Generation of squeezed states of radiation at submultiple Rabi resonances

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I show that the nonlinear mixing of an intense pump and a weak probe of frequencies ω_2 and $\omega_2 - \Omega$, where Ω is the pump's generalized Rabi frequency, leads to the spontaneous generation of the radiation at the submultiple of the Rabi frequency, that is, at $\omega_2 + (\Omega/2)$. I prove that such radiation at $\omega_2 + (\Omega/2)$ has strong squeezing properties that are nearly unaffected by the quantum noise of the medium.

Considerable work has been done on the spontaneous generation[1–4] of the coherent radiation at frequencies other than the applied frequencies. Such generation of radiation is fundamentally important and is also a potential source[5] for the squeezed states of the radiation field. Much of the current activity has been devoted to the spontaneous generation of the radiation at the frequencies ω_3 and ω_4 such that $\omega_3 + \omega_4 = 2\omega_2$, where ω_2 is the pump frequency. A coherent combination of the fields at ω_3 and ω_4 exhibits squeezing properties. Boyd *et al.*[6] demonstrated a different kind of four-wave mixing. They showed that an intense pump characterized by a generalized Rabi frequency Ω $[=(\Delta^2 + 4|g_2|^2)^{1/2}$; $\Delta = \omega_0 - \omega_{2j}$; $|g_2| = |\mathbf{d} \cdot \epsilon/\hbar|]$; ω_0 is the atom's frequency] can lead to the generated waves at the Rabi sidebands $\omega_2 \pm \Omega$.

Normally four-wave mixing is described in terms of the third-order nonlinearities of the medium. However, the generation of the Rabi sidebands has to be described by a nonlinear susceptibility that depends on all orders of the intensity of the pump. Suppose that we consider the interaction of a system with an intense field of frequency ω_2 and a weak field of frequency ω_3. First-order calculations in the weak field show that the induced polarization can be written as

$$\begin{aligned} P(\mathbf{r}, t) &= \exp(-i\omega_3 t + i\mathbf{k}_3 \cdot \mathbf{r})\epsilon(\omega_3)p_a \\ &\quad + \exp[-i(2\omega_2 - \omega_3)t + i(2\mathbf{k}_2 - \mathbf{k}_3) \cdot \mathbf{r}] \\ &\quad \times \epsilon^2(\omega_2)\epsilon^*(\omega_3)p_m + \text{c.c.} \end{aligned} \tag{1}$$

Here the coefficients p_a and p_m depend on all orders of the intensity of the pump. These are functions of the various frequencies. $p_a(p_m)$ describes the linear and nonlinear absorption or gain at the frequency ω_3 (the generation of the coherent radiation at $2\omega_2 - \omega_3$). The analytical expressions for the susceptibilities p_a and p_m are well known.[6,7] Note that p_a and p_m are related to the traditional susceptibilities of nonlinear-optics literature as

$$\begin{aligned} p_a &= \chi^{(1)}(\omega_3) + \chi^{(3)}(-\omega_2, \omega_2, \omega_3)|\epsilon(\omega_2)|^2 \\ &\quad + \chi^{(5)}(-\omega_2, \omega_2, -\omega_2, \omega_2, \omega_3)|\epsilon(\omega_2)|^4 + \ldots, \end{aligned} \tag{2}$$

$$\begin{aligned} p_m &= \chi^{(3)}(\omega_2, \omega_2, -\omega_3) + \chi^{(5)}(\omega_2, \omega_2, -\omega_3, \omega_2, -\omega_2)|\epsilon(\omega_2)|^2 \\ &\quad + \chi^{(7)}(\omega_2, \omega_2, -\omega_3, \omega_2, -\omega_2, \omega_2, -\omega_2)|\epsilon(\omega_2)|^4 + \ldots . \end{aligned} \tag{3}$$

The quantum theory of the generation at $\omega_2 \pm \Omega$ can be developed along the lines of my earlier work,[4] and this theory can be used to study the strictly quantum properties of the field, such as squeezing. The quantum-mechanical fluctuations (noise) of the medium are significant at frequencies in the neighborhood of ω_2, $\omega_2 \pm \Omega$. The work of Mollow[8] supports this concept. Any noise in the medium tends to inhibit the amount of achievable squeezing.[1] Thus, to obtain significant squeezing, we have to generate radiation at frequencies at which the medium's fluctuations are insignificant. In this paper, I demonstrate how this can be achieved by using the resonance at $\omega_2 \pm (\Omega/2)$. Such a resonance is referred to as submultiple Rabi resonance.[9–12]

I start by giving a brief discussion of the submultiple Rabi resonance.[9] First, we assume that the probe field starts becoming strong. Thus we have to include terms in Eq. (1) that are of higher order in the probe field. In general the induced polarization will have the structure

$$\begin{aligned} P(r, t) = \sum_{n=-\infty}^{\pm\infty} &\exp[in(\omega_2 - \omega_3)t - i\mathbf{r} \cdot (\mathbf{k}_2 - \mathbf{k}_3)n] \\ &\times \exp(i\mathbf{k}_2 \cdot \mathbf{r} - i\omega_2 t)\Psi_n. \end{aligned} \tag{4}$$

The coefficients Ψ_n depend on all orders of the intensities of both pump and probe fields. Thus Ψ_n's are expected to have the form

$$\Psi_n = \sum_{p,q} \Psi_n{}^{(p,q)}\epsilon^p(\omega_2)\epsilon^q(\omega_3). \tag{5}$$

The usual four-wave mixing will be described by $\Psi_{-1}{}^{(2,1)}$. To deal with the pump-induced saturation effects we rewrite Eq. (5) as

$$\Psi_n = \sum \phi_n{}^{(q)}\epsilon^q(\omega_3). \tag{6}$$

The coherent generation of the waves at $\omega_2 \pm \Omega$ can be described by $\Phi_{-1}{}^1$.

Interesting new effects arise when we examine the response functions $\phi_n{}^{(q)}$ for $q > 1$. Consider, for example, the

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generation of the coherent radiation at $3\omega_2 - 2\omega_3$. To lowest order in both the fields this generation is described by $\Psi^{(3,2)}$ [which is equivalent to the traditional fifth-order nonlinearity $\chi^{(5)}(\omega_2, \omega_2, \omega_2, -\omega_3, -\omega_3)$]. For a saturating pump, the generation of radiation at $3\omega_2 - 2\omega_3$ can be studied in terms of the function $\Phi_{-2}^{(2)}$. In what follows I treat only the coherently generated radiation from a two-level system interacting with resonant pump and probe fields. The nonlinear mixing of arbitrarily high order was investigated earlier.[9] The generated signal exhibits resonances at

$$\omega_3 - \omega_2 = 0, \pm\Delta, \pm\Delta/2, \Delta = \omega_0 - \omega_2. \tag{7}$$

The saturation of the pump changes resonance positions to

$$\omega_3 - \omega_2 = 0, \pm(\Delta^2 + 4|g|^2)^{1/2}, \pm 1/2(\Delta^2 + 4|g|^2)^{1/2}. \tag{8}$$

Thus the coherently generated signal at $3\omega_2 - 2\omega_3$ also exhibits a resonance at a frequency that is displaced from the central frequency by an amount equal to one half of the generalized Rabi frequency. The resonances at $\omega_3 = \omega_2 \pm (\Delta^2 + 4|g|^2)^{1/2}$ are central to the present work. The explicit form of the nonlinear response follows from optical Bloch equations for a two-level system driven by two fields of frequencies ω_2 and ω_3. The result to all orders in ϵ_2 but to second order in ϵ_3 is

$$\begin{aligned}\phi_{-2}^{(2)}\epsilon_3^{\,2} = \Phi \equiv \frac{4ig_2^{\,3}g_3^{\,2}}{T_2}\left[1 + \frac{4g_2^{\,2}T_1T_2}{(1+\Delta^2T_2^{\,2})}\right]^{-1} \\ \times (1 + i\Delta T_2)^{-1}[2 + 3i(\omega_3 - \omega_2)T_2] \\ \times [2 + i(\omega_3 - \omega_2)T_2]P^{-1}[i(\omega_3 - \omega_2)] \\ \times P^{-1}[2i(\omega_3 - \omega_2)].\end{aligned} \tag{9}$$

Here $2g_3(2g_2)$ is the Rabi frequency associated with the probe field of frequency $\omega_3(\omega_2)$, T_1 and T_2 are the usual longitudinal and transverse relaxation times, respectively, and $P(z)$ is the polynomial

$$P(z) = \left(z + \frac{1}{T_1}\right)\left[\left(z + \frac{1}{T_2}\right)^2 + \Delta^2\right] + 4g_2^{\,2}\left(z + \frac{1}{T_2}\right). \tag{10}$$

Such a polynomial is familiar[13] from optical Bloch equations. The zeros of $P(z)$ yield the resonances at the central frequency ω_2 and at Rabi frequencies $\omega_2 \pm \Omega$. Thus the usual sideband generation is determined by $P(z)$. The subharmonic or submultiple Rabi resonance arises from the roots of $P(2z)$. The nonlinear response Φ can be expressed as a Taylor series in terms of the traditional susceptibilities

$$\begin{aligned}\Phi = \chi^{(5)}(\omega_2, \omega_2, \omega_2, -\omega_3, -\omega_3)\epsilon^3(\omega_2)\epsilon^{*2}(\omega_3) \\ + \chi^{(7)}(\omega_2, \omega_2, \omega_2, +\omega_2, -\omega_2, -\omega_3, -\omega_3)\epsilon^3(\omega_2)|\epsilon(\omega_2)|^2 \\ \times \epsilon^{*2}(\omega_3) + \ldots.\end{aligned} \tag{11}$$

Thus the nonlinear response Φ is in a sense the renormalized form of $\chi^{(5)}$. Figure 1 shows the behavior of the function $|\Phi|$ as a function of the parameter $(\omega_3 - \omega_2)T_2$ for an intense pump and for several values of the detuning of the pump. The resonance marked II (I) is the submultiple Rabi resonance (analog of the usual Rabi resonance). Figure 1 shows that the submultiple resonance is much more pronounced than the usual Rabi resonance.

We next examine the quantum properties of the radiation generated in the vicinity of the submultiple Rabi resonance. In particular we look for the phase-noise (squeezing) characteristics of the generated field. We thus consider an effective quantum-mechanical Hamiltonian that will describe the generation of the fields at a subharmonic of the Rabi frequency. Note that the induced polarization leads to coherent radiation of frequency $3\omega_2 - 2\omega_3$ in the direction $3\mathbf{k}_2 - 2\mathbf{k}_3$. Hence the following effective Hamiltonian describes the interaction between the fields at the frequencies ω_2, ω_3 and $3\omega_2 - 2\omega_3$:

$$H_{\text{eff}} = \Phi_{-2}^{\,2}\epsilon^{*2}(\omega_3)[-\mathbf{d}\cdot\epsilon^*(3\omega_2 - 2\omega_3)] + \text{c.c.}, \tag{12}$$

where $\mathbf{d}$ is the dipole matrix element. If we chose $\omega_3 \cong \omega_2 + (\Omega/2)$, then $3\omega_2 - 2\omega_3 \approx \omega_2 - \Omega$. Thus if the system is driven by a strong pump of frequency ω_2 and a weak field at $\omega_2 - \Omega$, then the spontaneous generation of the field of frequency ω, in the region of the subharmonic of the Rabi frequency $\omega_2 + (\Omega/2)$, can take place. If we treat the generated field at $\omega_2 + (\Omega/2)$ quantum mechanically and the remaining fields semiclassically, then the effective Hamiltonian [Eq. (12)] reduces to

$$\begin{aligned}H_{\text{eff}} = \beta a^{+2} + \text{h.c.}, \\ \epsilon\left(\omega_2 + \frac{\Omega}{2}\right) = i\left[\frac{2\pi\hbar\left(\omega_2 + \frac{\Omega}{2}\right)}{V}\right]^{1/2} a,\end{aligned} \tag{13}$$

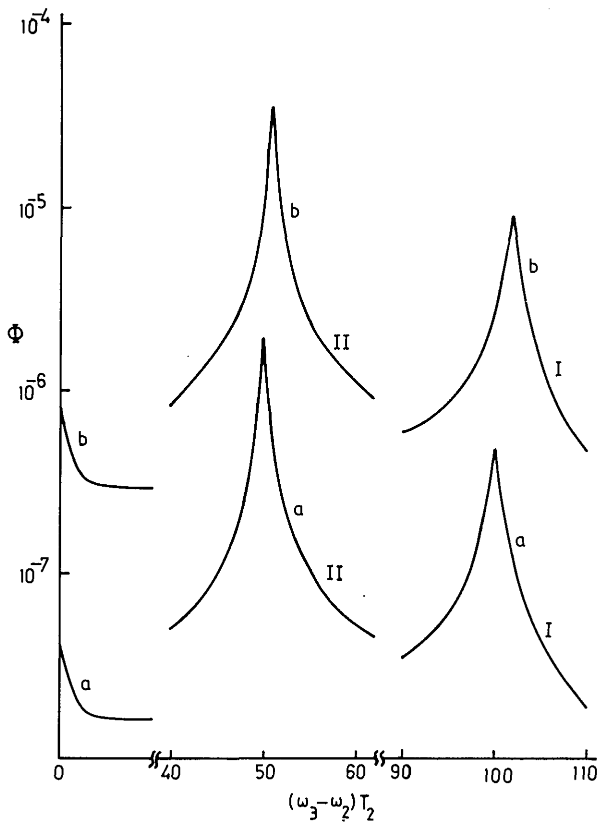

Fig. 1. The nonlinear mixing coefficient $\Phi \propto |\phi_{-2}^{(2)}|$ of the frequencies ω_2 and ω_3, to produce coherent radiation at $3\omega_2 - 2\omega_3$, as a function of $\omega_3 - \omega_2$ for an intense pump with Rabi frequency 100. Curves a and b are for a pump atom-detuning parameter equal to zero and 20, respectively. All parameters are in units of $1/T_2$, which has been chosen as $50/T_1$.

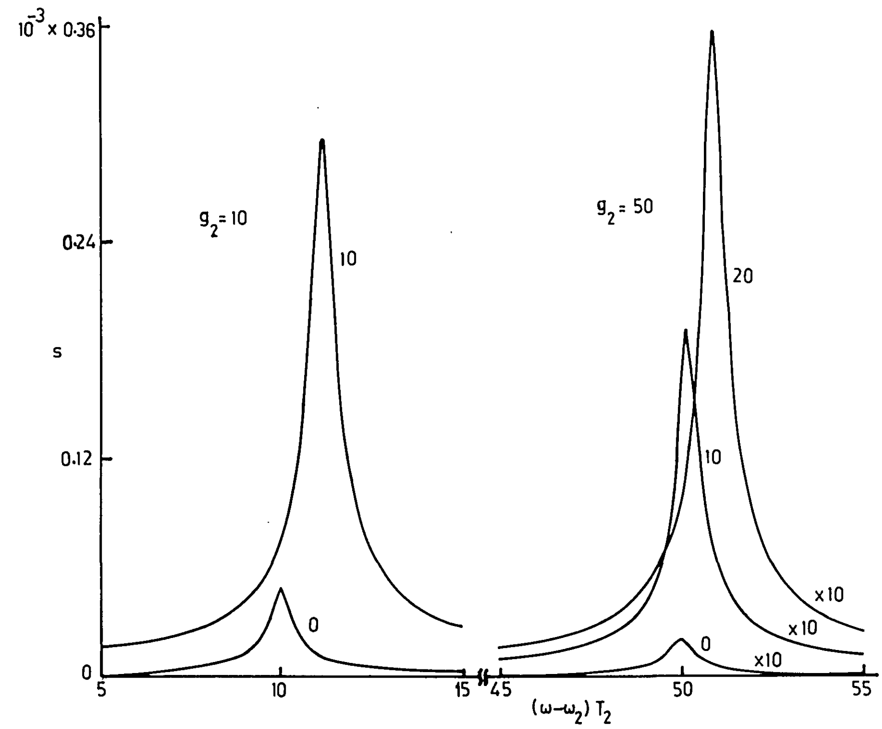

Fig. 2. A plot of the parameter s, which determines the amount of squeezing of the radiation generated in the region of submultiple Rabi resonance for two values of the pump's Rabi frequency. The curves are labeled by the values of the detuning and the Rabi frequency.

where

$$\beta = i\left[\frac{2\pi\hbar\left(\omega_2 + \dfrac{\Omega}{2}\right)}{V}\right]^{1/2}[\mathbf{d}\cdot\epsilon^*(\omega_2 - \Omega)]. \tag{14}$$

The Hamiltonian in Eq. (13) has the standard form of a two-photon Hamiltonian[14] whose exact solution[15,16] leads to the following results for the mean number of photons and the phase fluctuations:

$$\langle a^+(t)a(t)\rangle = \sinh^2(2|\beta|t), \tag{15}$$

$$\pm 1/4\langle[a(t)e^{i\theta} \pm a^+(t)e^{-i\theta}]^2\rangle = (1/4)e^{\pm 4|\beta|t}, \quad -i\beta = |\beta|e^{-2i\theta}. \tag{16}$$

We assume that the field $\epsilon[\omega_2 + (\Omega/2)]$ at time $t = 0$ is in the vacuum state. Equation (13) is also similar to the usual Hamiltonian for the downconversion[16] process, which was recently used[17] to get considerable squeezing. The nonlinearity for the present problem is given by Eq. (14), which can be enhanced by using the resonant character of Φ. Thus the enhancement of the nonlinearity could be considerable because of the resonance coming from $P^{-1}[2i(\omega_3 - \omega_2)]$.

The squeezing parameter $4|\beta|t$ can be written in the form $4|\beta|t = (2\alpha Ls)$ multiplied by the Rabi frequency of the field at $\omega - \Omega$, where

$$\begin{aligned} s = {} & \frac{4i}{T_2}{g_2}^3\left[1 + \frac{4{g_2}^2T_1T_2}{(1 + \Delta^2{T_2}^2)}\right]^{-1} \\ & \times P^{-1}[i(\omega_3 - \omega_2)]P^{-1}[2i(\omega_3 - \omega_2)] \\ & \times \{2 + 3iT_2(\omega_3 - \omega_2)\}\{2 + iT_2(\omega_3 - \omega_2)\} \\ & \times \{1 + i\Delta T_2\}^{-1}. \end{aligned} \tag{17}$$

The behavior of s near the submultiple Rabi resonance is shown in Fig. 2. Substantial values of the squeezing parameter can be achieved by choosing αL, detuning, ω_3, etc. The effect can be further enhanced by putting the resonant system into a cavity. The amount of achievable squeezing depends on the spontaneous noise of the medium. We thus examine the question of whether the quantum fluctuations due to spontaneous emission from atoms are significant in the interesting region ($\approx\omega_2 + \Omega/2$).

Mollow[8] examined the spectrum of the spontaneous noise produced by an atom driven by an external field of frequency ω_2. He found that the quantum fluctuations of the medium are important at frequencies ω_2 and $\omega_2 \pm \Omega$. Because we have considered the generation of photons at $\omega_2 + (\Omega/2)$, the squeezing in the field generated at $\omega_2 + (\Omega/2)$ is not much affected by the quantum noise of the medium. This characteristic will be true if the probe field at $\omega_2 - \Omega$ does not lead to noise terms at $\omega_2 - (\Omega/2)$. I will now show that the simultaneous action of a probe field of frequency ν and the pump leads to noise predominantly at frequencies $\omega - \omega_2 = \pm\Omega, \pm 2\Omega, 0$. To see this, we examine the structure of the dipole–dipole correlation function $\Gamma(\tau, t) \equiv \langle s^+(t + \tau)s^-(t)\rangle$ for a two-level atom interacting with two fields of frequencies ω_2 and ν. The Fourier transform of $\lim_{t\to\infty}\Gamma(\tau, t)$ will yield the spectrum of noise. The correlation function Γ can be obtained from the solution of optical Bloch equations and the regression theorem. In fact we can derive equations for the correlation matrix C with elements $\Gamma(\tau, t)$, $\langle s^-(t + \tau)s^-(t)\rangle$, and $\langle s^z(t + \tau)s^-(t)\rangle$, and these can be written in the form

$$\begin{aligned} \frac{\partial C}{\partial \tau} = {} & AC + I\,\langle s^-(t)\rangle + B_+e[-ix(t + \tau)]C \\ & + B_-\exp[+ix(t + \tau)]C, \qquad x = \nu - \omega_2. \end{aligned} \tag{18}$$

The matrices $B_{\pm}$ depend on the probe field in the vicinity of $\omega_2 - \Omega$. The matrix A depends on the strong pump. The explicit form of these matrices is not needed.

If we introduce the Laplace transform $\hat{C}(z, t)$ of $C(\tau, t)$, then in the long-time limit C is expected to have the structure

$$\hat{C}(z, t) = \sum_{-\infty}^{+\infty} \exp(-inxt)\hat{C}^{(n)}(z),$$

$$\langle s^{-}(t)\rangle = \sum_{-\infty}^{+\infty} \exp(-inxt)s^{(n)}. \quad (19)$$

From Eqs. (18) and (19), we obtain

$$z\hat{C}^{(n)} - C^{(n)}(0, t) = A\hat{C}^{(n)} + \frac{I}{z}s^{(n)} + B_{-}\hat{C}^{(n+1)}(z - ix) + B_{+}\hat{C}^{(n-1)}(z + ix). \quad (20)$$

We need to compute $C^{(0)}$ to second order in $B_{\pm}$. The inversion of the matrix $(z - A)$ leads to the polynomial $P(z)$. Using Eq. (20), we can show that $\hat{C}^{(0)}$ to second order in $B_{\pm}$ involves the denominators $P(z)$ and $P(z \pm ix)$. Thus the spectrum of noise will peak at the frequencies

$$\omega - \omega_2 = 0, \pm\Omega, \qquad \omega - \omega_2 \pm x = 0, \pm\Omega. \quad (21)$$

For the present problem the probe field is in the vicinity of the Rabi sideband $\omega_2 - \Omega$. Equations (21) then show that the spectrum of spontaneous noise will consist of the spectral peaks at

$$\omega - \omega_2 = 0, \pm\Omega, \pm2\Omega. \quad (22)$$

Therefore the spontaneous noise appears predominantly at the Rabi sidebands and at the harmonics of the Rabi sidebands. Thus the subharmonic region corresponding to $\omega = \omega_2 + (\Omega/2)$ is relatively noise-free.

In conclusion, I have shown how the subharmonic Rabi resonances can be used to generate radiation with strong squeezing properties. This region of the submultiple Rabi resonances is also relatively free from the spontaneous noise of the resonant medium.

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