sin \left( \frac{kvt}{2} \right) \right] \times$$

$$\times \cos \left( \frac{kvt}{2} \right) \exp \left( -\frac{v^2}{v_0^2} \right) \frac{dv}{v_0}.$$  

(9c)

We discuss in the following section the signal shapes of the reduced polarization $\Pi(t)$ given by equation (9c) and of the resulting transient response of the resonator.

3. Theoretical signal shapes. — 3.1 Reduced gas polarization. — Our study is limited to the case of a nearly constant beam waist (Eq. (9c)). Numerical calculations (not reported here) have shown that nutation signals are not strongly distorted as long as the beam waist change along the resonator axis is less than 15% ($R_0/l \gg 4$) whereas larger beam waist changes lead to an extra smoothing of the Rabi oscillations.

The reduced polarization $\Pi(t)$ has been plotted in figure 2 for various ratios of the most probable Doppler shift ($kv_0$) to the maximum Rabi angular frequency ($\omega_1$). The signal rise time is slower (first maximum for $\omega_1 t = 2.8$ instead of 1.8) and the Rabi oscillations are dramatically damped by inhomogeneous effects (relative amplitude of the 2nd oscillation : 5% instead of 60%). Let us also notice that $\Pi(t)$ remains in the upper half plane, namely in the absorptive domain in contrast with the travelling wave case [16].

3.1.1 Negligible molecular motion case ($kv_0 \ll \omega_1$).

— The reduced polarization $\Pi(t)$ derived from equation (9c) takes a very simple analytic form in this case:

$$\Pi(t) = 4 \frac{1 - J_0(\omega_1 t)}{\omega_1 t}.$$  

(10)

The corresponding shape (Fig. 2, thick solid line) differs strongly from the corresponding shape obtained in a plane standing wave ($\Pi(t) = 2 J_1(\omega_1 t)$). The signal rise time is slower (first maximum for $\omega_1 t = 2.8$ instead of 1.8) and the Rabi oscillations are dramatically damped by inhomogeneous effects (relative amplitude of the 2nd oscillation : 5% instead of 60%). Let us also notice that $\Pi(t)$ remains in the upper half plane, namely in the absorptive domain in contrast with the travelling wave case [16].

3.1.2 Prevailing molecular motion case ($kv_0 \gg \omega_1$).

— In this case, the molecules move so fast that they pass through several nodes of the optical field during a Rabi period.

Equation (9c) is then conveniently rewritten down as:

$$\Pi(t) = \frac{\pi^{1/2}}{kv_0} \frac{1}{\omega_1 t} \left\{ 16 \operatorname{erf} \left( \frac{kv_0 t}{2} \right) \right\} \times$$

$$\times \int_0^{\infty} \left[ 1 - J_0(\omega_1 t \sin u) \right] \frac{\cos u}{\omega_1 t \sin u} \frac{du}{\sin u}.$$  

(11)

with $\sin u = \sin \omega_1 t \sin u$. The error function [15] $\operatorname{erf}(kv_0 t/2)$ describes the short time behaviour of $\Pi(t)$ and is obtained by a development of equation (9c) at the first order in $\omega_1 t$. The maximum value $\pi^{1/2} \omega_1 /kv_0$ of $\Pi(t)$ is then reached for times $t$ such as $\omega_1 /kv_0 \ll \omega_1 t < 1$ [7]. As expected, the
Rabi oscillations are smeared out by molecular motions and the time evolution of $\Pi(t)$ is reduced to a simple transient pulse with a duration of the order of the Rabi period (see Fig. 2, thick dashed line). Let us however notice a small overshoot in the negative lower half plane. This feature, which contrasts with the negligible molecular motion case, is due to the fast molecules which are $\pm kv$ off resonance ($v : z$-component of the molecular velocity). They then give a contribution to the polarization evolving as $\sin (kvt)$, i.e. unaffected by the $\omega_1$-inhomogeneities [17].

3.1.3 Intermediate cases ($\omega_1 \approx kv_0$). — The shapes plotted for some intermediate cases show that the motional effects have rapidly a great influence on the Rabi oscillation amplitude. As an example, when the Rabi frequency is equal to the Doppler width ($\omega_1 = kv_0$, see Fig. 2, thin solid line), all the oscillations disappear and except for the rise time, the signal is quite similar to the one obtained in the Doppler limit.

3.2 Resonator Transient Response. — As indicated before, only the fundamental gaussian mode driven by the external field is significantly excited by the induced polarization. The optical field inside the resonator is then conveniently written as :

$$\tilde{E}(r, t) = \{ E_n + \tilde{\alpha}(t) \} u_n(r) \quad (12)$$

$u_n(r)$ describes the $n$th fundamental mode, $E_n$ the maximum field amplitude in the empty resonator (0th order solution) and $\tilde{\alpha}(t)$ its first order change due to the induced polarization. The output field, proportional to $E_n + \tilde{\alpha}(t)$ is received on a non linear detector which, at the first order approximation gives a signal proportional to $\tilde{\alpha}(t)$ (heterodyne detection). Some detectors have even a constant conversion factor in a large domain of incident power and in this domain, the time-dependent detected signal will be directly proportional to $\tilde{\alpha}(t)$ whatever $E_n$ is.

In all cases, $\tilde{\alpha}(t)$ appears to be the quantity of experimental interest. By substituting equation (12) in the starting equation (1) or more exactly in the corresponding approximate scalar equation and by projecting on the $n$th fundamental mode, we get straightforwardly :

$$\tilde{\alpha}(t) + \tau_{ph} \frac{d}{dt} \tilde{\alpha}(t) = - (Q/e_0) n\mu II(t) \quad (13)$$

where $\tau_{ph} = 2Q/\omega$ is the mean photon lifetime inside the resonator [5, 6] (resonator rise time to a step excitation) and where $II(t)$ is the reduced polarization (see Eq. (9)). Equation (13) is the general equation governing the evolution of a system of time constant $\tau_{ph}$ submitted to a time — dependent external excitation. Its solution depends dramatically on the relative values of the time constant $\tau_{ph}$ and of the times characterizing the evolution of the $II(t)$ source term, especially the Rabi period $2\pi/\omega_1$.

In the bad quality cavity limit ($\omega_1 \tau_{ph} \ll 1$) [18], the signal $\tilde{\alpha}(t)$ follows exactly the time dependence of the reduced polarization $II(t)$ (adiabatic approximation). On the contrary, in the good quality cavity limit ($\omega_1 \tau_{ph} \gg 1$) [18], the resonator cannot follow $II(t)$ which acts as a pulse excitation (sudden approximation). This excitation is entirely defined by its impulse $\int_0^\infty II(t') \, dt'$. The resonator response is then easily written :

$$\tilde{\alpha}(t) = -(Q\mu/2e_0) \exp(-t/\tau_{ph}) \int_0^\infty II(t') \, dt' \quad (14)$$

and the signal is reduced to a single exponential of time constant $\tau_{ph}$.

In figure 3 have been plotted typical signal shapes obtained in the negligible molecular motion case ($kv_0 \ll \omega_1$) for different values of $\omega_1 \tau_{ph}$. The Rabi oscillations disappear as soon as $\omega_1 \tau_{ph} > 3$ but on the other limit the fall time constant remains twice...
larger than $\tau_{\text{ph}}$ for $\omega_1 \tau_{\text{ph}}$ as large as 30. This can be attributed to the slow falling down of the $\Pi(t)$ function (see Eq. (10)) evolving as $(\omega_1 t)^{-3/2}$ for $\omega_1 t \gg 1$. Similar results are obtained in the Doppler limit, excepted for the pulse response which is more marked since the $\Pi(t)$ duration decreases when $k v_0 / \omega_1$ becomes larger.

4. Experiment. — The previous theoretical results have shown that good conditions for the observation of Rabi oscillations in a standing wave need a moderate Doppler effect and a rather bad quality resonator (3). Thus, the microwave range seems suitable for this purpose, as it will be easy to get a saturation broadening much larger than the Doppler broadening.

The experiments were done on the $J=0 \rightarrow J=1$ line of CH$_3$F at 51 GHz [19]: this very strong transition is non degenerate, and has a large dipole moment matrix element ($\mu \sim 0.75$ Debye) combined with a moderate Doppler broadening ($k v_0 / 2 \pi \sim 57$ kHz at room temperature). The working pressure was of the order of 1 mtorr, leading to a moderate collisional broadening

$$\Delta \nu_{\text{coll}} / \nu \sim 20 \text{ kHz/mtorr}.$$  

A plane cylindrical resonator between Stark plates was used [20]. This device allowed the use of the Stark switching technique: the e.m. field was continuously applied in the cavity, so that only the transient absorption was detected when the molecular frequency was switched.

The excited modes were TE types with the electric field parallel to the Stark plates allowing for $\Delta M = \pm 1$ selection rules.

Due to the Stark plates, the e.m. field is slightly different from a gaussian beam: the modes are described by sine functions in the $y$-direction perpendicular to the Stark plates and a gaussian dependence in the other transverse direction. By introducing the curvilinear coordinates (see Eq. (6)), the electric field of the $n$th TE$_n$ fundamental mode is shown to be [20]:

$$\hat{E}^{(n)} = E_n \sin \left( k_g z - \Phi(z)/2 \right) \sin \left( n y / b \right) \left( w_0 / w(z) \right)^{1/2} x \times \exp \left\{ -x^2 / w^2(z) \right\} \quad (15)$$

where $b = 15$ mm is the plate spacing and $k_g$ is the propagation constant of the TE$_n$ fields between two parallel plates ($k_g^2 = k^2 - \pi^2 / b^2$).

$w_0$, $w(z)$ and $\Phi(z)$ are given by substituting $k_g$ to $k$ in equations (6b, c, d), and $\Phi(l)/2$ to $\Phi(l)$ in equation (6e). The curvature radius $R_0$ is about 5 m and the resonator length is about 1 m leading to a 10% beam waist variation along the cavity. Except for a complete numerical treatment, the previous theory applies and leads to quite similar behaviours within a 10% difference.

The driven fundamental mode had a quality factor of $Q \sim 29$ 000 which gives at 51 GHz a photon lifetime of 180 ns. The nearest higher order mode (TE$_3$) was 15 MHz apart, had a quality factor of about 10 000 and, as expected, was never excited in our experiments (see discussion of section 2.3).

The driving microwave power was supplied by a klystron phase locked at the zero Stark field line frequency. The resonator eigenfrequency was also tuned at this frequency. A uniline (25 dB rejection) avoided coupling problems between the cavity and the klystron.

The Stark field was switched by a 1 500 volt pulse generator (rise time $\sim 100$ ns). The corresponding molecular frequency switching was about 8 MHz.

The detection of the output field was achieved by a Schottky barrier diode whose conversion factor is nearly constant with a suitable bias for incident power up to a few mW. The detected signal ($\hat{a}(t)$) was treated in a 256-channel digital averager (channel width : 10 ns) working at a 3 kHz repetition rate. Although ground-loop currents were avoided by a careful grounding of the experiment, residual spurious signals were always eliminated by subtracting the averaged signals obtained with and without gas. Up to $2 \times 10^6$ cycles were done without great problems of resonator thermal drifts (total averaging duration $\sim 10$ min.).

Experiments were done with various microwave powers inside a 40 dB range. Special attention was paid to the low and strong power limits corresponding to the bad or good quality cavity limit [18] in the theoretical discussion (section 3.2). Figure 4

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(3) This latter prediction agrees with the analysis given in Refs. [4] and [5].
gives a good example of a negligible motion case with a bad quality cavity behaviour. The Rabi frequency is about six times as large as the Doppler line width and the Rabi period about 16 times as long as the photon lifetime. In this case, the Rabi oscillations are well marked. The experimental points were fitted by a least square method to the theoretical shape taking account of the actual mode distribution (including the small beam waist variation), the residual Doppler effect and the photon lifetime. A correction of the type \(1 - \exp(-\Gamma t)\) was also added to take account of residual intermolecular and wall collisions \((\Gamma = 1/\tau\) is the collision rate \(\sim \omega_1/25\) in the experiment). Owing to these improvements, the agreement between the calculated shape and the experimental points is fairly good, the residual standard error being only 0.8 \% the maximum amplitude. This agreement must be attributed to the precise knowledge of the microwave field distribution in contrast with the travelling wave case [16]. A good confidence can then be reasonably given to the corresponding value of the Rabi frequency \((\omega_1/2 \pi = 350\ \text{kHz})\) which was used as a reference to determine its value in other experimental cases by a simple calibrated attenuator reading.

As the microwave power is increased, the Doppler line width becomes quite negligible with respect to the Rabi frequency but at the same time the corresponding Rabi period becomes shorter than the photon lifetime and we fall in the good quality cavity limit.

![Fig. 5. — Experimental transient resonator response in the good quality cavity limit. CH\(_3\)F pressure = 1 mtorr, collision rate \(\Gamma\) (with \(\Gamma/2 \pi = 23\ \text{kHz}\), microwave field \(E_0 \approx 3\ \text{kV/m}\), Rabi frequency \(\omega_1/2 \pi \approx 12\ \text{MHz}\), saturation parameter \(\omega_2/\Gamma \approx 520\), \(\omega_1 \tau_{ph} \approx 14\), \(k_{R} / \omega_1 \approx 5 \times 10^{-5}\). The time constant of the exponential fall is about 0.25 \(\mu\)s.](image)

As an example, figure 5 gives the result obtained when the microwave power entering the cavity was set at its maximum value. The corresponding Rabi frequency (determined by reference to the previous case) is 12 MHz (microwave field \(\sim 3\ \text{kV/m}\), \(\omega_1 \tau_{ph} \sim 14\)). The signal has the theoretically predicted shape (see Fig. 3b the curves related to \(\omega_1 \tau_{ph} = 10\) and 30). Its rise time, slower than expected, was in fact bounded by the Stark voltage rise time (\(~ 100\ \text{ns}\) ). We also noticed that the signal amplitude increased as the microwave power was decreased. This surprising feature may be explained as follows. We were in a power domain for which the detector conversion factor was nearly constant and the detected signal was then proportional to the first order field change \(\vartheta(t)\) namely in the good quality cavity limit to \(\int_0^\infty \Pi(t')\ dt'\) (see section 3.2, particularly equation (14)). In all cases, \(\Pi(t')\) is only dependent of \(\theta = \omega_1 t' (dt' = d\theta/\omega_1)\) and we get:

\[
\vartheta(t) \sim \frac{1}{\omega_1} \int_0^\infty \Pi(\theta)\ d\theta.
\]

The detected signal is then theoretically in inverse ratio to the microwave field. This is in qualitative agreement with our experimental observations. However a quantitative comparison was not possible since, in the case of large microwave power, the saturation broadening (namely the Rabi frequency) was larger than the Stark shift and the absorption was then not fully switched off-on. Anyway, this last point affects only the signal amplitude, not its shape which is mainly related to the resonator response. This point was checked in an experiment made at a lower Stark field.

5. Conclusion. — We have presented an extensive study of the optical nutation of 2-level systems in a passive resonator for the negligible relaxation case (infinite saturation). The obtained integral form of the induced polarization takes account of the molecular velocities in the standing waves and of the exact optical field mode distribution inside the cavity. These two effects combine to give very weak or even unexisting Rabi oscillations:

i) on the one hand, longitudinal motions through the standing waves lead to a kind of transit time effect, which is obviously in close connection with the Doppler effect;

ii) on the other hand, we have pointed out the necessity of the gas polarization projection over limited transverse extension modes. This more physically meaningful model leads to a result quite different from the plane standing wave case [7]. Molecular motions are not involved in this process, but the molecular distribution inside the inhomogeneous optical beam leads to various Rabi frequencies, and then to a strong damping of the nutation signal. As each molecule gives a signal projected in the used mode, this effect does not depend on the actual size of the optical beam, but is only related to the total inhomogeneity of the field. Only these inhomogeneous effects are present when the Rabi frequency \(\omega_1\) is much larger than the Doppler line width and an analytical result has been given at this limit.
The influence of the resonator quality factor on the transient response has also been examined. In the bad quality cavity limit \((Q \ll \omega/2 \omega_1)\), the output field follows adiabatically the transient induced gas polarization whereas in the good quality cavity limit \((Q \gg \omega/2 \omega_1)\), this polarization acts only as an impulse excitation.

The reported experiment at a millimetre wavelength is in full agreement with the proposed theoretical model.

These results show that the Doppler effect and, most of all, the inhomogeneities in the optical field would have a great influence on Rabi oscillations predicted in a optical device which is switched from the cooperative to the non cooperative state \([4, 5, 6]\). More recently photon echoes in standing wave fields \([21, 22, 23, 24]\) have been observed and studied: the Doppler effect is shown to play an important role in the echo formation, but one can wonder whether the preparation step (\(\pi/2\) pulse) and the phase reversal step (\(\pi\) pulse) are not to be more precisely examined. The theory presented in this paper is well suitable for such a purpose and can be extended to take account of eventual level degeneracies \([25, 26]\).

Besides, our model can be applied to the calculation of the two photon optical nutation \([27]\) in a gaussian standing wave \([28]\).

References