Supernutation

A. I. Burshteĭn and A. Yu. Pusep

Institute of Chemical Kinetics and Combustion, Siberian Branch, USSR Academy of Sciences, Tomsk
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A calculation is made of the dependence of the frequency of energy exchange between N two-level atoms and a monochromatic field on the initial number of photons n_0 in the resonator. If $n_0 \ll N$, the frequency is governed by the number of atoms, but if $n_0 \gg N$, it is governed by the number of photons; the frequency vanishes for $n_0 = N$, revealing the unsatisfactory nature of the quasiclassical approximation. It is shown that one can use the anomalously high frequency of the supernutation, which appears in the $n_0 \ll N$ case, in the measurement of short relaxation times and oscillator strengths of transitions which are in resonance with the field. It is also shown that oscillations of the $n_0 < N$ type (with full modulation of the field) can be obtained in a cw gas laser by a sudden increase in the resonator Q factor, and nutational oscillations ($n_0 \gg N$) can be obtained by a sudden increase in the pump power.

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A monochromatic wave acting on a two-level system induces periodic oscillations of its population (nutation) of frequency[1–4]

$$\omega_1 = 2dE/\hbar = D\sqrt{n_0}, \tag{1}$$

where d is the matrix element of the dipole moment; E is the field intensity; n_0 is the number of photons in the resonator; $D = 4d(\pi\omega_0/\hbar V)^{1/2}$ (ω_0 is the frequency of light and V is the resonator volume). It is usually assumed that the field is sufficiently high and, therefore, its intensity is unaffected ($E \approx$ const) even if the wave interacts simultaneously with many atoms. This is indeed true under conditions in the usual nutation experiments when a high-power practically unattenuated light beam traverses a resonant medium.

However, if the interacting light beam and the medium form a closed system inside a high-Q resonator in the absence of pump radiation, both the frequency of energy exchange between the optical field and the atoms in the medium and the dynamics of the process may change considerably. This is due to the fact that, in a bounded volume, the initial number of photons n_0 and the nunber of atoms N are finite and the energy losses due to the excitation of atoms have a slight influence on the field amplitude, so that the usual estimate of the nutation frequency given by Eq. (1) remains valid only if $n_0 \gg N$. However, if $n_0 \leqslant N$, we cannot ignore the reaction of the medium on the field. This point has been mentioned before[5,6] but its consequences have not yet been fully worked out.

The chief consequence is that, if $n_0 < N$, the depth of modulation of the field is equal to the field intensity and, if $n_0 \ll N$, the frequency of energy exchange is proportional to the square root of the number of atoms and not of the number of photons: $\omega_1 = D\sqrt{N} \gg D\sqrt{n_0}$, i.e., it is considerably higher than one would expect on the basis of Eq. (1). This phenomenon, called supernutation in [7], can be observed by detecting the radiation leaking out of a resonator and it can be used, like the conventional nutation, in measuring the decay decrement of the process in the form of the relaxation times of the atoms.

We shall describe the state of a system of atoms using the concept of the energy (effective) spin R.[8] The energy of the system is $\mathscr{E}_a = \hbar\omega_0 R_3 = -\hbar\omega_0 R\cos\varphi$, where R is the maximum projection of the spin ($R \leq N/2$), R_3 is the projection of the spin on the "energy axis," φ is the angle between the spin direction and the energy axis, and ω_0 is the atomic transition frequency. It follows from the equations of motion of the spin in a monochromatic field, which is in resonance with an atomic transition,[4] that $\varphi = 2de/\hbar$ or

$$\dot{\varphi} = D\sqrt{n}, \tag{2}$$

where n is the number of atoms in the resonator at a given moment.

The law of conservation of energy

$$n + R_3 = n - R\cos\varphi = \text{const} \tag{3}$$

makes it possible to allow for the reaction of the atomic ensemble on the field, and Eqs. (2)–(3) can be used to obtain the following coupled system of equations:

$$\dot{\varphi} = Du, \quad \dot{u} = -{}^1/_2 DR\sin\varphi, \tag{4}$$

where $u = \sqrt{n}$. If the decay due to the finite value of the Q factor of the resonator is introduced into the system (4), this system transforms to the equations obtained by Faĭn.[9]

The system (4) is identical with the equations describing the motion of a physical pendulum. Solving these equations, we find the oscillation frequency as a function of the total energy of the system on condition that we initially have $n = n_0$ and that all the atoms are unexcited, i.e., we shall assume that $R = N/2$ and $\varphi(0) = 0$. We then obtain

$$\omega_1 = \begin{cases} \pi D\sqrt{n_0}/2K(\sqrt{N/n_0}), & n_0 > N \\ \pi D\sqrt{N}/2K(\sqrt{n_0/N}), & n_0 < N \end{cases} \tag{5}$$

Here, K is a complete elliptic integral of the first kind.

The dependence $\omega_1(n_0)$ found in this way is shown in Fig. 1. It is worth noting that the nutation frequency vanishes at $n_0 = N$. This result demonstrates the limited validity of the classical description of the field in the vicinity of the singularity $n_0 = N$. A consistent quantum calculation shows that ω_1 passes through a minimum in this region but does not vanish. This can be proved by considering a system composed of a quantum field oscillator and two atoms described by the Hamiltonian

$$\hat{H} = \omega_0(\hat{a}^+\hat{a} + \hat{R}_3) - {}^1/_2 D(\hat{a}^+\hat{R}_- + \hat{a}\hat{R}_+), \tag{6}$$

where $\hat{a}^+$ and $\hat{a}$ are the photon creation and annihilation operators; $\hat{R}_+$ and $\hat{R}_-$ are the raising and lowering operators. If we assume that, at t = 0, the oscillator is in the n_0-th excited state ($n_0 = 1, 2, 3, \ldots$), we obtain periodic oscillations with frequencies represented by circles in Fig. 1. At the minimum ($n_0 = N = 2$), we have $\omega_1 = D\sqrt{3/2}$.

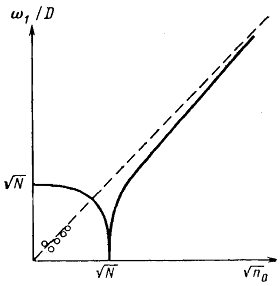

FIG. 1. Dependence of the nutation frequency on the initial number of photons in the resonator. The dashed line corresponds to $\omega_1 = 2dE/\hbar = D\sqrt{n_0}$.

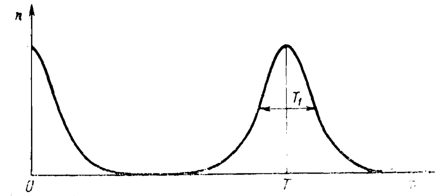

FIG. 2. Time dependence of the number of photons near the singularity; $\omega_1 = 2\pi/T$, $n_0/N = 0.99$, $T_1/T = 0.23$.

In the case of a system composed of three atoms or more, the solutions are no longer periodic for $n_0 \geq 3$.[10] This essentially quantum result remains valid also for macroscopically large numbers $N \approx n_0$, as also shown by Senitsky.[10] It is interesting to note that the quasiclassical approximation is of a "spiky" nature (Fig. 2). in the range $|n_0 - N| \ll N$. The profile of a spike is nearly identical with that of a 2π pulse (the McCall-Hahn solution[11]) and the frequency can be obtained from the general formula (5):

$$\omega_1 = \pi D N^{1/2} \ln^{-1}(16N/|n_0 - N|).$$

A similar expression was derived by Kazantsev and Smirnov[5] for the $(n_0 - N) > 0$ case (to the right of the singularity).

We shall now consider the limiting cases $n_0 \gtrless N$. If $n_0 \gg N$, we find that Eq. (5) gives the usual formula (1). This is supported also by a consistent quantum calculation of this case carried out in [12]. It also follows from Eq. (5) that, in the opposite case of $n_0 \ll N$ (weak oscillations), we obtain

$$\omega_1 = D\sqrt{N}, \tag{7}$$

i.e., the nutation frequency is much higher than that expected in the external field approximation (1) when $\omega_1 = D\sqrt{n_0}$. This excess of the oscillation frequency over the standard estimate (1), because of its dependence on the number of atoms, was called by us supernutation,[7] by analogy with superradiance.[8,13]

Supernutation, like ordinary nutation, is not affected by the nature (classical or quantum) of the treatment. In fact, small oscillations correspond to low degrees of excitation of the energy spin which then behaves as a harmonic oscillator.[14] Assuming in Eq. (6) that

$$\hat{R}_+ = (2R)^{1/2}\hat{b}^+, \quad \hat{R}_- = (2R)^{1/2}\hat{b}, \quad \hat{R}_3 = \hat{b}^+\hat{b} - R, \tag{8}$$

where $\hat{b}^+$ and b are the Bose operators, we can readily determine the frequency of energy exchange between two coupled oscillators, which is obviously governed by the change in the quantity $M = \hat{a}^+\hat{a} - \hat{b}^+\hat{b}$. In the Heisenberg representation, we obtain the equations of motion which are encountered also in the exciton theory:1)

$$\frac{d}{dt}\hat{M} = iD(2R)^{1/2}(\hat{a}^+\hat{b} - a\hat{b}^+), \quad \frac{d}{dt}(a^+\hat{b} - \hat{a}\hat{b}^+) = iD(2R)^{1/2}\hat{M}. \tag{9}$$

It is clear from these equations that the oscillation frequency is always $D(2R)^{1/2}$, i.e., it is twice the coupling constant and is independent of the initial conditions.

This conclusion, drawn from the quantum theory of radiation, is in exact agreement with the quasiclassical estimate (7) if we bear in mind that $R = N/2$. Moreover, in a linear description of an absorbing medium, which is equivalent to the description of Eq. (8), only this result can be obtained,[15] whereas, in reality, there is a stringent condition which limits the validity of the generality of the nonlinear approach: $n_0 \ll N$. This sets the upper limit to the power of the radiation that can cause supernutation.

Since, in the supernutation regime, an atomic system is only slightly excited, this system can be described—like the field—purely classically by the polarization induced in the medium. On the other hand, nutational oscillations are a typical example of a nonlinear reaction of the medium to the field, due to the finite extent of the energy spectrum of an atomic system, which must be quantized to discern the effect under consideration. Only in the exceptional case when the number of photons is exactly equal to the number of atoms is it necessary to quantize both matter and field to obtain the nonzero result.

As expected, in almost all the experiments involving detection or use of energy exchange (oscillations) between a field and atoms,[16] the number of photons is less than the number of active atoms. Nevertheless, the proportionality of the oscillation frequency to the field is clear evidence that these oscillations are nutational. It is not clear whether this is due to the considerable length of the pulses interacting with the atomic system. The continuous arrival of new photons in a sample prevents complete deactivation of the field oscillator and thus destroys the effect under consideration. Supernutation in an illuminated target can be expected only if the length of the exciting π pulse is considerably less than the thickness of the sample and the extinction in the sample is sufficiently high for complete absorption of the light in a layer comparable with the pulse length. In this case, supernutation is manifested not only by temporal but also by spatial oscillations of the optical energy density.

The experimental situation can also be reversed, as demonstrated recently by Feld et al.[17] when the population is first inverted by simultaneous excitation of all the atoms, and photons are generated in the system due to superradiance resulting from an adjacent transition. Although in this case we have the extremal situation in which the maximum number of photons n_0 is exactly equal to the number of active atoms, the radiation density rapidly decays because the system remains open and the oscillation regime is soon displaced to the left of the singularity into the supernutation region. This interpretation of the experiments of Feld et al., proposed by Emel'yanov and Klimontovich,[18] requires only that the supernutation frequency be higher than the time necessary for a photon to escape from the sample.

However, the most direct method for observing energy exchange oscillations between the field and the medium is the simple switching of a laser which induces transient processes. We shall analyze the dynamics of these processes in the Appendix, bearing in mind that they are of considerable interest as providing a means for measuring the relaxation times and oscillator strengths of the transitions resulting in laser action.

APPENDIX

We shall consider the possibility of a transition to nutational and supernutational oscillations from cw operation of a single-mode laser. A laser can be described in terms of the semiclassical theory by introducing relaxation terms in the system (4). We shall express the initial equations in terms of the variables $R_1 = -R\sin\varphi$ and $R_3 = -R\cos\varphi$:

$$\dot{R}_1 = DuR_3 - R_1/\tau, \tag{10a}$$

$$\dot{R}_3 = -DuR_1 - (R_3 - R_0)/\tau, \tag{10b}$$

$$\dot{u} = {}^1/_2 DR_1 - \nu u. \tag{10c}$$

The quantities R_0 and the relaxation times of the population τ introduced in these equations are governed not only by the relaxation processes in the medium but also by the action of the pump radiation. Moreover, it is assumed that the relaxation time of the polarization is also τ (molecular laser case) and ν is used to denote the reciprocal of the photon lifetime in the resonator.

An analysis of the stability of the cw solutions of the system (10) shows[19,20] that laser action is possible only if $\eta = D^2 R_0\tau/2\nu > 1$, where the steady-state values are

$$n_s = u_s^2 = (\eta - 1)/D^2\tau^2, \quad R_{1s} = R_0(\eta-1)^{1/2}/\eta,$$
$$R_{3s} = R_0/\eta, \quad R_s = (R_{1s}^2 + R_{3s}^2)^{1/2} = R_0/\eta^{1/2} \tag{11}$$

and the stimulated emission regime is unstable in the peninsular region shown in Fig. 3.

We shall now demonstrate that stable emission should change to nutation as a result of a steep increase in the pump power, i.e., an increase in the value of R_0. In fact, nutation represents harmonic oscillations of the vector $\mathbf{R}(t)$ in the presence of a field of constant amplitude and frequency $\omega_1 = Du$. A sudden change in R_0 (at a moment t_0) alters instantaneously the steady-state solutions of Eqs. (10a) and (10b):

$$R_{1s}' = (\eta-1)^{1/2} R_0'/\eta, \quad R_{3s}' = R_0'/\eta$$

(in this case, η is defined in terms of R_0), but $\mathbf{R}(0) = \mathbf{R}_S \neq \mathbf{R}_S'$ and nutational oscillations begin near $\mathbf{R}_S'$

$$R_1(t) = R_{1s}' + (R_{1s} - R_{1s}')\cos\omega_1 t + (R_{3s} - R_{3s}')\sin\omega_1 t,$$
$$R_3(t) = R_{3s}' + (R_{3s} - R_{3s}')\cos\omega_1 t - (R_{1s} - R_{1s}')\sin\omega_1 t. \tag{12}$$

Substituting the solution (12) by way of a trial in the system (10), we readily show that it satisfies this system provided

$$\omega_1 \gg \nu, \quad \omega_1 \gg 1/\tau, \quad u_s^2 \gg |R_0 - R_0'|\eta^{-1/2}.$$

In view of Eq. (11), these conditions are equivalent to the following:

$$\eta^{1/2} \gg \nu\tau, \quad \eta^{1/2} \gg 1, \quad \eta^{1/2}/2\nu\tau \gg |1 - R_0'/R_0|.$$

The first two conditions can be satisfied simultaneously only in the region identified by the horizontal shading in Fig. 3. The nutation frequency is $\omega_1 = \eta^{1/2}/\tau$.

Dynamic oscillations with full modulation of the field can be obtained as a result of a sudden increase in the resonator Q factor, i.e., by replacing ν with $\nu_1 < \nu$. We shall find the conditions under which such oscillations occur by rewriting the system (10) in terms of the variables φ, u, and R (replacing ν with ν_1):

$$\dot{\varphi} = Du + \frac{R_0}{R\tau}\sin\varphi, \quad \dot{u} = -\frac{DR}{2}\sin\varphi - \nu_1 u,$$
$$\dot{R} = -\frac{R + R_0\cos\varphi}{\tau}. \tag{13}$$

The required oscillations are those which hardly affect the relaxation processes and which are, in fact, described by the system (4) and not by (13). Thus, we must require that the relaxation terms in Eq. (13) affect only slightly the quantities u, φ, and R during one oscillation $1/\omega_1$, i.e., we must postulate the inequalities

$$\nu_1 \ll \omega_1, \quad |\dot{\varphi} - Du| = \frac{R_0}{R\tau}|\sin\varphi| \ll \omega_1,$$
$$\frac{|\dot{R}|}{R} = \frac{1}{\tau}\left|1 + \frac{R_0}{R}\cos\varphi\right| \ll \omega_1, \tag{14}$$

from which we can deduce—because $R_0/R(0) = R_0/R_S = \eta^{1/2} > 1$—only two inequalities: $\nu_1 \ll \omega_1$ and $\omega_1\tau \gg \eta^{1/2}$.

If these two conditions are satisfied, it readily follows from (4) that the frequency of dynamic energy oscillations

$$\omega_1 = \pi D(2R_s)^{1/2}/2K(\sqrt{n_0/2R_s}), \quad n_0 < 2R_s \tag{15}$$

differs from the frequency (5) by the replacement of N with

$$2R_s = 2R_0/\eta^{1/2}. \tag{16}$$

Moreover, n_0 (representing the number of photons in the resonator for $\varphi = 0$) is not arbitrary but is given by the initial conditions (11):

$$n_0 = R_s + R_{3s} + n_s = \frac{R_0}{\eta^{1/2}} + \frac{R_0}{\eta} + \frac{\eta - 1}{D^2\tau^2} = R_0\left[\frac{1}{\eta^{1/2}} + \frac{1}{\eta} + \frac{\eta - 1}{2\eta\nu\tau}\right]. \tag{17}$$

The condition for full modulation of the field $n_0 < 2R_S$, which is the condition of validity of Eq. (15), is evidently equivalent to the following relationship between the laser parameters before switching:

$$\nu\tau > {}^1/_2(1 + \eta^{1/2}). \tag{18}$$

We can easily show that this condition is definitely satisfied if the inequalities (14) are obeyed. In fact, using Eqs. (15) and (16) and $\eta = D^2R_0\tau/2\nu$, we find that, far from the singularity $n_0 = 2R_S$, the condition (14) reduces to

$$(\nu\tau)^{1/2} \gg \eta^{1/4}, \tag{19}$$

and hence we obtain Eq. (18).

Thus, oscillations with full modulation of the field (far from the singularity $n_0 = 2R_S$) can be obtained if $\nu_1 \ll \omega_1 \sim \eta^{1/4}(\nu/\tau)^{1/2}$ and the inequality (19) are satisfied. The condition (19) corresponds to the region with vertical shading in Fig. 3. It is important to note that these oscillations are of completely different origin from the oscillations with a slight deviation from steady-state conditions investigated by Kazantsev[21] at the limit of the instability region. In the oscillations under discussion here, the deviations are large: there is full modu-

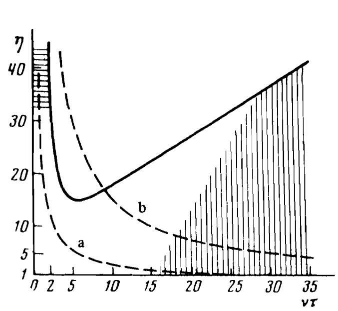

FIG. 3. Regions of unstable laser action, nutational oscillations, and oscillations accompanied by full modulation of the field. The hyperbolas a and b correspond to A = 50 and A = 300.

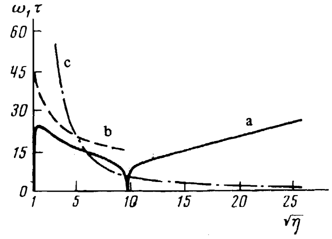

FIG. 4. Oscillation frequency ω_1, supernutation frequency $D\sqrt{2R_S}$, and reciprocal of the photon lifetime in a resonator ν plotted as a function of $\sqrt{\eta}$ for $A = 10^3$: a) $\omega_1\tau$; b) $D\tau\sqrt{2R_S}$; c) $\nu\tau$.

lation of the energy of not only the field but also of the atomic system.

The above treatment allows us to examine in detail the dependence of the frequency of the oscillations resulting from the switching of a laser (extra pulsations of the radiation [22]) on the initial laser characteristics. It is sufficient to note that there is a one-to-one correspondence between the quantities R_S and n_0, used to describe the oscillation frequency in Eq. (15), and the laser parameters $\nu\tau$ and η; this correspondence is established in Eqs. (16) and (17). Therefore, every point in Fig. 3 corresponds to a definite frequency because a formula similar to Eq. (15) also exists in the $n_0 > 2R_S$ case [Eq. (5), in which N is replaced with $2R_S$].

We shall now consider the change in the frequency in that section of the region where the solutions are defined and which corresponds to a change only in the initial Q factor of the resonator (i.e., to different values of ν) when all the other laser characteristics are constant:

$$\eta = A/2\nu\tau, \quad A = D^2\tau^2 R_0 = \text{const}, \quad \tau = \text{const}. \tag{20}$$

This section corresponds to the hyperbolas in Fig. 3. When the condition (19) is not obeyed (this means that $A^{1/2} \gg \eta^{3/4}$), it is not possible to have oscillations accompanied by full modulation of the field, so that we are only interested in the case when $A^{1/2} \gg 1$. In this case, the change in the frequency is of the form shown in Fig. 4.

A comparison of Figs. 1 and 4 shows that there are two zeros of ω_1 in the latter case. This is due to nonmonotonic variation of the ratio $n_0/2R_S$ when η is increased along the hyperbola (20). As long as the excess over the laser threshold is slight, the number of photons n_0 decreases faster than $R_S = R_0/\eta^{1/2}$, but then n_0 begins to fall more slowly and even to rise. Consequently, the oscillation regime is displaced from the singularity to the range of full modulation of the field and then returns through the singularity, approaching the usual nutational process.

Thus, the region between the two zeros of Fig. 4 correspond to full modulation of the field and the rectilinear asymptote $\omega_1 = \eta^{1/2}/\tau$ to the usual nutation. Almost throughout the region between the two zeros, the curve $\omega_1(\eta^{1/2})$ is close to its limit, which is the supernutation frequency $D(2R_S)^{1/2} = (2A)^{1/2}/\tau\eta^{1/4}$, which (in these coordinates) is not constant because of variation of R_S. Actual detection of oscillations of this frequency requires that the rate of dissipative processes be much lower than ω_1 and, since $\nu \geqslant \omega_1$, as can be seen from Fig. 4, a sudden increase in the Q factor is not only the means but the condition for observing the effect in question.

[1)]The authors are grateful to S. A. Moskalenko, who drew their attention to this point.

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