Quantum Dynamics of Molecular Nanomagnets in a Resonant Cavity and the Maser Effect

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Abstract

We study the dynamics of molecular nanomagnets through a fully quantum mechanical model describing high-spin and high-anisotropy magnetic molecules subjected to a time-dependent magnetic field along the quantization axis, which continuously inverts the population of spin states. Crystals of molecular nanomagnets placed inside a resonant cavity interact with a quantized electromagnetic field. Relaxation of excited states takes place by means of spin-photon interaction, allowing stimulated emission of radiation and creating a maser effect.

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I. INTRODUCTION

Recent interest on high-spin high-anisotropy molecular nanomagnets has grown significantly due to the spectacular magnetic effects they exhibit, namely the pronounced magnetic hysteresis and the quantization of the magnetic moment\(^1\). Potential applications of such magnets for current and future technologies include information storage, construction of nanomagnetic Maser-like devices, and quantum computation. The name Single-Molecule Magnets (SSM) has been coined to mean that individual molecules act as magnets. They can be prepared in long-lived excited quantum states by simply applying a magnetic field\(^2\), and exhibit a stepwise magnetic hysteresis in a time-dependent magnetic field. Quantum relaxation of spin states at low temperatures is very sluggish, and has been intensively studied within the framework of the Landau-Zener-Stueckelberg (LZS) effect\(^3\)–\(^5\).

A detailed study of quantum spin transitions and the effect of the environment in an experiment with a rotating magnetic field, was done in Ref. 6. Their approach follows the lines of the LZS theory, taking as the starting point the following hamiltonian\(^6\)–\(^8\):

\[
\mathcal{H} = -DS_z^2 - g\mu_B \mathbf{B} \cdot \mathbf{S},
\]

where \(D\) is the uniaxial anisotropy constant, \(g\) is the gyromagnetic factor, \(\mu_B\) is the Bohr magneton, \(\mathbf{B}\) is the magnetic field and \(\mathbf{S}\) is the molecular spin in units of \(\hbar\). In the absence of an applied field \(\mathbf{B}\), the ground state is double degenerate, corresponding to the states with parallel (\(m = S\)) and anti-parallel (\(m = -S\)) projections of the magnetic moment along the quantization axis. Throughout this paper, we will consider only these two states (\(m = -S\) and \(m = +S\)) as relevant to the problem\(^6\). In this context, a striking effect is predicted\(^9\), where molecular nanomagnetic crystals exhibit a giant magnetic relaxation due to Dicke superradiance of electromagnetic waves\(^10\). In recent electron spin resonance (EPR) experiments, it has been observed a pronounceable resonant absorption of electromagnetic radiation by molecular nanomagnets\(^11\)–\(^15\). In turn, they become a powerful source of coherent electromagnetic radiation when the wavelength of the emitted photons exceeds the linear size of crystals. When this condition is achieved, the molecules can coherently interact with the radiation emmitted, and the phase of the emitted photons may be considered the same throughout the sample\(^9\). Inside a resonant cavity, molecular magnets exhibit a strong dependence of the magnetization on the geometry of the cavity and this effect was observed experimentally, providing strong evidence for the coherent microwave radiation given off by
the crystals. These observations open the possibility of building nanomagnetic microwave lasers pumped by magnetic fields\textsuperscript{16}.

Our aim in this paper is to study the effect of stimulated radiation from molecular nanomagnetic crystals, by considering a fully quantum mechanical model, in which the electromagnetic radiation enters as a quantized field. The essential ingredients in our model include:

\begin{itemize}
  \item[i)] Hamiltonian (1) describing individual molecules, with a time-varying magnetic field $B_z(t)$ along the quantization axis. This way, the low and high energy states will be continuously changing with time, creating the effect of spin-states population inversion. Note that this is different from most models proposed in the literature, where the magnetic field $B_z$ is constant, and the time-dependent field is applied in a transverse direction;
  
  \item[ii)] a quantized electromagnetic field inside a resonant cavity, which allows the relaxation of excited states by means of coherent photon emission, enhancing the radiation field.
\end{itemize}

Finally, we will analyze the semiclassical limit, in which the photon-field can be treated as a classical electromagnetic field.

The above program will be developed in the present contribution. The content of this paper can be described as follows: in the next Section, we formulate the theoretical basis for analyzing spin dynamics, discussing the Hamiltonian. In Section III, we analyze the dynamics of a pure quantum mechanical state initially prepared in one of the double degenerate ground states of the system. We compute the correlation amplitude between the initial and the evolved state. Analytical approximated expressions are obtained to be compared with numerical results. In Section IV, we consider the photon field as a classical variable and obtain, in close analytical way, the conditions for a maser-like effect. Finally, in the last Section a few conclusions and remarks are added.

\section{Hamiltonian and Quantum Dynamics}

We start considering the two lowest-level states ($m = +S$ and $m = -S$) of spin Hamiltonian (1) and a second quantized term in photon variables describing the radiation
field in the cavity:

\[
\mathcal{H}_0 = -DS^2 \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} - g\mu_B S B_0(t) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \hbar \omega a^\dagger a \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix},
\]

(2)

where the Pauli matrix

\[
\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}
\]

yields the splitting of the levels in the presence of a time-dependent magnetic field \( B_0(t) \) applied along the quantization axis. The operator \( a^\dagger(a) \) is the photon creation (annihilation) operator. Inside a resonant cavity, only photons with a preselected frequency \( \omega \) can be emitted or absorbed. The basis of \( \mathcal{H}_0 \) is given through the kets \( \{|S, n >\} \) and \( \{|-S, n >\} \), being \( n \) the number of photons in a given state. We will consider that a general state can be written as follows:

\[
|\Psi > = \{ A_S |S > + A_{-S} |-S >\} \otimes \sum_{n=0}^{\infty} \alpha_n |n >
\]

The interaction between the molecular spin and the dipolar component of the electromagnetic radiation inside the cavity is considered as a perturbation:

\[
\mathcal{H}_I = -\hbar \Gamma \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} (a^\dagger + a),
\]

(3)

where the Pauli matrix

\[
\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}
\]

flip the molecule from one level to the other (with the emission or absorption of a photon). One can see that our model consists of a two level system coupled to a quantized harmonic oscillator. Despite the simplicity of the above mentioned model, no exact analytic solution is yet known\(^{17}\). Now, we consider the quantum dynamics in the interaction picture, taking into account that the non-perturbed (diagonal) hamiltonian \( \mathcal{H}_0 \) is time dependent. In this case, operators evolve according to the equation:

\[
\frac{i\hbar}{\partial t} \frac{\partial \mathcal{O}}{\partial t} = [\mathcal{O}, \mathcal{H}_0]
\]

and for the interaction Hamiltonian we get

\[
\mathcal{H}_I(t) = e^{iW(t)} \mathcal{H}_I e^{-iW(t)},
\]

(4)
being

\[ W(t) = \frac{1}{\hbar} \int_0^t \mathcal{H}_0(t') dt' \]  \hspace{1cm} (5)

The temporal evolution of an initial state ket \(|\Psi_0\rangle\) will be given by the unitary evolution operator \(U_I\) in the interaction picture. The latter can be written in the form of a Dyson series:

\[
U_I(t, 0) = 1 - i \frac{\hbar}{\Gamma} \int_0^t dt_1 e^{iW(t_1)} \mathcal{H}_I e^{-iW(t_1)} + \\
+ \left( -i \frac{\hbar}{\Gamma} \right)^2 \int_0^t dt_1 \int_0^{t_1} dt_2 e^{iW(t_1)} \mathcal{H}_I e^{-iW(t_1)} \int_0^{t_2} dt_3 e^{iW(t_2)} \mathcal{H}_I e^{-iW(t_2)} + \ldots ,
\]  \hspace{1cm} (6)

with \(|\Psi(t)\rangle = U_I(t, 0)|\Psi_0\rangle\). We can express the interaction hamiltonian \(\mathcal{H}_I\) in the form below:

\[
e^{iW(t)} \mathcal{H}_I e^{-iW(t)} = -\hbar \Gamma \begin{pmatrix} 0 & H_{12} \\ H_{21} & 0 \end{pmatrix},
\]  \hspace{1cm} (7)

being

\[
H_{12} = a^{\dagger} \exp \left[ i \left( \omega t - \frac{2g\mu_BS}{\hbar} \int_0^t B_0(t') dt' \right) \right] + \\
a \exp \left[ -i \left( \omega t + \frac{2g\mu_BS}{\hbar} \int_0^t B_0(t') dt' \right) \right]
\]  \hspace{1cm} (8)

\[
H_{21} = H_{12}^{\dagger} = a^{\dagger} \exp \left[ i \left( \omega t + \frac{2g\mu_BS}{\hbar} \int_0^t B_0(t') dt' \right) \right] + \\
+ a \exp \left[ -i \left( \omega t - \frac{2g\mu_BS}{\hbar} \int_0^t B_0(t') dt' \right) \right]
\]  \hspace{1cm} (9)

One must remember that \(H_{12}\) and \(H_{21}\) are infinite-dimensional in the Fock space of photons. We write \(U_I\) in the following form

\[
U_I = 1 + i \Gamma \int_0^t dt_1 \begin{pmatrix} 0 & H_{12}(t_1) \\ H_{21}(t_1) & 0 \end{pmatrix} - \\
- \Gamma^2 \int_0^t dt_1 \int_0^{t_1} dt_2 \begin{pmatrix} H_{12}(t_1)H_{21}(t_2) & 0 \\ 0 & H_{21}(t_1)H_{12}(t_2) \end{pmatrix} + \ldots
\]  \hspace{1cm} (10)

The above expressions admit any time-varying magnetic field \(B_0(t)\). For sake of convenience, we will restrict our attention to the sinusoidal dependence:

\[
B_0(t) = B_0 \cos(\Omega t),
\]  \hspace{1cm} (11)

and in this case (8) and (9) reduce to

\[
H_{12}(t) = a^{\dagger} \exp \left[ i \left( \omega t - r \sin(\Omega t) \right) \right] + a \exp \left[ -i \left( \omega t + r \sin(\Omega t) \right) \right],
\]  \hspace{1cm} (12)

\[
H_{21}(t) = a^{\dagger} \exp \left[ i \left( \omega t + r \sin(\Omega t) \right) \right] + a \exp \left[ -i \left( \omega t - r \sin(\Omega t) \right) \right],
\]  \hspace{1cm} (13)
where we have defined
\[ r = \frac{2g\mu_B S B_0}{\hbar \Omega}. \]  

The frequency \( \Gamma \) in \( \mathcal{H}_I \) represents the coupling with the dipolar field, and is small when the wavelength of the radiation field is far longer than the molecular dimension. In this case, first order perturbation theory applies, leading to:
\[ U_I = 1 + i\Gamma \mathcal{M}_1 + O(\Gamma^2) \]  

and
\[ \mathcal{M}_1 = \begin{pmatrix} 0 & a^\dagger F_1 + a F_2 \\ a^\dagger F_2^* + a F_1^* & 0 \end{pmatrix}. \]

The dynamics of the system will be governed by the functions \( F_1(t) \) and \( F_2(t) \):
\[
F_1(t) = \int_0^t dt_1 \exp[i(\omega t_1 - r \sin(\Omega t_1))]| = \frac{J_0(r)}{i\omega}(e^{i\omega t} - 1) + \\
+2 \sum_{m=0}^{\infty} \left[ \frac{J_{2m+2}(r)}{\omega^2 - (2m + 2)^2 \Omega^2} \Lambda_{1,m}(t) - i \frac{J_{2m+1}(r)}{\omega^2 - (2m + 1)^2 \Omega^2} \Lambda_{2,m}(t) \right],
\]
\[
F_2(t) = \int_0^t dt_1 \exp[-i(\omega t_1 + r \sin(\Omega t_1))]| = \frac{J_0(r)}{i\omega}(1 - e^{-i\omega t}) + \\
+2 \sum_{m=0}^{\infty} \left[ \frac{J_{2m+2}(r)}{\omega^2 - (2m + 2)^2 \Omega^2} \Lambda_{1,m}^*(t) - i \frac{J_{2m+1}(r)}{\omega^2 - (2m + 1)^2 \Omega^2} \Lambda_{2,m}^*(t) \right],
\]

being \( J_{2m+1}(r) \) and \( J_{2m+2}(r) \) the Bessel functions of first kind, and
\[
\Lambda_{1,m}(t) = i\omega - i\omega e^{i\omega t} \cos[(2m + 2)\Omega t] - (2m + 2)\Omega e^{i\omega t} \sin[(2m + 2)\Omega t]
\]
\[
\Lambda_{2,m}(t) = -i\omega e^{i\omega t} \sin[(2m + 1)\Omega t] + (2m + 1)\Omega e^{i\omega t} \cos[(2m + 1)\Omega t] - (2m + 1)\Omega.
\]

From the above formulae, a resonant behavior occurs when the photon frequency \( \omega \) is an integer multiple of \( \Omega \), say \( \omega = m\Omega \), and the dominant behavior of \( F_1 \) and \( F_2 \) near that condition is given by:
\[
F_1 \approx \frac{J_m(r)[(-1)^m i(1 - e^{2i\omega t}) + 2i\omega]}{2\omega} \]
\[
F_2 \approx (-1)^m F_1^* = (-1)^m \frac{J_m(r)[(-1)^m i(1 - e^{-2i\omega t}) + 2i\omega]}{2\omega}
\]

In the next Section we will analyze the dynamics of a pure quantum mechanical state initially prepared as an eigenstate of \( \mathcal{H}_0 \).
III. DYNAMICS OF QUANTUM STATES

To fix ideas, consider a quantum state prepared in the magnetic state \( |S> \) of the molecule, in the form:

\[
|\Psi_0> = |S> \sum_{n=0}^{\infty} \alpha_n |n>
\]

where the mean number of photons at \( t = 0 \) is given by

\[
n_0 = \langle a^\dagger a \rangle_0 = \sum_{n=0}^{\infty} n |\alpha_n|^2.
\]

To first order perturbation theory, the evolved state \( |\Psi(t)> = U_I |\Psi_0> \) reads

\[
|\Psi(t)> \approx \frac{1}{\sqrt{1 + \Gamma^2[(n_0 + 1)|F_2|^2 + n_0|F_1|^2]} \times \left[ |\Psi_0> + i\Gamma |S> + \sum_{n=0}^{\infty} \alpha_n \left( F_2^* \sqrt{n + 1} |n + 1> + F_1^* \sqrt{n} |n - 1> \right) \right].
\]

Observe that we have renormalized the evolved state. The correlation probability function defined as \( C(t) = |\langle \Psi_0 |\Psi(t) >|^2 \), which is simply the probability of finding the system in the initial state at later times, yields

\[
C(t) = \frac{1}{1 + \Gamma^2[(n_0 + 1)|F_2|^2 + n_0|F_1|^2]},
\]

and the above expression can be approximated at the resonant condition \( \omega = m\Omega \) by the behavior of \( F_1 \) and \( F_2 \) given in (21) and (22):

\[
C(t) \approx \frac{1}{1 + \Gamma^2|F_1|^2(2n_0 + 1)},
\]

with

\[
|F_1|^2 = \frac{|J_m(r)|^2}{4\omega^2} \left( 4\sin^2 \omega t + 4t\omega \sin 2\omega t + 4\omega^2 t^2 \right).
\]

For long times \( (t\omega \gg 1) \), the leading behavior is quadratic in time

\[
|F_1|^2 \approx |J_m(r)|^2 t^2,
\]

and we can approximate the above expression as follows:

\[
C(t) \approx \frac{1}{1 + t^2/\tau^2},
\]

with the correlation time \( \tau \) defined as:

\[
\frac{1}{\tau} = \Gamma |J_m(r)|\sqrt{2n_0 + 1}.
\]
The value $1/\tau$ can be interpreted as the rate of emission of photons, since the nanomagnet will relax to lower energy states by emitting photons. For large values of the constant $r$, maxima of the transition rate will be given by $r = 2gS_{\mu B}B_0/(\hbar \omega/m) \sim \zeta \pi$, with $\zeta \gg 1$ a constant depending on the order of the Bessel function, yielding

$$\left(\frac{1}{\tau}\right)_{\text{max}} \approx \Gamma \sqrt{\frac{\hbar (\omega/m)}{\pi gS_{\mu B}B_0}} \sqrt{2n_0 + 1}.$$

Next, we show some examples. To illustrate the evolution of a quantum state, we consider the initial mean number of photons $n_0 = 0$, i.e. the vacuum for the cavity field, and the molecular spin in the $S$ state (in this case the initial state is $|\Psi_0 > = |S, 0 >$). The resonant cavity is adjusted to the frequency $f = \omega/2\pi = 10$ GHz and the spin-photon interaction constant is taken as $\Gamma = 2$ GHz. In Fig. 1 to 3, we show the correlation probability $C(t)$ as a function of time, for given values of $r$ and $m = \omega/\Omega$. At a given ratio $m = \omega/\Omega$, the parameter $r$ was set to give the maximum value of the Bessel function $J_m(r)$. The parameters for the first two figures satisfy the resonant condition ($m$ an integer). One can see that the behavior for $m$ even or odd is qualitatively the same. Except for the stepwise character of the correlation function, $C(t)$ can be estimated by the expression (27), with the relaxation time given by (28). Note the irreversible character of $C(t)$ at the resonance, due to the relaxation of the molecule by successive photon emissions.

A quite different situation occurs for a non-integer value of the ratio $\omega/\Omega$, as shown in Fig. 3 for $m = 9, 31$. There are revivals of the initial state at latter times, in the form of an absorption-emission cycle not completely periodic.

The above results were obtained within first order perturbation theory. Consequently, the long-time behavior should include corrections due to multiple photon processes. However, we believe that the essential physics is contained in the results discussed above. To support this view, in the next section we analyze the radiation field using the semiclassical approximation. We predict a masser effect at resonances.

IV. SEMICLASSICAL THEORY

The semiclassical theory is based on two fundamental assumptions: i) the cavity radiation field is described by a coherent state, which is the most nearly classical state (it minimizes the uncertainty relations); ii) the total density matrix is written as a product,
whose factors are related to spin and photon degrees of freedom. In other words, we assume that spin variables are uncorrelated with those of the field (Sargent). For a coherent state $|z\rangle$, we have the results

$$a |z\rangle = z |z\rangle,$$

$$\langle z| a^\dagger = \langle z| z^*,$$

$$\langle n\rangle = |z|^2.$$ (29)

(30)

(31)

So, the semiclassical approximation can be thought to be obtained by replacing the photon operators by complex numbers. Choosing the amplitude of the coherent state as real, we get $a^\dagger, a \rightarrow \sqrt{\langle n\rangle}$. Then the equation of motion for the mean number of photons $\langle n\rangle$ will be given by:

$$\frac{dA}{dt} = -\Gamma \sin(\omega t)[\rho_{12}e^{ir\sin(\Omega t)} + \rho_{21}e^{-ir\sin(\Omega t)}]$$

with $A = \sqrt{\langle n\rangle}$ The density matrix $\rho$ of the spin system satisfies the following set of coupled equations:

$$\frac{\partial M}{\partial t} = 4i\Gamma[\rho_{21}e^{-ir\sin(\Omega t)} - \rho_{12}e^{ir\sin(\Omega t)}]A \cos(\omega t)$$

$$\frac{\partial \rho_{12}}{\partial t} = -2i\Gamma e^{-ir\sin(\Omega t)} MA \cos(\omega t)$$

$$\frac{\partial \rho_{21}}{\partial t} = 2i\Gamma e^{ir\sin(\Omega t)} MA \cos(\omega t),$$

where $M = \rho_{11} - \rho_{22}$. In general the solution must be accomplished by numerical methods. A simple solution is obtained if one assumes $\rho_{12} = \rho_{21}^* = iN$, being $\rho_{12} - \rho_{21} = 2iN$, i.e. the off-diagonal elements depends on a single real parameter. It can be shown that the other possibility, $\rho_{12} = \rho_{21} = R$ does not lead to a maser effect. For the former case we have:

$$\frac{\partial M}{\partial t} = 8\Gamma N A \cos(r \sin(\Omega t)) \cos(\omega t),$$

$$\frac{\partial N}{\partial t} = -2\Gamma MA \cos(r \sin(\Omega t)) \cos(\omega t),$$

$$\frac{dA}{dt} = 2\Gamma N \sin(r \sin(\Omega t)) \sin(\omega t).$$

An approximate solution near the values $A_0 \approx 0, N_0$ and $M_0$ for the photon field $A$ is easily obtained yielding:

$$A(t) = A_0 + \Gamma N_0 \text{Re}(F_1 - F_2)$$
In the case of $\omega = (2m + 1)\Omega$ the above solution for $A$ grows linearly with time:

$$A(t) \approx A_0 + 2\Gamma N_0 J_{2m+1}(r) \left( t - \frac{\sin(2\omega t)}{2\omega} \right)$$  \hspace{1cm} (39)$$

and consequently, the number of photons increases quadratically with time. In contrast, for $\omega = (2m + 2)\Omega$ the approximate solution is nearly constant, with $A(t) \approx A_0$, displaying ripples around this value. This even-odd symmetry breaking at resonance, is at variance with the pure quantum case treated in the previous section, where both instances presented similar behaviors at resonance, i.e. an increasing of the photon number due to relaxation of the molecular states, no matter if $m$ was even or odd. In any case, for the semiclassical approximation, a maser effect is predicted for $\omega = (2m+1)\Omega$. To illustrate this phenomenon, we show in Figures 4 to 7, the photon number as a function of time, for some temperatures. In Fig. 4 and 5, we take the limit $k_B T \to 0$, and the initial photon number $n_0 \to 0(A_0 \to 0)$. Other parameter values are $M_0 = 0$ and $N_0 = 0.5$. In Fig. 4, we have chosen $m = 9$ and $r = 10$, while for Fig. 5, $m = 10$ and $r = 12$ (the corresponding value of $r$ is always chosen to maximized the Bessel function). From Fig. 4, it is clear that the overall behavior of the photon number for $m$ odd is closely related to the solution given by expression (39), and shown by the dashed line, corresponding to a quadratic increase of the number of photons with time. In contrast, when $m$ is even, there is an oscillatory behavior, at least for short times, which reminds the quantum revivals. Figures 6 and 7 refer to examples at finite temperatures for $m$ odd. For Fig. 6, $T = 24$ K, corresponding to the initial value $n_0 = k_B T/(\hbar \omega) = 49$ for the number of photons, while Fig. 7 is at room temperature ($T = 300$K), corresponding to $n_0 = 625$ photons. Note that the higher the initial number of photons, the faster the photon number increases with time. The effect is paramount in the latter case. Note also the different time scale.

It practice, the photon number will increase until a saturation limit. The divergent behavior here obtained is due to the fact that losses are not taken into account. Such losses are provided by photons leaving the cavity and by excitation of other energy states. In fact, the real system which we are concerned here, is not as simple as a two-level object. In addition, the cavity field should include contributions from other photon modes, which we neglected from the beginning.
V. CONCLUSIONS

We have developed the quantum dynamics of molecular nanomagnets, taking into account the quantized photon field of the cavity. In first approximation, the magnetic molecules were treated as two-level systems. The temporal evolution of quantum states were studied in details in this paper, considering a time-varying magnetic field \( B_z(t) = B_0 \cos(\Omega t) \), applied along the quantization direction. The above field produces the necessary population inversion in a periodic way.

The spin system couples with the dipolar component of the cavity field, allowing transitions between both molecular states with photon emission and absorption. The emission of photons is enhanced at the resonant condition, when the ratio between photon frequency \( \omega \) and the applied field frequency \( \Omega \) is an integer number \( m = \omega / \Omega \). At resonance, the energy is pumped from the applied magnetic field to the cavity radiation field, inducing a relaxation process depicted in Fig. 1 and 2. This irreversible process can be understood, if one considers that the probability of creating a photon increases with the number of photons. This striking phenomenon was illustrated in the previous section, via the semiclassical theory. In turn, the case of non-integer \( m \) produces a revival of the initial state, which means that emission and absorption of photons occur in almost periodic sequences, very similar to Rabi periods.

Qualitative insights can be obtained via the semiclassical approximation, which is more suitable for practical purposes at room temperatures. Within this theory, the macroscopic state of the cavity field is described by the coherent states introduced by Glauber\(^{19} \). In this case, a maser effect is obtained at resonance, when \( m \) is an odd number. The maser effect is enhanced by temperature, when the initial photon number is increased.
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Figure Captions

Figure 1: Correlation function $C(t)$ as a function of time, considering $\Gamma = 2 \times 10^9 \text{ s}^{-1}$, $\omega = 2\pi \times 10^{10} \text{ rad/s}$, $\Omega = \omega/m$, $m = 8$ and $r = 9.64$.

Figure 2: Correlation function $C(t)$ with parameters $\Gamma$ and $\omega$ kept the same as for the previous figure, $m = 11$ and $r = 11.94$.

Figure 3: Correlation function $C(t)$ for a non-integer ratio $\omega/\Omega$. In this case we have chosen $m = 9.31$ and $r = 11.45$.

Figure 4: Number of photons for the field $A(t) = \sqrt{\langle n \rangle}$ in the semiclassical approximation using $f = \omega/2\pi = 10$ GHz, $\Gamma = 1$ GHz, $M_0 = 0$, $N_0 = 0.5$, $n_0 = 0$, $m = 9$ and $r = 10$.

Figure 5: Number of photons for the field $A(t) = \sqrt{\langle n \rangle}$ in the semiclassical approximation using $f = \omega/2\pi = 10$ GHz, $\Gamma = 1$ GHz, $M_0 = 0$, $N_0 = 0.5$, $n_0 = 0.25$, $m = 10$ and $r = 12$.

Figure 6: Number of photons for the field $A(t) = \sqrt{\langle n \rangle}$ in the semiclassical approximation with $f$, $\Gamma$, $M_0 = 0$ and $N_0 = 0.5$ kept the same as the previous case, $n_0 = 49$, $m = 9$ and $r = 10$.

Figure 7: Number of photons for the field $A(t) = \sqrt{\langle n \rangle}$ in the semiclassical approximation using $n_0 = 625$, $m = 5$ and $r = 6.5$. Other parameters kept the same as for the previous figures.
$C(t) = \left| \langle \Psi_0 | \Psi(t) \rangle \right|^2$

Normalized Time ($T = \Omega t / 2\pi$)
$C(t) = |\langle \Psi_0 | \Psi(t) \rangle|^2$

Normalized Time ($T = \Omega t / 2\pi$)
Number of Photons $n(t) = [A(t)]^2$ vs. $t$(ns)
Number of Photons $n(t) = [A(t)]^2$
