Violation of Cauchy-Schwarz and Bell's inequalities in four-wave mixing

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Certain nonclassical effects in four-wave mixing are studied within the framework of a quantum theory of multiwave mixing. It is shown that the Cauchy-Schwarz and Bell's inequalities are violated for large detuning of the pump frequency from the side-mode frequencies.

I. INTRODUCTION

In quantum optics, nonclassical effects (where P representation is not positive definite) exhibit some exciting phenomena including squeezing, photon antibunching, and violation of the Cauchy-Schwarz and Bell's inequalities. In recent years, squeezing has been the subject of great attention because of its potential applications in weak-signal detection and optical communication systems, and a lot of interesting work has been done in this regard. Yuen and Shapiro[1] first proposed squeezing in degenerate four-wave mixing. Reid and Walls[2] also gave a detailed microscopic model for degenerate four-wave mixing and predicted squeezing. Recently, Holm, Sargent, and Capron[3] predicted squeezing in nondegenerate four-wave mixing and marked the regions of strong squeezing. Kilin[4] predicted squeezing in forward degenerate four-wave mixing. Experimentally squeezed states have been observed in four-wave mixing by Slusher *et al.*[5] and Levenson *et al.*[6] The second-order coherence function provides information to test photon antibunching and violation of the Cauchy-Schwarz inequality.[7] Kimble *et al.*[8] and Cresser *et al.*[9] observed photon antibunching in resonance fluorescence from atoms in an atomic beam. Walther and Diedrich[10] recently observed this effect in resonance fluorescence from a single ion in an ion trap. Violation of the Cauchy-Schwarz inequality has also been studied in many systems including the two-photon laser,[11] the parametric amplifier,[12] Jaynes-Cumming–type model systems,[13] resonance fluorescence,[14] and the three-photon hyper-Raman process.[15] Clauser, and more recently, Grangier *et al.*, observed the violation of the Cauchy-Schwarz inequality in an atomic two-photon cascade system.[16] Bell's inequality[17,18] provides a test of quantum mechanics. Aspect and coworkers[19–21] performed experiments to test Bell's theorems and found good agreement with quantum-mechanical predictions. Reid and Walls[22,23] and Drummond[24] showed that the states of radiation fields which give violation of Bell's inequalities cannot be represented in terms of singular positive P representation. In a recent paper Reid and Walls proposed[22] an experiment for a two-mode source to test the violation of Bell's inequality. They also predicted the limits for the violation of Bell's inequality in such systems.

In this paper we discuss the violation of the Cauchy-Schwarz and Bell's inequalities in a nondegenerate four-wave mixing process within the framework of the theory developed by Sargent, Holm, and Zubairy.[25] This theory describes a number of interesting topics in quantum optics, including resonance fluorescence, saturation spectroscopy, laser, and optical-bistability instabilities, and three- and four-wave mixing. The theory treats a classical plane running wave of intense frequency and one or two weak quantized plane running waves. In Sec. II, we derive a Fokker-Planck equation for the Q representation from the reduced density-matrix equation for the fields. We then present an exact steady-state expression for the Q representation for the field modes. Various correlation functions needed for the study of nonclassical effects can be obtained from this expression in a straightforward manner. In Sec. III we calculate appropriate fourth-order correlation functions and discuss the violation of the Cauchy-Schwarz and Bell's inequalities in four-wave mixing. We plot these inequalities against detunings of the pump frequency to the side-mode frequencies and mark the region where these inequalities are violated.

II. EQUATION OF MOTION FOR THE FIELD DENSITY MATRIX

In four-wave mixing the pump frequency is servo-locked at frequency ν_2 and the side-mode frequencies are locked at frequencies ν_1 and ν_3, respectively, so that the mode-locking condition $\nu_2-\nu_1=\nu_3-\nu_2$ is satisfied. We take the pump field to be arbitrarily intense, and treat it classically up to all orders. Side modes of frequencies ν_1,ν_3 are considered weak and are treated quantum mechanically up to the second order in the coupling constant. The atomic system consists of two-level atoms where transition takes place between the ground state and some excited states.

The slowly varying field density operator equation of motion for the system is[25]

$$\begin{aligned}\rho=&-A_1(\rho a_1 a_1^\dagger - a_1^\dagger \rho a_1)\\&-(B_1+\nu/2Q_1)(a_1^\dagger a_1\rho - a_1\rho a_1^\dagger)\\&+C_1(a_1^\dagger a_3^\dagger\rho - a_3^\dagger\rho a_1^\dagger)+D_1(\rho a_3^\dagger a_1^\dagger - a_1^\dagger\rho a_3^\dagger)\\&+[1\leftrightarrow 3]+\text{H.c.}\ ,\end{aligned}\tag{1}$$

where $1\leftrightarrow 3$ represents the same terms with subscripts 1 and 3 interchanged, ν/Q is the cavity loss rate, and a_i

and $a_i^\dagger$ are the annihilation and creation operators for the ith mode. The expressions of the coefficients A_1, B_1, C_1, D_1 appearing in Eq. (1) are given in Ref. 25. We interpret $A_1 + A_1^*$ as the spectrum of resonance fluorescence, while $A_1 - B_1$ is the semiclassical complex gain and/or absorption coefficient and $C_1 - D_1$ is the semiclassical complex-coupling coefficient.

In four-wave mixing the strong pump field consists of two oppositely directed running waves, forming a standing wave. Due to spatial hole burning (SHB) of the upper and lower population difference, the atoms in the different locations receive different amount of saturation. So the coefficients used in Eq. (1) should be averaged over the spatial hole burning for one wavelength. The resulting expressions for the averaged coefficients A_1, B_1, C_1, D_1, are[26]

$$\langle A_1 \rangle_{\rm SHB} = \frac{g^2 D_1}{\mathcal{L}_2 - d}\left[\frac{-b_1 + c/\mathcal{L}_2}{(1+4I_2\mathcal{L}_2)^{1/2}} - \frac{-b_1 + c/d}{(1+4I_2 d)^{1/2}} + c\left[\frac{1}{d} - \frac{1}{\mathcal{L}_2}\right]\right], \tag{2a}$$

$$\langle B_1 \rangle_{\rm SHB} = \frac{g^2 D_1}{\mathcal{L}_2 - d}\left[\frac{-b_2 + c/\mathcal{L}_2}{(1+4I_2\mathcal{L}_2)^{1/2}} - \frac{d - b_2 + c/d}{(1+4I_2 d)^{1/2}} + c\left[\frac{1}{d} - \frac{1}{\mathcal{L}_2}\right]\right], \tag{2b}$$

$$\langle C_1 \rangle_{\rm SHB} = -\frac{g^2 U_1^* U_3^* D_1}{\mathcal{L}_2 - d}\left[\frac{-b_3 + c/\mathcal{L}_2}{(1+4I_2\mathcal{L}_2)^{1/2}} - \frac{-b_3 + c/d}{(1+4I_2 d)^{1/2}} + c\left[\frac{1}{d} - \frac{1}{\mathcal{L}_2}\right]\right], \tag{2c}$$

$$\langle D_1 \rangle_{\rm SHB} = \frac{-g^2 U_1^* U_3^* D_1}{\mathcal{L}_2 - d}\left[\frac{-b_4 + c/\mathcal{L}_2}{(1+4I_2\mathcal{L}_2)^{1/2}} - \frac{-b_4 + c/d}{(1+4I_2 d)^{1/2}} + c\left[\frac{1}{d} - \frac{1}{\mathcal{L}_2}\right]\right], \tag{2d}$$

where

$$b_1 = \frac{\mathcal{L}_2}{2} + \frac{\gamma D_2^* F}{4}\left[1 + \frac{\Gamma}{i\Delta}\right], \tag{3a}$$

$$b_2 = \frac{\mathcal{L}_2}{2} + \frac{\gamma F}{2}\left[D_3^* - \frac{D_2^*}{4}\left[1 - \frac{\Gamma}{i\Delta}\right]\right], \tag{3b}$$

$$b_3 = \frac{-\gamma D_2 F}{4}\left[1 + \frac{\Gamma}{i\Delta}\right], \tag{3c}$$

$$b_4 = \frac{\gamma}{2} F\left[D_3^* + \frac{D_2}{4}\left[1 - \frac{\Gamma}{i\Delta}\right]\right], \tag{3d}$$

$$c = \frac{\gamma \mathcal{L}_2 F D_3^*}{4}, \tag{3e}$$

$$d = \frac{\gamma}{2} F (D_1 + D_3^*) . \tag{3f}$$

The complex Lorentzian denominator D_n is

$$D_n = \frac{1}{\gamma + i(\omega - \nu_n)}, \tag{3g}$$

and the dimensionless quantities such as the Lorentzian $\mathcal{L}_2$ intensity I_2, and the population pulsation term F are defined by the following equations:

$$\mathcal{L}_2 = \gamma^2 / [\gamma^2 + (\omega - \nu_2)^2] , \tag{3h}$$

$$I_2 = \mu_0^2 |A_2|^2 T_1 T_2 / \hbar^2 , \tag{3i}$$

$$F = \frac{\Gamma}{\Gamma + i\Delta} . \tag{3j}$$

The constants $\Gamma (=1/T_1)$ and $\gamma (=1/T_2)$ are the upper-to-lower-level decay constant and dipole decay constant, respectively. Also, g is the atom-field coupling constant, ω is the atomic transition frequency, and $\Delta = \nu_2 - \nu_1$ is the beat frequency between the models 1 and 2. The coefficients A_3, B_3, C_3, D_3 are obtained by interchanging ν_1 and ν_3 in the expressions of A_1, B_1, C_1, D_1. The system we consider here involves atomic transitions that take place between ground level and some excited state.

III. FOKKER-PLANCK EQUATION OF MOTION FOR THE FIELD

In this section we derive a Fokker-Planck equation for the Q representation for the field modes from the equation of motion for the reduced field density matrix, and then solve it exactly in steady state. The disadvantage of using the more commonly used P representation in the present problem is that it is non-positive-definite for the nonclassical field. The Q representation, however, is positive definite for such fields.

The Q representation for the field of modes 1 and 3 is defined as

$$Q(\alpha_1, \alpha_3) = \frac{1}{\pi} \langle \alpha_1, \alpha_3 | \hat{\rho} | \alpha_1, \alpha_3 \rangle , \tag{4}$$

where $|\alpha_1, \alpha_3\rangle$ is the eigenstate of a_i, with eigenvalues α_i, i.e.,

$$a_i | \alpha_1, \alpha_3 \rangle = \alpha_i | \alpha_i, \alpha_3 \rangle \quad (i = 1, 3) . \tag{5}$$

The expectation value of any antinormally ordered function $T(a_1, a_1^\dagger, a_3, a_3^\dagger)$ can be evaluated from $Q(\alpha_1, \alpha_3)$ using

$$\langle T(a_1, a_1^\dagger, a_3, a_3^\dagger) \rangle = \int T(\alpha_1, \alpha_1^*, \alpha_3, \alpha_3^*) Q(\alpha_1, \alpha_3) d^2\alpha_1 d^2\alpha_3 . \tag{6}$$

It follows, on taking the expectation values of various terms appearing in Eq. (1) with respect to the coherent state $|\alpha_1, \alpha_3\rangle$, that

$$\dot{Q}=-\left[A_1\frac{\partial}{\partial\alpha_1}\alpha_1+A_3\frac{\partial}{\partial\alpha_3}\alpha_3\right]Q+\left[B_1\left[\frac{\partial}{\partial\alpha_1}\alpha_1+\frac{\partial^2}{\partial\alpha_1\partial\alpha_1^*}\right]+B_3\left[\frac{\partial}{\partial\alpha_3}\alpha_3+\frac{\partial^2}{\partial\alpha_3\partial\alpha_3^*}\right]\right]Q$$

$$-\left[C_1\frac{\partial}{\partial\alpha_1}\alpha_3^*+C_3\frac{\partial}{\partial\alpha_3}\alpha_1^*\right]Q+\left[D_1\left[\frac{\partial}{\partial\alpha_1}\alpha_3^*+\frac{\partial^2}{\partial\alpha_1\partial\alpha_3}\right]+D_3\left[\frac{\partial}{\partial\alpha_3}\alpha_1^*+\frac{\partial^2}{\partial\alpha_3\partial\alpha_1}\right]\right]Q+\text{c.c.} \qquad (7)$$

After performing rather lengthy calculations we get the exact steady-state solution of the Fokker-Planck equation, which satisfies the above equation. The solution is of the form

$$Q(\alpha_1,\alpha_3)=\frac{1}{N}\exp[-(L_{11}|\alpha_1|^2+L_{33}|\alpha_3|^2+L_{13}\alpha_1\alpha_3+L_{31}\alpha_1^*\alpha_3^*)] , \qquad (8)$$

where the normalization constant

$$N=\int\exp[-(L_{11}|\alpha_1|^2+L_{33}|\alpha_3|^2+L_{13}\alpha_1\alpha_3+L_{31}\alpha_1^*\alpha_3^*)]d^2\alpha_1 d^2\alpha_3 \qquad (9)$$

and

$$L_{11}=\langle a_3a_3^\dagger\rangle/(\langle a_1a_1^\dagger\rangle\langle a_3a_3^\dagger\rangle-|\langle a_1a_3\rangle|^2) , \qquad (10a)$$

$$L_{33}=\langle a_1a_1^\dagger\rangle/(\langle a_1a_1^\dagger\rangle\langle a_3a_3^\dagger\rangle-|\langle a_1a_3\rangle|^2) , \qquad (10b)$$

$$L_{13}=-\langle a_1^\dagger a_3^\dagger\rangle/(\langle a_1a_1^\dagger\rangle\langle a_3a_3^\dagger\rangle-|\langle a_1a_3\rangle|^2) , \qquad (10c)$$

$$L_{31}=-\langle a_1a_3\rangle/(\langle a_1a_1^\dagger\rangle\langle a_3a_3^\dagger\rangle-|\langle a_1a_3\rangle|^2) . \qquad (10d)$$

The expectation values of normally ordered correlation functions are obtained from antinormally ordered correlation functions using the commutation relation $[a_i,a_i^\dagger]=1$. The expectation values $\langle a_1^\dagger a_1\rangle$, $\langle a_1a_3\rangle$ appearing in Eqs. (10a)–(10d) are related to various coefficients by the following expressions:

$$\langle a_1^\dagger a_1\rangle=\frac{1}{D}\{A_1[\beta_3(|\beta_1+\beta_3|^2)-(\beta_1+\beta_3)\kappa_1^*\kappa_3+\text{c.c.}]$$
$$+A_3(\beta_1+\beta_3+\text{c.c.})|\kappa_1|^2+\kappa_1^*(C_1+C_3)[(\beta_3+\beta_3^*)(\beta_1^*+\beta_3^*)-\kappa_1^*\kappa_3+\kappa_1\kappa_3^*]+\text{c.c.}\} , \qquad (11)$$

$$\langle a_1a_3\rangle=\frac{1}{D}\{-\kappa_3[\kappa_1^*\kappa_3-\kappa_1\kappa_3^*-(\beta_3+\beta_3^*)(\beta_1^*+\beta_3^*)](A_1+A_1^*)$$
$$+(\beta_1+\beta_1^*)[\beta_1^*(\beta_3+\beta_3^*)(C_1+C_3)-\kappa_3(\kappa_1^*C_1+\kappa_1^*C_3-\text{c.c.})]+[1\leftrightarrow 3]\} . \qquad (12)$$

The denominator D in Eqs. (11) and (12) is given by

$$D=(\beta_1+\beta_1^*)(\beta_3+\beta_3^*)|\beta_1+\beta_3|^2+(\kappa_1^*\kappa_3-\kappa_1\kappa_3^*)^2-\{(\beta_1+\beta_1^*)\kappa_1^*\kappa_3(\beta_1+\beta_3)+[1\leftrightarrow 3]+\text{c.c.}\} , \qquad (13)$$

with $\beta_1=(B_1+\nu/2Q-A_1)$, $\kappa_1=(C_1-D_1)$. The quantity $\langle a_3^\dagger a_3\rangle$ is given by Eq. (11) with $1\leftrightarrow 3$ interchanged. The above expectation values are evaluated from the following equations of motion:

$$\frac{d}{dt}\langle a_1^\dagger a_1\rangle=(A_1-B_1-\nu/2Q_1)\langle a_1^\dagger a_1\rangle+(C_1-D_1)\langle a_1^\dagger a_3^\dagger\rangle+A_1+\text{c.c.} , \qquad (14)$$

$$\frac{d}{dt}\langle a_1a_3\rangle=(A_1-B_1-\nu/2Q_1)\langle a_1a_3\rangle+(C_1-D_1)\langle a_3^\dagger a_3\rangle+C_1+[1\leftrightarrow 3] . \qquad (15)$$

The equation of motion for the number operator $\langle a_3^\dagger a_3\rangle$ can be obtained by interchanging $1\leftrightarrow 3$ in Eq. (14). The quantity $\langle a_1^\dagger a_3^\dagger\rangle$ is determined by taking the complex conjugate of Eq. (15). In steady state the time dependence of these equations vanishes and we get four simultaneous equations. After solving these equations algebraically we obtain the expectation values given by Eqs. (11) and (12).

IV. NONCLASSICAL EFFECTS

In this section we evaluate the expectation values of fourth-order correlation functions for the field. We predict that the Cauchy-Schwarz inequality and Bell's inequality are violated under certain conditions. We also plot the appropriate correlation functions and identify the regions where these inequalities are violated.

A. Squeezing

Recently, Holm, Sargent, and Capron[3] predicted squeezing in four-wave mixing using the present approach. We briefly review some of the calculations for the sake of completeness. We define the linear superposition of the coupled-mode annihilation operator by

$$d=\frac{1}{\sqrt{2}}(a_1+a_3)e^{i\theta} . \qquad (16)$$

Then canonically conjugate Hermitian amplitude operators are given by

$$d_1=\tfrac{1}{2}(d+d^{\dagger}) , \tag{17a}$$

$$d_2=\frac{1}{2i}(d-d^{\dagger}) . \tag{17b}$$

The squared variances are given by

$$\Delta d_i^2=\tfrac{1}{4}+\tfrac{1}{4}(\langle a_1^{\dagger}a_1\rangle+\langle a_3^{\dagger}a_3\rangle\pm\langle a_1a_3e^{2i\theta}+\text{c.c.}\rangle) , \tag{18a}$$

where $i=1,2$ for $-,+$, respectively.

The minimum variance is obtained for Δd_1^2 with phase angle choice,

$$\langle a_1a_3\rangle e^{2i\theta}=|\langle a_1a_3\rangle| , \tag{18b}$$

hence

$$\Delta d_1^2=\tfrac{1}{4}+\tfrac{1}{4}(\langle a_1^{\dagger}a_1\rangle+\langle a_3^{\dagger}a_3\rangle-2|\langle a_1a_3\rangle|) . \tag{19}$$

In Fig. 1 we plot this variance versus detuning $(\nu_1-\nu_1)T_2$ for pump intensity $I_2=50$, with detuning of the pump frequency from the atomic transition frequency $(\omega-\nu_2)T_2=8$ and $C=5$, where $C=g^2/\gamma(\nu/Q)$. This figure shows the regions of strong squeezing.

B. Violation of the Cauchy-Schwarz inequality

The Cauchy-Schwarz inequality is violated in those systems where the correlation between the photons of different modes is larger than the correlation of the photon of the same mode. As mentioned before, the violation of the Cauchy-Schwarz inequality represents a nonclassical effect in the sense that the P representation of the field is not positive definite. This can be seen by the following argument.

In the case of two modes for a non-negative two-mode P representation $P(\xi_1,\xi_2)$, we get

$$\int\int\int\int(|\xi_1|^2|\eta_2|^2-|\xi_2|^2|\eta_1|^2)^2P(\xi_1,\xi_2)\times P(\eta_1,\eta_2)d^2\xi_1d^2\xi_2d^2\eta_1d^2\eta_2 , \tag{20}$$

This gives the following quantum analogue of the Cauchy-Schwarz inequality:

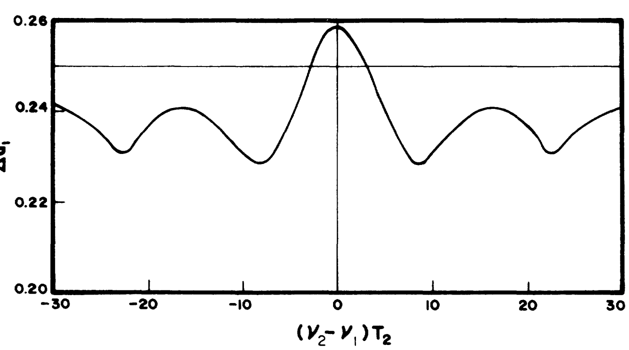

FIG. 1. Minimum squared variance Δd_1^2 vs detuning $(\nu_2-\nu_1)T_2$ for $I_2=50$, $(\omega-\nu_2)T_2=8$, and $C=10$.

$$G_{ij}\le[G_{ii}G_{jj}]^{1/2} , \tag{21}$$

where the second-order coherence function of light is defined as

$$G_{ij}=\frac{\langle a_i^{\dagger}a_j^{\dagger}a_ia_j\rangle}{\langle a_i^{\dagger}a_i\rangle\langle a_j^{\dagger}a_j\rangle} , \tag{22}$$

where a_i and $a_i^{\dagger}$ are the destruction and creation operators for the ith mode.

Hence, the Cauchy-Schwarz inequality is violated when

$$G_{ij}>[G_{ii}G_{jj}]^{1/2} . \tag{23}$$

It is evident that this happens when $P(\xi_1,\xi_2)$ is not positive definite. Hence to investigate the violation of the Cauchy-Schwarz inequality in two modes whose destruction operators are a_1 and a_3, we need to evaluate the following quantity:

$$\lambda=\frac{\langle a_1^{\dagger}a_3^{\dagger}a_1a_3\rangle}{\langle a_1^{\dagger}a_1\rangle\langle a_3^{\dagger}a_3\rangle}-\frac{(\langle a_1^{\dagger}a_1^{\dagger}a_1a_1\rangle\langle a_3^{\dagger}a_3^{\dagger}a_3a_3\rangle)^{1/2}}{\langle a_1^{\dagger}a_1\rangle\langle a_3^{\dagger}a_3\rangle} . \tag{24}$$

The Cauchy-Schwarz inequality is violated if $\lambda>0$. It follows, on substituting for $Q(\alpha_1,\alpha_3)$ from Eq. (8), that

$$\begin{aligned}\langle a_1a_1a_1^{\dagger}a_1^{\dagger}\rangle&=\int|\alpha_1|^4Q(\alpha_1,\alpha_3)d^2\alpha_1d^2\alpha_3 ,\\&=\frac{1}{N}\frac{\partial^2N}{\partial L_{11}^2} ,\\&=2\langle a_1a_1^{\dagger}\rangle^2 .\end{aligned} \tag{25}$$

By using the commutation relation $[a_i,a_i^{\dagger}]=1$, we convert this correlation function into a normally ordered correlation function, so that

$$\langle a_1^{\dagger}a_1^{\dagger}a_1a_1\rangle=2\langle a_1^{\dagger}a_1\rangle . \tag{26}$$

In a similar manner, we obtain

$$\langle a_3^{\dagger}a_3^{\dagger}a_3a_3\rangle=2\langle a_3^{\dagger}a_3\rangle , \tag{27a}$$

$$\langle a_1^{\dagger}a_3^{\dagger}a_1a_3\rangle=\langle a_1^{\dagger}a_1\rangle\langle a_3^{\dagger}a_3\rangle+|\langle a_1a_3\rangle|^2 . \tag{27b}$$

On substituting for the fourth-order correlation functions in terms of second-order correlation functions from Eqs. (25)–(27) into Eq. (24), the following simple expression for λ is obtained:

$$\lambda=\frac{|\langle a_1a_3\rangle|^2}{\langle a_1^{\dagger}a_1\rangle\langle a_3^{\dagger}a_3\rangle}-1 . \tag{28}$$

The Cauchy-Schwarz inequality is therefore violated when

$$\langle a_1a_3\rangle>[\langle a_1^{\dagger}a_1\rangle\langle a_3^{\dagger}a_3\rangle]^{1/2} . \tag{29}$$

From Eq. (29) we conclude that the Cauchy-Schwarz inequality is violated when the combination tone contribution exceeds the square root of the product of the number operator contribution. In Fig. 2 we plot λ versus detuning $(\nu_2-\nu_1)T_2$ for the same parameters as used in Fig. 1. It is obvious from the figure that the Cauchy-Schwarz in-

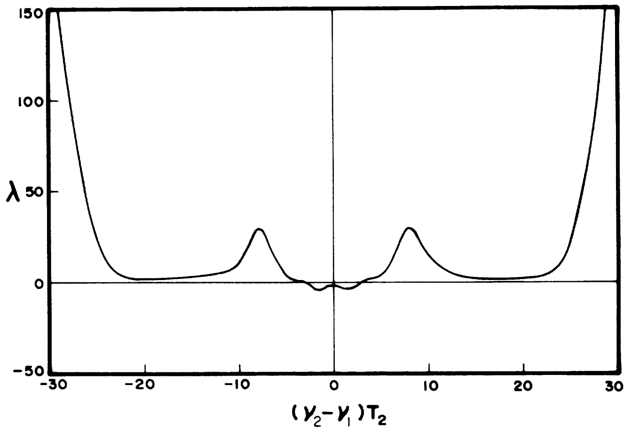

FIG. 2. λ vs detuning $(\nu_2-\nu_1)T_2$ for the same parameters as used in Fig. 1.

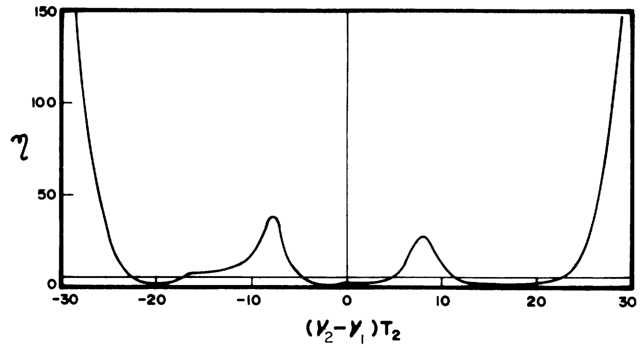

FIG. 3. η vs detuning $(\nu_2-\nu_1)T_2$ for the same parameters as used in Fig. 1.

equality is satisfied for zero detuning or small detunings, where the system behaves classically.

C. Violation of Bell's inequality

Recently Reid and Walls[22] performed detailed calculations to describe violation of Bell's inequality in four-mode, two-mode, and dissipative systems. In two-mode systems their model consists of a two-mode source with destruction operators a_1 and a_3 (see Fig. 6 of Ref. 22). Light from each of the two output modes is then split after passing through two 50% beam splitters. These two modes are then recombined on two other beam splitters which are oriented on two different angles θ and ϕ. Light is detected by photomultiplier tubes. The detected modes are the superposition of different combinations of the modes a_1 and a_3 and are of the form

$$c_1=a_1\cos\theta+a_3\sin\theta\ , \tag{30a}$$

$$c_2=-a_1\sin\theta+a_3\cos\theta\ , \tag{30b}$$

$$d_1=a_1\cos\phi-a_3\sin\phi\ , \tag{30c}$$

$$d_2=a_1\sin\phi+a_3\cos\phi\ . \tag{30d}$$

Then the correlation of detected modes is

$$\begin{aligned}\langle c_1^\dagger c_1 d_1^\dagger d_1\rangle &= \langle a_1^\dagger a_3^\dagger a_1 a_3\rangle \sin^2(\theta-\phi)\\ &\quad+\langle a_1^\dagger a_1^\dagger a_1 a_1\rangle(\cos^2\theta\cos^2\phi+\sin^2\theta\sin^2\phi)\ ,\end{aligned} \tag{31a}$$

$$\begin{aligned}\langle c_1^\dagger c_1 d_2^\dagger d_2\rangle &= \langle a_1^\dagger a_3^\dagger a_1 a_3\rangle \cos^2(\theta-\phi)\\ &\quad+\langle a_1^\dagger a_1^\dagger a_1 a_1\rangle(\cos^2\theta\sin^2\phi+\sin^2\theta\cos^2\phi)\ .\end{aligned} \tag{31b}$$

If we define the quantity

$$E(\theta,\phi)=\frac{\langle :(c_1^\dagger c_1-c_2^\dagger c_2)(d_1^\dagger d_1-d_2^\dagger d_2):\rangle}{\langle :(c_1^\dagger c_1+c_2^\dagger c_2)(d_1^\dagger d_1+d_2^\dagger d_2):\rangle}\ , \tag{32}$$

the condition for Bell's inequality becomes

$$-2\le E(\theta,\phi)-E(\theta,\phi')+E(\theta',\phi)+E(\theta',\phi')\le 2\ , \tag{33}$$

for $\theta=0$, $\phi=\pi/8$, $\theta'=\pi/4$, $\phi'=3\pi/8$, the violation of Bell's inequality is attained when

$$\frac{\langle a_1^\dagger a_3^\dagger a_1 a_3\rangle}{\langle a_1^\dagger a_3^\dagger a_1 a_3\rangle+\langle a_1^\dagger a_1^\dagger a_1 a_1\rangle}\ge 0.707\ . \tag{34}$$

By using Eqs. (25)–(27) and rearranging, it follows from Eq. (34) that Bell's inequality is violated if

$$\eta=\frac{\langle a_3^\dagger a_3\rangle}{\langle a_1^\dagger a_1\rangle}+\frac{|\langle a_1 a_3\rangle|^2}{\langle a_1^\dagger a_1\rangle^2}\ge 5\ . \tag{35}$$

In Fig. 3 we plot η versus detuning $(\nu_2-\nu_1)T_2$ for the same parameters as used in Fig. 1. It is clear from Fig. 3 that Bell's inequality is violated for large detunings where number operator $\langle a_1^\dagger a_1\rangle$, $\langle a_3^\dagger a_3\rangle$ contribution is negligible but $\langle a_1 a_3\rangle$ remains significant.

V. CONCLUSION

Our calculations show that nonclassical effects arise when combination tone contribution exceeds the expectation values for number operators. At large detuning the contribution of the number operators $\langle a_1^\dagger a_1\rangle$, $\langle a_3^\dagger a_3\rangle$ becomes negligible but the combination tone operator $\langle a_1 a_3\rangle$ is still effective. Hence at large detunings the Cauchy-Schwarz and Bell's inequalities are violated strongly. At zero detuning or small detunings the system behaves classically so the Cauchy-Schwarz and Bell's inequalities are valid in these regions.

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