Ringing in the resonance fluorescence spectrum of a driven two-level atom under bichromatic excitation in a broadband squeezed vacuum

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We present analytical results for the steady-state resonance fluorescence spectrum produced by a driven two-level atom excited under two strong coherent fields in the presence of a broadband squeezed vacuum. Here the central frequency of the squeezed vacuum is considered to be resonant as well as off-resonant with the average driving field frequency while maintaining resonance between the atomic transition frequency and the average driving field frequency. We observe a ringing phenomenon in the fluorescence spectrum due to the small frequency difference in the bichromatic field and this phenomenon is found to be sensitive to the detuning of the carrier frequency of squeezed vacuum with respect to the central frequency of the bichromatic field. [S1050-2947(98)00810-5]

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In the recent past, certain radiative properties of one and many two-level atoms embedded in a broadband squeezed bath have been the topic of keen investigation in quantum optics. Among them the most interesting prediction is that a two-level atom strongly driven by a coherent field and interacting with a broadband squeezed vacuum bath may exhibit a narrowing (subnatural linewidth) or broadening of the central peak of the usual three-peaked Mollow spectrum of resonance fluorescence, depending on the relative phase of driving field and squeezed vacuum [1]. In the studies carried out in Refs. [1–6] it is assumed that the frequency ω_L of the driving field is exactly equal to the carrier frequency of the squeezed vacuum. When the central (carrier) frequency of the broadband squeezed vacuum is not in resonance with both the driving field frequency ω_L and the atomic transition frequency ω_a, the analysis of first harmonic quadrature components of both the absorption spectrum and the fluorescent intensity is presented in [7]. Specifically, the absorption profile has a hole burning structure in the weak field case, and two unequal absorption or amplification peaks in the strong field case [7]. Recently, some results were reported on the resonance fluorescence spectrum of a driven two-level atom in an off-resonant squeezed vacuum [8,9]. In order to explore further new features in the spectral properties of the emitted fluorescence field we consider here a system of two-level atom (transition frequency ω_a) coherently driven by a bichromatic laser field (frequency components $\omega_1=\omega_a-\delta$ and $\omega_2=\omega_a+\delta$) in a broadband squeezed vacuum whose central frequency may not be at resonance with either the average driving field frequency [$\omega_s=(\omega_1+\omega_2)/2$] or the atomic transition frequency [10]. Because of the nonzero detuning of the squeezed vacuum field as well as the presence of the bichromatic driving field the model Bloch equations have time-dependent periodic coefficients, so they do not have an analytic solution in general [7–10]. Here we show that the analytic solution of the model Bloch equations under bichromatic excitation is possible in the intense field approximation [3]. However, the steady-state fluorescent spectrum so obtained is quite sensitive to the value of δ as well as on the detuning of the central frequency of the squeezed vacuum with respect to the central frequency of the driving field. The small frequency difference δ in the bichromatic fields gives rise to the phenomenon of ‘‘ringing,’’ i.e., an oscillatory behavior or a train of pulselike features in the fluorescence spectrum of a strongly driven two-level atom. This spectrum is also sensitive to the relative phase of the squeezed vacuum field. It is both qualitatively as well as quantitatively different from the fluorescent spectrum obtained at $\delta=0$ and at the exact resonance [1–3] condition of the central driving field frequency with the frequency of the squeezed vacuum field.

In an ordinary vacuum (without squeezing) the observed resonance fluorescent spectrum of a two-level-like Ba atom under intense bichromatic field excitation consists [11] of many sidebands separated by δ and the number of sidebands increases with increase in Rabi frequency (Ω_0) (the dressed-state analysis of this spectrum is given in [12]). Here we show that, in the squeezed vacuum case, under the condition of strong driving field ($\Omega_0 \gg \gamma, \delta$) we observe ringing behavior in the usual Mollow triplet for the above system. This ringing is sensitive to the detuning of the carrier frequency of the squeezed vacuum with respect to the average frequency (ω_s) of the driving fields. To quantify this phenomenon we give an analytic expression for the steady-state fluorescence spectrum.

We assume a broadband squeezed vacuum centered around ω_{sv}. The squeezing bandwidth is much broader than the spectral width of fluorescence so that the fluorescent spectrum falls well within the bandwidth of the squeezed vacuum modes. Then the squeezed vacuum appears as δ-correlated squeezed white noise to the atom [1]. The master equation for this system can readily be obtained [1–3] in a frame rotating at the driving field frequency ω_L:

$$\begin{aligned}\dot{\rho}=&i[H,\rho]-\frac{\gamma}{2}(N+1)(S_+S_-\rho+\rho S_+S_- -2S_-\rho S_+)\\&-\frac{\gamma}{2}N(S_-S_+\rho+\rho S_-S_+-2S_+\rho S_-)\\&-\gamma|M|e^{i\phi_s}e^{-2i(\omega_{sv}-\omega_s)t}S_+\rho S_+\\&-\gamma|M|e^{-i\phi_s}e^{2i(\omega_{sv}-\omega_s)t}S_-\rho S_-,\end{aligned}\tag{1}$$

with

$$H=\tfrac{1}{2}\Delta S_z+\Omega_0[S_+e^{i\phi_L}\cos(\delta t)+\text{H.c.}], \tag{2}$$

where γ is Einstein's A coefficient, and Ω_0 and ϕ_L are, respectively, the Rabi frequency and the phase of the driving field (whose two amplitudes are equal). N and M are the squeezed vacuum parameters with $|M|\leqslant\sqrt{N(N+1)}$, $\Delta=\omega_s-\omega_a$, ω_a is the atomic Bohr frequency, ω_{SV} is the central frequency of the broadband squeezed vacuum, and ϕ_s is the phase of the squeezed vacuum.

With the help of the master equation (1) we can readily obtain the model Bloch equation in the following form:

$$\begin{aligned}\langle\dot{S}_+\rangle&=-\left(\frac{\gamma}{2}(2N+1)+i\Delta\right)\langle S_+\rangle-\gamma M\\&\quad\times\exp(2ik\delta t)\langle S_-\rangle-2i\Omega_0\cos(\delta t)\langle S_z\rangle,\\\langle\dot{S}_-\rangle&=\langle\dot{S}_+\rangle^*,\\\langle\dot{S}_z\rangle&=-\frac{\gamma}{2}-\gamma(2N+1)\langle S_z\rangle+(i\Omega_0\langle S_-\rangle\\&\quad-i\Omega_0\langle S_+\rangle)\cos(\delta t).\end{aligned} \tag{3}$$

Here the parameter $k=(\omega_{\text{SV}}-\omega_s)/\delta$ is the measure of detuning of the squeezed vacuum carrier frequency ω_{SV} from the central frequency ω_s of the driving field. So $k=0$ implies that the squeezed vacuum is centered on the central frequency, $k=1$ means the squeezed vacuum centered on the first odd harmonic, $k=2$ means that it is centered on the first even harmonic of δ, and so on [10].

Note that within the same model Eq. (1) and for exact atomic detuning $(\Delta=0)$, the authors of Ref. [10] have investigated the nonoscillatory component of the steady-state fluorescent (scattered) spectrum numerically in the cases where ω_{SV} is close tuned to the central spectral line as well as to the first odd and the first even sideband, respectively. Their main interesting result is the occurrence of an anomalous spectral feature such as hole burning and dispersive profiles in the central peak which can easily be observed because of the absence of coherent scattering at line center under bichromatic excitations. What we are going to present here is the analytic expression of the complete fluorescent spectrum (including all harmonics) from the model Bloch equation (3) for the exact atomic detuning condition $(\Delta=0)$. To simplify our model equations to a great deal we restrict ourselves to the case of strong field $(\Omega_0\gg\gamma,\delta)$ [3,9] so that we can study the underlying features analytically. For this purpose we transform [3,9] the atomic operators as follows:

$$\begin{aligned}R_x&=S_y,\\R_y&=-(\Delta/2\Omega)S_x+(\Omega_0/\Omega)S_z,\\R_z&=(\Omega_0/\Omega)S_x+(\Delta/2\Omega)S_z,\end{aligned} \tag{4}$$

where $\Omega^2=\Omega_0^2+\Delta^2/4$. Next we go to the interaction picture defined by

$$\begin{aligned}\overline{\psi}&=e^{2i\Omega tR_z}\psi,\\\overline{\psi}&=\text{Col}[\langle\overline{R}_+\rangle,\langle\overline{R}_-\rangle,\langle\overline{R}_z\rangle],\\\psi&=\text{Col}[\langle R_+\rangle,\langle R_-\rangle,\langle R_z\rangle].\end{aligned} \tag{5}$$

With this transformation, the resulting equations split into two parts: one containing non-rapidly-oscillating terms and the other involving factors like $\exp(\pm2i\Omega t)$, $\exp(\pm4i\Omega t)$, etc. If we make the secular approximation at this stage (e.g., $\Omega\gg\gamma,\delta$) and neglect the rapidly oscillating terms and then revert back to the Schrödinger picture, we obtain

$$\begin{aligned}\langle\dot{R}_+\rangle&=\{2i\Omega[r+(1-r)\cos(\delta t)]-\gamma_+\}\langle R_+(t)\rangle,\\\langle\dot{R}_z\rangle&=-\gamma_0\langle R_z\rangle-\sqrt{r}\gamma/2,\end{aligned} \tag{6}$$

where

$$\begin{aligned}\gamma_+&=(\gamma/2)[(2N+1)(3-r)/2-|M|(1-r)\cos(2k\delta t+\Phi)],\\\gamma_0&=\gamma[(2N+1)(1+r)/2+|M|(1-r)\cos(2k\delta t+\Phi)],\\r&=\Delta^2/4\Omega^2\end{aligned} \tag{7}$$

in which $\Phi=2\phi_L-\phi_s$ (ϕ_L is the phase of the external driving field, and ϕ_s is the phase of the squeezed vacuum field).

The solution of Eqs. (6) can be obtained very easily. The fluorescent spectrum is given as the Fourier transform of the two-time correlation function of the atomic dipole operators

$$S(D)=\frac{2}{T}\,\text{Re}\left(\int_0^T dt\int_0^{T-t}d\tau\, e^{-iD\tau}\langle S_+(t+\tau)S_-(t)\rangle\right), \tag{8}$$

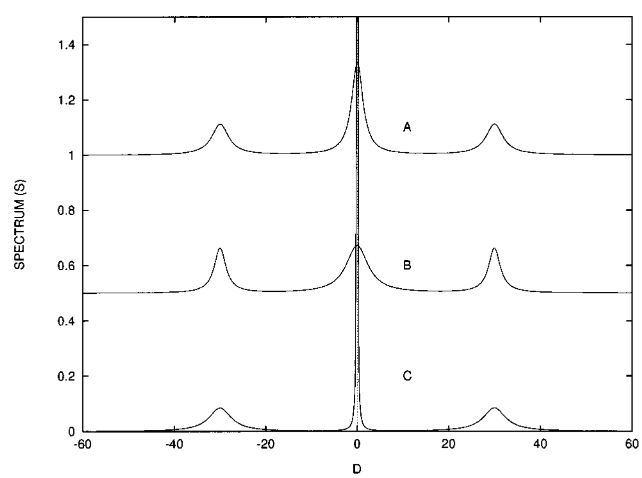

FIG. 1. Steady-state fluorescent spectrum S (in arbitrary units) of a two-level atom under monochromatic excitation in thermal (curve A) and squeezed (curves B and C) baths with driving field in resonance with the atomic transition $(\Delta=0)$. Here $2\Omega_0/\gamma=30$ and $D=(\omega-\omega_a)/\gamma$. Curve A $(S+1.0)$ is for $N=1$, $|M|=0$; curve B $(S+0.5)$ is for $N=1$, $|M|=\sqrt{[N(N+1)]}$, $\Phi=0$, and curve $C(S)$ is for $N=1$, $|M|=\sqrt{[N(N+1)]}$, $\Phi=\pi$.

where $D=\omega-\omega_a$ is the frequency offset of the (Fabry-Pérot) detector line center with respect to the atomic frequency ω_a, and the time T is the integrating time of the detector. We have to set $T\to\infty$ in the above expression (8) to get the steady-state fluorescent spectrum. In Eq. (8) we need to calculate the two-time averages $\langle S_+(t+\tau)S_-(t)\rangle$ which in fact can easily be obtained using quantum regression theorem. The correlation function appearing in Eq. (8) can then be expanded in R operators and making use of Eq. (6) we get the steady-state spectrum

$$S(D)=\frac{1}{4}\sum_{n=-\infty}^{\infty} I_n\left(\frac{b}{4k\delta}\right)e^{-(b/4k\delta)\sin(\Phi)}\times\sum_{m=-\infty}^{\infty} J_m\left(\frac{2\Omega_0}{\delta}\right)\frac{(3a/4)\cos[n(\Phi-\pi/2)]+[D+(m-2kn)\delta]\sin[n(\Phi-\pi/2)]}{(3a/4)^2+[D+(m-2kn)\delta]^2}+\frac{1}{4}\sum_{n=-\infty}^{\infty} I_n\left(\frac{b}{4k\delta}\right)e^{-(b/4k\delta)\sin(\Phi)}\times\sum_{m=-\infty}^{\infty} J_m\left(\frac{2\Omega_0}{\delta}\right)\frac{(3a/4)\cos[n(\Phi-\pi/2)]+[D-(m+2kn)\delta]\sin[n(\Phi-\pi/2)]}{(3a/4)^2+[D-(m+2kn)\delta]^2}+\frac{1}{2}\sum_{n=-\infty}^{\infty} I_n\left(\frac{-b}{2k\delta}\right)e^{(b/2k\delta)\sin(\Phi)}\frac{(a/2)\cos[n(\Phi-\pi/2)]+(D-2kn\delta)\sin[n(\Phi-\pi/2)]}{(a/2)^2+(D-2kn\delta)^2} \tag{9}$$

in which $a=\gamma(1+2N)$, $b=\gamma|M|$ and for simplicity we have kept atomic detuning $\Delta=0$ (or $r=0$). Also, in Eq. (9) there appears the Bessel function $J_m(x)$ and the modified Bessel function of first kind $I_n(x)$. The limiting case of $\delta\to0$ has to be taken properly to recover the results of the monochromatic excitation case.

The expression of $S(D)$ [in Eq. (9)] contains two types of terms. For the first category of terms in which $m=0$, $n=0$ (the part around the central peak of the Mollow triplet), only the magnitude of the spectral features is a function of δ but the positioning of the spectral feature is independent of δ. Also, the spectral shape for this category of terms is independent of the relative phase Φ between the driving field and the squeezed vacuum field. It is not surprising to have such terms as these terms represent the zero order (nonoscillatory component) of the fluorescent spectrum. The complete harmonics of the fluorescent spectrum are contained in the second category of terms for which $n\neq0$, $m\neq0$ in general. For these terms both the magnitude as well as the position of the spectral features are dependent on the parameter δ. Also, there appears the relative phase Φ in these terms. It implies that under the strong driving field condition with bichromatic excitation ($\delta\neq0$) the fluorescent spectrum is sensitive to the relative phase between the driving field and the squeezed vacuum field. The authors of Ref. [10] have also presented similar results numerically for large δ values. Our analytic result, Eq. (9), contains information about all the harmonics present (under intense field limit) in the fluorescence spectrum and thus reveals certain spectral features not mentioned in earlier studies under the secular approximation. We will now elaborate our results with the help of figures.

We will first demonstrate how good our secular approximation is under the strong driving field with monochromatic excitation ($\delta=0$). For this purpose, in Fig. 1, we have plotted the steady-state fluorescent spectrum for a broadband squeezed bath with the driving field $2\Omega_0/\gamma=30$, atomic detuning $\Delta/\gamma=0$, and monochromatic excitation condition $\delta/\gamma=0$. Here we have kept $|M|=0$, $N=1$, i.e., only a thermal bath for curve A, but a broadband squeezed bath ($N=1$, $|M|=\sqrt{[N(N+1)]}$) for both curves B and C, along with $\Phi=0$, for curve B and $\Phi=\pi$, for curve C. Clearly, the spectral features are in a very good agreement with previously reported results [1–3], e.g., when $\Phi=0$ the central peak broadens and equality of heights of the three peaks occurs, but when $\Phi=\pi$ the central peak narrowed down considerably and its height increases enormously. After establishing these well-known results obtained with our method, we next proceed for the $\delta\neq0$ (bichromatic excitation) case. However, we select δ very small in order to maintain the validity of our secular approximation and thus obtain some different spectral features. In Fig. 2 we have depicted the steady-state fluorescence spectrum for a broadband squeezed vacuum ($N=1$, $|M|=\sqrt{[N(N+1)]}$, $\Phi=0$) with $2\Omega_0/\gamma=30$, $\Delta/\gamma=0$, $\delta/\gamma=0.3$. Curve A is for $k=0$, i.e., the squeezed vacuum's carrier frequency is in resonance with the central frequency of the driving field. Comparing Fig. 2 (curve A) with Fig. 1 (curve B) essentially brings out the effect of bichromatic excitation (in which the difference in frequencies of the two excitation fields is kept very small) over the monochromatic excitation. What we observe is a kind of ringing phenomenon in between the central peak and the side peak region of the resonance fluorescence spectrum. For intense driving fields we can observe such ringing only at very low δ's. (At higher values of δ, this phenomenon gets degenerate perhaps because the secular approximation starts deteriorating.) Here we have selected $\delta/\gamma\ll2\Omega_0/\gamma$ so the secular approximation must be maintained. This ringing could be attributed essentially due to the oscillatory intensity distribution pattern of the new sidebands generated along with a kind of Mollow triplet as a result of bichromatic excitation by intense fields. The new sidebands generated in the process are located at $\omega=\omega_L\pm n\delta$. We can quantify our statement by diagonalizing the Hamiltonian (2) with $\Delta=0$. The eigenvalues of this Hamiltonian are $\pm n\delta$ and the popu-

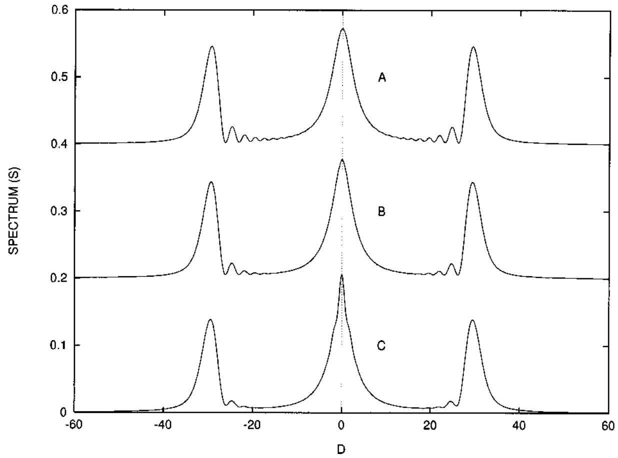

FIG. 2. Steady-state fluorescent spectrum S (in arbitrary units) of a two-level atom under bichromatic ($\delta/\gamma=0.3$) excitation in a squeezed bath with driving field in resonance with the atomic transition ($\Delta=0$). Here $2\Omega_0/\gamma=30$, $D=(\omega-\omega_a)/\gamma$, $N=1$, $|M|=\sqrt{[N(N+1)]}$, $\Phi=0$. Curve A ($S+0.4$): carrier frequency of squeezed bath is in resonance with central frequency of the driving field ($k=0$); curve B ($S+0.2$): carrier frequency of squeezed bath is centered on the first odd harmonic of δ ($k=1$); curve C (S): carrier frequency of squeezed bath is centered on the first even harmonic of δ ($k=2$).

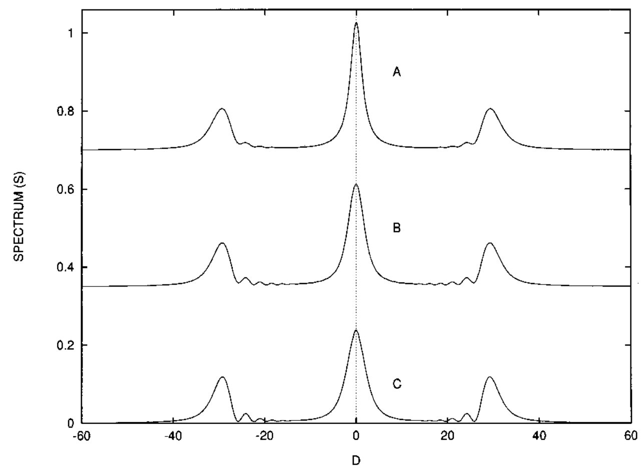

FIG. 3. The same as Fig. 2 but for $\delta/\gamma=0.35$, $\Phi=-\pi/2$. Curve A ($S+0.7$): carrier frequency of squeezed bath is in resonance with central frequency of the driving field ($k=0$); curve B ($S+0.35$): carrier frequency of squeezed bath is centered on the first odd harmonic of δ ($k=1$); curve C (S): carrier frequency of squeezed bath is centered on the first even harmonic of δ ($k=2$).

lation of the dressed states in manifold $\mathfrak{N}$ is associated with the Bessel function of argument $2\Omega_0/\delta$ consistent with Ref. [12]. Also, in the high Rabi frequency limit, higher dressed-state manifolds get populated, leading to transition to lower dressed-state manifolds and finally to the ground state [10]. The Bessel function $J_m(2\Omega_0/\delta)$ oscillates with its index m and hence the population distribution of the dressed states also oscillates, meaning for certain values of m (keeping $2\Omega_0/\delta$ unchanged), the amplitude of Bessel functions goes down to zero, resulting in zero population of that dressed state and hence that particular spectral feature is completely disappearing. So, it is the oscillatory Bessel functions giving rise to oscillating population distribution in the dressed states and that along with the proximity of the $\pm n\delta$ sidebands are responsible for the apparent oscillatory intensity pattern in the resonance fluorescence spectrum. Because of the small value of δ, the side peaks at $\omega_L\pm n\delta$ are very close to each other and thus their individual spectral envelopes of different heights are overlapping with the neighboring peaks considerably for the selected squeezed vacuum parameters and hence the overall enveloping pattern looks like a ''ringing'' with usual side peaks. This physical explanation is just a guideline toward an understanding of this phenomenon, but a more appropriate physical picture is certainly required because $\delta\ll\Omega_0,\gamma$.

In curve B of Fig. 2 we have centered the squeezed vacuum on the first odd harmonic ($k=1$) of δ. The effect of this change is visible in the sense that the ringing phenomenon starts diffusing or damping out. This effect becomes more pronounced if we center the squeezed vacuum on the first even harmonic ($k=2$, curve C of Fig. 1). Comparison of curves A, B, and C in Fig. 2 gives us the inference that the phenomenon of ringing reduces if we start centering the squeezed vacuum away from the central frequency with $\Phi=0$. Other effects of centering the squeezed vacuum on the first even harmonic are the appearance of structures around the central peak that are symmetrically located about the $D=0$ line, and the change in the width of the central peak. However, the sensitivity of the ringing phenomenon on the off centering of the squeezed vacuum critically depends on the phase Φ. We have set $\Phi=-\pi/2$ in Fig. 3 with $\delta/\gamma=0.35$ and keeping all other parameters the same as in Fig. 2. Here too like Fig. 2 we get the ringing phenomenon in the fluorescence spectrum (curve A, Fig. 3). But as we off center the squeezed vacuum frequency and move to first odd harmonic (curve B, Fig. 3) and first even harmonic (curve C, Fig. 3) of δ, to our surprise the ringing becomes more prominent. This behavior is related to the product of $J_m(x)I_n(y)e^{-y\sin(\Phi)}$ appearing in the expression of $S(D)$, Eq. (9), along with phase (Φ) at each n. Beside this the central peak starts reducing in its height and broadening as we move from $k=0$ to $k=2$. As for $\Phi=\pi/2$ the ringing features in the spectrum are reduced relative to the above cases of $\Phi=0,-\pi/2$.

In conclusion, the bichromatic excitation (δ small) along with the presence of the resonant or off-resonant squeezed vacuum field brings out noticeable changes in the resonance fluorescence spectrum of a strongly driven two-level atom. We have considered all harmonics of the spectrum in our calculations. The bichromatic field as such produces ringing in the Mollow triplet which becomes sensitive to the phase of the squeezed vacuum as well as centering the location of the carrier frequency of the squeezed vacuum with respect to the driving field frequency. Observation of the spectrum in the absence of the squeezed vacuum for a two-level atom under intense bichromatic field excitation has shown [11] that the fluorescence spectrum contains no Rabi sidebands of the Mollow triplet, instead there is a series of sidebands (comblike structure) separated by half the frequency difference between the two components of driving fields. The number of sidebands increases with increase in Rabi frequency [11,12]. What we have studied here is the resonance fluorescence in a different regime of parameter value δ/γ valid under secular approximation. The ringing we have observed has its frequency directly related to the periodic nature of $J_m(x)$ with m and it is in contrast to the previously mentioned results in the literature. Also, the ringing phenomenon is sensitive to both the centering of carrier frequency as well as the relative phase of the squeezed vacuum. With the recent availability of squeezed radiation [13] (with $N=0.5$) and analogously to the experiment reported in [11] on Ba atoms under intense bichromatic excitation in normal vacuum, the results presented here offer the possibility of observing such ringing effect in the fluorescence spectrum of two-level systems embedded in a squeezed radiation bath.

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