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Intermodal entanglement in Raman processes

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The operator solution of a completely quantum mechanical Hamiltonian of the Raman processes is used here to investigate the possibility of obtaining intermodal entanglement between different modes involved in the Raman processes [e.g., pump mode, Stokes mode, vibration (phonon) mode and anti-Stokes mode]. Intermodal entanglement is reported between (a) pump mode and anti-Stokes mode, (b) pump mode and vibration (phonon) mode, (c) Stokes mode and vibration phonon mode, and (d) Stokes mode and anti-Stokes mode in the stimulated Raman processes for variation of the phase angle of complex eigenvalue α_1 of pump mode a. Some incidents of intermodal entanglement in the spontaneous and the partially spontaneous Raman processes are also reported. Further, it is shown that the specific choice of coupling constants may produce genuine entanglement among Stokes mode, anti-Stokes mode, and vibration-phonon mode. It is also shown that the two-mode entanglement not identified by Duan's criterion may be identified by Hillery-Zubairy criteria. It is further shown that intermodal entanglement, intermodal antibunching, and intermodal squeezing are independent phenomena.

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I. INTRODUCTION

Entanglement is one of the most important resources for quantum communication and quantum information processing. For example, it is well known that entanglement is essential for teleportation, dense coding, quantum information splitting, etc. Thus we need entangled states to perform various important tasks related to quantum information theory. To do so, first we need a protocol to check whether or not a state generally mixed is entangled. This is a very important issue in quantum information science and several inseparability criteria have been proposed for this purpose ([1], and references therein). In 1996, Peres [2] proposed the first inseparability criterion based on negative eigenvalues of the partial transpose of the composite density operator. This criterion is sufficient and necessary for the detection of entanglement in (2×2) and (2×3) dimensional states but is not necessary for higher dimensional states (see [3], and references therein). Since the pioneering work of Peres, several other inseparability inequalities have been reported for two-mode and multimode states [3–14]. Most of these criteria only provide sufficient condition of inseparability. Further, these criteria may be classified into two sets [10]: A) set of criteria which cannot be directly tested through experiments [4,5] and B) set of criteria which can be tested experimentally [2,3,6–10]. Experimentally testable inequalities involve variance or higher order moments of some observables. Since the expectation values of physical observables can be measured experimentally, these set of inseparability criteria can be tested experimentally.

The aim of the present work is not to study the inseparability criteria in detail but to study the possibility of generation of a multipartite entangled state in two-photon stimulated Raman processes, as depicted in Fig. 1. The scheme is essentially a sequential double Raman process that can produce Stokes and anti-Stokes photons that show a highly nonclassical correlation [11] and macroscopic entanglement in a two-photon laser [12]. To study two-mode entanglement in two-photon Raman processes it would be reasonable to use three criteria from set B. To be precise, we have chosen the two criteria of Hillery and Zubairy [7,8] and the criterion of Duan *et al.* [3]. Since all three of these criteria are only sufficient, a particular criterion can detect only a subset of all sets of entangled states. Consequently, application of a single criterion may yield an incomplete result. This is why we have used three experimentally testable inseparability criteria for our investigation of intermodal entanglement in stimulated, spontaneous, and partially spontaneous Raman processes.

Nonclassical properties of these Raman processes have been extensively studied. Initial studies were restricted to the short-time approximation [15–17]. But recently some of the present authors have reported different nonclassical effects (such as squeezing, antibunching, intermodal antibunching, and sub-shot-noise photon number correlation) in stimulated and spontaneous Raman processes [18–21] without using traditional short-time approximation technique. Our solution of Raman processes, which does not involve short-time approximation, is found to reveal many facets of nonclassical effects which were undetected by short-time approximation technique. However,the possibility of observing intermodal entanglement has not been rigorously studied so far. This fact motivated us to study intermodal entanglement in doubleRaman processes. The present investigation is relevant for quantum communication for two reasons: first, because entanglement is an essential resource for quantum communication and, second, because the spontaneous Raman process

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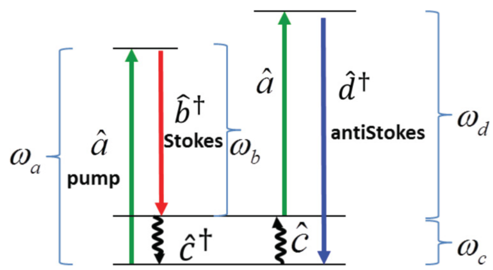

FIG. 1. (Color online) Two-photon stimulated Raman scheme. The pump photon is converted into a Stokes photon and a phonon. The pump photon can also mix with a phonon to produce an anti-Stokes photon.

is reported to be useful in the realization of quantum repeaters [22,23], which has its application in long-distance high-fidelity quantum communication.

Here it is worthy to note that Peřinová *et al.* [24] have recently studied the possibility of observing entanglement in the Raman process using the method of invariant subspace. They followed an independent approach and have numerically computed the time dependence of a measure of entanglement. Earlier, Kuznetsov *et al.* [25] studied entanglement in the stimulated Raman process considering only two modes (Stokes mode and phonon mode) and taking the pump mode as the classical light source. Naturally Kuznetsov *et al.*'s work illustrated an incomplete scenario and failed to observe intermodal entanglement involving the anti-Stokes mode and pump mode. To circumvent this limitation we have used here a completely quantum mechanical Hamiltonian. Further, Pathak, Křepelka, and Peřina [26] have recently investigated the possibilities of observing intermodal entanglement in Raman processes using the same Hamiltonian but with a short-time approximated solution. Their work is restricted by the intrinsic limitations of the short-time approximation. Such limitations may be circumvented by the analytical methods recently developed by us to study the stimulated Raman scheme [18–21]. Those methods are systematically used here and a relatively complete scenario of intermodal entanglement in Raman processes is presented. Interestingly, we have observed intermodal entanglement between (i) pump mode and anti-Stokes mode and (ii) Stokes mode and anti-Stokes mode. These two intermodal entanglements were not observed in earlier analytic studies [25,26]. The beauty of the present study lies in the fact that analytic expressions for the separability criterion are obtained by a completely quantum mechanical treatment where all four modes are considered quantum mechanical. If we look closely into the methodology adopted in the earlier studies, we quickly find that the approach adopted in the present paper is simpler and easily extendable to other physical systems which are described by bosonic Hamiltonians.

The paper is organized as follows. In Sec. II we describe the Hamiltonian of spontaneous and stimulated Raman processes and its operator solution. In Sec. III we use the solution to show that it is possible to observe intermodal entanglement in Raman processes. The inseparability criteria used for this purpose are also described in this section. Finally, Sec. IV is dedicated to conclusions and a brief summary of the results of the present study, and we also discuss the mutual relations among different nonclassical phenomena observed in Raman processes.

II. MODEL HAMILTONIAN

The Hamiltonian [15–21,27] of our interest is

$$\begin{aligned} H = {} & \omega_a a^\dagger a + \omega_b b^\dagger b + \omega_c c^\dagger c + \omega_d d^\dagger d \\ & + g(ab^\dagger c^\dagger + \mathrm{H.c.}) + \chi(acd^\dagger + \mathrm{H.c.}), \end{aligned} \tag{1}$$

where H.c. stands for the Hermitian conjugate. Throughout the present paper, we use $\hbar = 1$. The annihilation (creation) operators $a(a^\dagger)$, $b(b^\dagger)$, $c\left(c^\dagger\right)$, and $d(d^\dagger)$ correspond to the laser (pump) mode, Stokes mode, vibration (phonon) mode, and anti-Stokes mode, respectively. They obey the well-known boson commutation relations. The quantities ω_a, ω_b, ω_c, and ω_d correspond to the frequencies of pump mode a, Stokes mode b, vibration (phonon) mode c, and anti-Stokes mode d, respectively. The parameters g and χ are the Stokes and anti-Stokes coupling constants, respectively. The coupling constant g (χ) denotes the strength of coupling between the Stokes (anti-Stokes) mode and the vibrational (phonon) mode and depends on the actual interaction mechanism. The dimension of g and χ is that of frequency, and consequently, gt and χt are dimensionless. Further, gt and χt are very small compared to unity. In order to study the possibility of intermodal entanglement, we need simultaneous solutions of the following Heisenberg operator equations of motion for various field operators:

$$\begin{aligned} \dot{a} &= -i(\omega_a a + gbc + \chi cd), \quad \dot{b} = -i(\omega_b b + gac^\dagger), \\ \dot{c} &= -i(\omega_c c + gab^\dagger + \chi a^\dagger d), \quad \dot{d} = -(\omega_d d + \chi ac). \end{aligned} \tag{2}$$

The above set of coupled nonlinear differential operator equations (2), is not exactly solvable in the closed analytical form under weak pump conditions. But when the pump is very strong the operator a can be replaced by a c number and the above set of equations (2), can be solved exactly [17]. In order to solve these equations under weak pump approximation we use the perturbative approach. Our solutions are more general than the well-known short-time approximation. Details of the calculations are given in our previous papers [18–21]. Here we just note that under the weak pump approximation, the solutions of Eq. (2) assume the following form:

$$\begin{aligned} a(t) = {} & f_1 a(0) + f_2 b(0)c(0) + f_3 c^\dagger(0)d(0) + f_4 a^\dagger(0)b(0)d(0) + f_5 a(0)b(0)b^\dagger(0) + f_6 a(0)c^\dagger(0)c(0) + f_7 a(0)c^\dagger(0)c(0) \\ & + f_8 a(0)d^\dagger(0)d(0), \\ b(t) = {} & g_1 b(0) + g_2 a(0)c^\dagger(0) + g_3 a^2(0)d^\dagger(0) + g_4 c^{\dagger 2}(0)d(0) + g_5 b(0)c(0)c^\dagger(0) + g_6 b(0)a(0)a^\dagger(0), \\ c(t) = {} & h_1 c(0) + h_2 a(0)b^\dagger(0) + h_3 a^\dagger(0)d(0) + h_4 b^\dagger(0)c^\dagger(0)d(0) + h_5 c(0)a(0)a^\dagger(0) + h_6 c(0)b(0)b^\dagger(0) + h_7 c(0)d^\dagger(0)d(0) \\ & + h_8 c(0)a^\dagger(0)a(0), \\ d(t) = {} & l_1 d(0) + l_2 a(0)c(0) + l_3 a^2(0)b^\dagger(0) + l_4 b(0)c^2(0) + l_5 c^\dagger(0)c(0)d(0) + l_6 a(0)a^\dagger(0)d(0). \end{aligned} \tag{3}$$

The functions f_i, g_i, h_i, and l_i are evaluated from the dynamics under the initial conditions. In order to apply the boundary condition, we put $t = 0$ in the first term in Eq. (3). It is clear that $f_1(0) = g_1(0) = h_1(0) = l_1(0) = 1$ and $f_i(0) = g_i(0) = h_i(0) = l_i(0) = 0$ (for $i = 2$, 3, 4, 5, 6, 7, and 8). Under these initial conditions the corresponding solutions for $f_i(t)$, $g_i(t)$, $h_i(t)$, and $l_i(t)$ are obtained as given in the Appendix.

The solutions Eqs. (3) and (A1)–(A4) are valid up to the second orders in g and χ provided the dimensionless interaction time $gt < 1$ and/or $\chi t < 1$ such that the perturbation theory is respected. For example, f_2 rises indefinitely with an increase in time t. Clearly, the divergent nature of the parameters f_i, g_i, h_i, and l_i becomes more pronounced as t is increased. The secular nature is a direct outcome of the perturbation theory. In the present investigation the secular term is not a problem since we consider a small interaction time. A small interaction time also ensures that the damping term contributes insignificantly. Here $\Delta\omega_1 = \omega_b + \omega_c - \omega_a$ and $\Delta\omega_2 = \omega_a + \omega_c - \omega_d$. Normally, the detunings $\Delta\omega_1$ and $\Delta\omega_2$ are extremely small. In the present investigation, we, however, assume that a small (nonzero) detuning is present and hence $\Delta\omega_1 \neq 0$ and $\Delta\omega_2 \neq 0$. Here we have used $|\Delta\omega_1| = 0.1$ MHz and $|\Delta\omega_2| = 0.19$ MHz. Of course, in Eqs. (A1)–(A4) we have neglected the terms beyond the second order in g and χ. Now we may use Eq. (3) to obtain the temporal evolution of the number operators of various modes as

$$\begin{aligned} N_a(t) = {} & |f_1|^2 a^\dagger(0)a(0) + |f_2|^2 b^\dagger(0)c^\dagger(0)b(0)c(0) + |f_3|^2 c(0)d(0)^\dagger c^\dagger(0)d(0) + \{f_1^* f_2 a^\dagger(0)b(0)c(0) + f_1^* f_3 a^\dagger(0)c^\dagger(0)d(0) \\ & + f_1^* f_4 a^\dagger(0)a^\dagger(0)b(0)d(0) + f_1^* f_5[a^\dagger(0)a(0) + a^\dagger(0)a(0)b^\dagger(0)b(0)] + f_1^* f_6 a^\dagger(0)a(0)c^\dagger(0)c(0) \\ & + f_1^* f_7 a^\dagger(0)a(0)c^\dagger(0)c(0) + f_1^* f_8 a^\dagger(0)a(0)d^\dagger(0)d(0) + f_2^* f_3 b^\dagger(0)c^{\dagger^2}(0)d(0) + \text{H.c.}\}, \end{aligned} \tag{4}$$

$$\begin{aligned} N_b(t) = {} & |g_1|^2 b^\dagger(0)b(0) + |g_2|^2 a^\dagger(0)c(0)a(0)c^\dagger(0) + \{g_1^* g_2 b^\dagger(0)a(0)c^\dagger(0) + g_1^* g_3 b^\dagger(0)a^2(0)d^\dagger(0) + g_1^* g_4 b^\dagger(0)c^{\dagger^2}(0)d(0) \\ & + g_1^* g_5[b^\dagger(0)b(0) + b^\dagger(0)b(0)c^\dagger(0)c(0)] + g_1^* g_6[b^\dagger(0)b(0) + b^\dagger(0)b(0)a^\dagger(0)a(0)] + \text{H.c.}\}, \end{aligned} \tag{5}$$

$$\begin{aligned} N_c(t) = {} & |h_1|^2 c^\dagger(0)c(0) + |h_2|^2 a^\dagger(0)b(0)a(0)b^\dagger(0) + |h_3|^2 a(0)d^\dagger(0)a^\dagger(0)d(0) + \{h_1^* h_2 c^\dagger(0)a(0)b^\dagger(0) + h_1^* h_3 c^\dagger(0)a^\dagger(0)d(0) \\ & + h_1^* h_4 c^\dagger(0)b^\dagger(0)c^\dagger(0)d(0) + h_1^* h_5[c^\dagger(0)c(0) + c^\dagger(0)c(0)a^\dagger(0)a(0)] + h_1^* h_6[c^\dagger(0)c(0) + c^\dagger(0)c(0)b^\dagger(0)b(0)] \\ & + h_1^* h_7 c^\dagger(0)c(0)d^\dagger(0)d(0) + h_1^* h_8 c^\dagger(0)c(0)a^\dagger(0)a(0) + h_2^* h_3 a^{\dagger^2}(0)b(0)d(0) + \text{H.c.}\}, \end{aligned} \tag{6}$$

and

$$\begin{aligned} N_d(t) = {} & |l_1|^2 d^\dagger(0)d(0) + |l_2|^2 a^\dagger(0)a(0)c^\dagger(0)c(0) + \{l_1^* l_2 a(0)c(0)d^\dagger(0) + l_1^* l_3 a^2(0)b^\dagger(0)d^\dagger(0) + l_1^* l_4 b(0)c^2(0)d^\dagger(0) \\ & + l_1^* l_5 c^\dagger(0)c(0)d^\dagger(0)d(0) + l_1^* l_6 d^\dagger(0)d(0)[1 + a^\dagger(0)a(0)] + \text{H.c.}\}. \end{aligned} \tag{7}$$

In the following section these number operators are used to study intermodal entanglement in Raman processes.

III. INTERMODAL ENTANGLEMENT

In order to investigate the intermodal entanglement for various coupled modes, we assume that all photon and phonon modes are initially coherent. In other words, the composite boson field consisting of photons and phonon are in the initial coherent state. Therefore, the composite coherent state arises from the product of the coherent states $|\alpha_1\rangle$, $|\alpha_2\rangle$, $|\alpha_3\rangle$, and $|\alpha_4\rangle$, which are the eigenkets of a, b, c, and d, respectively. Thus the initial composite state is

$$|\psi(0)\rangle = |\alpha_1\rangle \otimes |\alpha_2\rangle \otimes |\alpha_3\rangle \otimes |\alpha_4\rangle. \tag{8}$$

It is clear that the initial state is separable. Now the field operator $a(t)$ operating on such a multimode coherent state gives rise to the complex eigenvalue $\alpha_1(t)$. Hence we have

$$a(0)|\psi(0)\rangle = \alpha_1|\psi(0)\rangle, \tag{9}$$

where $|\alpha_1|^2$ is the number of input photons in pump mode a. In a similar fashion we have three more complex amplitudes, $\alpha_2(t)$, $\alpha_3(t)$, and $\alpha_4(t)$, corresponding to the Stokes, vibrational (phonon), and anti-Stokes field mode operators b, c, and d, respectively. Clearly, for a spontaneous process, the complex amplitudes are $\alpha_2 = \alpha_3 = \alpha_4 = 0$ and $\alpha_1 \neq 0$. For a partial spontaneous process, the complex amplitude α_1 and any one of the remaining three eigenvalues are not equal to 0, while the other two complex amplitudes are 0. On the other hand, for a stimulated process, the complex amplitudes are not necessarily 0. In our present investigation we consider $\alpha_1 = |\alpha_1|\, e^{-i\phi}$ and the other eigenvalues for the Stokes, vibrational (phonon), and anti-Stokes field modes are real. The aim of the present work is to investigate the possibility of intermodal entanglement in spontaneous, partially spontaneous, and stimulated Raman processes. To do so, let us begin with the investigation of two-mode entanglement using Hillery and Zubairy's criteria.

A. Two-mode entanglement

There are two criteria due to Hillery and Zubairy [7,8]. The first one is

$$\langle N_a N_b\rangle - |\langle ab^\dagger\rangle|^2 < 0. \tag{10}$$

On the other hand, the second criterion is given by

$$\langle N_a\rangle\langle N_b\rangle - |\langle ab\rangle|^2 < 0. \tag{11}$$

From here onward we refer to these criteria as the HZ-1 and HZ-2 criteria, respectively. In addition to these two criteria, we also use Duan's inseparability criterion due to Duan *et al.* [3]:

$$\langle \Delta a^\dagger \Delta a\rangle\langle \Delta b^\dagger \Delta b\rangle - |\langle \Delta a \Delta b\rangle|^2 < 0. \tag{12}$$

In the criteria Eqs. (10)–(12), a and b are annihilation operators for two arbitrary modes. They are not limited to the pump mode and the Stokes mode.

We note that all the above criteria are only sufficient (not necessary) for detection of entanglement. Keeping this fact in mind, we have applied all these criteria to study intermodal entanglement between different modes of the Raman Hamiltonian and have observed intermodal entanglement in various situations.

Let us first investigate the possibility of two-mode entanglement in the Raman process using the HZ-1 criterion. From Eqs. (3)–(5) and (8) we obtain

$$\begin{aligned}\langle N_a N_b\rangle &- |\langle a b^\dagger\rangle|^2 \\ &= |f_3|^2|\alpha_2|^2|\alpha_4|^2 + |g_2|^2(|\alpha_1|^4 - |\alpha_1|^2|\alpha_2|^2).\end{aligned} \tag{13}$$

Consequently, for a spontaneous Raman process Eq. (13) reduces to

$$\langle N_a N_b\rangle - |\langle a b^\dagger\rangle|^2 = |g_2|^2|\alpha_1|^4. \tag{14}$$

It is clear that the right-hand side (r.h.s.) of Eq. (14) is always positive. Hence the HZ-1 criterion does not show any signature of intermodal entanglement between pump and Stokes modes in the spontaneous Raman process. For a partially spontaneous Raman process ($|\alpha_1| \neq 0$, $|\alpha_2| \neq 0$, $|\alpha_3| = |\alpha_4| = 0$), Eq. (13) reduces to

$$\langle N_a N_b\rangle - |\langle a b^\dagger\rangle|^2 = |g_2|^2(|\alpha_1|^2 - |\alpha_2|^2)|\alpha_1|^2. \tag{15}$$

It is clear that entanglement is possible in the partially spontaneous Raman process only when $|\alpha_2|^2 > |\alpha_1|^2$; i.e., the number of Stokes photons is more than the number of pump photons, which is not the usual case. According to the HZ-1 criterion of Eq. (10), it is clear that the negative values on the r.h.s. of Eq. (13) would indicate the presence of intermodal entanglement between the pump mode and the Stokes mode in the stimulated Raman process. To investigate the possibility of intermodal entanglement in the stimulated Raman process we have used $\chi = g = 10^5$ Hz, $|\alpha_1| = 10$, $|\alpha_2| = 8$, $|\alpha_3| = 0.01$, and $|\alpha_4| = 1$ [28]. We have plotted the r.h.s. of Eq. (13) in Fig. 2(a), which does not show any signature of intermodal entanglement between the pump mode and the Stokes mode in the stimulated Raman process.

Here we would like to note that once we have an analytic expression for the HZ-1 or HZ-2 or Duan criteria in the stimulated Raman process, it is straightforward to study the special cases of (i) a spontaneous Raman process, where $\alpha_2 = \alpha_3 = \alpha_4 = 0$ but $\alpha_1 \neq 0$, and (ii) a partial spontaneous Raman process, where $\alpha_1 \neq 0$ and any one of the other three α_i ($i = 2,3,4$) is nonzero.

The same technique used in the above case is now adopted to obtain the following equations for the study of intermodal entanglement in the stimulated Raman process using the HZ-1 criterion:

$$\begin{aligned}\langle N_b N_c\rangle - |\langle b c^\dagger\rangle|^2 &= |g_2|^2(3|\alpha_1|^2|\alpha_3|^2 + 3|\alpha_1|^2|\alpha_2|^2 + |\alpha_1|^2 - |\alpha_2|^2|\alpha_3|^2) + |h_3|^2|\alpha_2|^2|\alpha_4|^2 \\ &\quad + [(h_1^* h_2 \alpha_1 \alpha_2^* \alpha_3^* + 2 g_4^* g_1 \alpha_2 \alpha_3^2 \alpha_4^* + h_2 h_3^* \alpha_1^2 \alpha_2^* \alpha_4^*) + \text{c.c.}],\end{aligned} \tag{16}$$

$$\langle N_a N_d\rangle - |\langle a d^\dagger\rangle|^2 = |f_3|^2(|\alpha_3|^2 + |\alpha_4|^4 + |\alpha_1|^2|\alpha_3|^2 - |\alpha_1|^2|\alpha_4|^2) - |l_2|^2(|\alpha_3|^2 + |\alpha_1|^2|\alpha_3|^2), \tag{17}$$

$$\langle N_b N_d\rangle - |\langle b d^\dagger\rangle|^2 = |g_2|^2|\alpha_1|^2|\alpha_4|^2 + [\{l_1^* l_3 \alpha_1^2 \alpha_2^* \alpha_4^* + \text{c.c.}\}], \tag{18}$$

$$\langle N_c N_d\rangle - |\langle c d^\dagger\rangle|^2 = |l_2|^2(2|\alpha_1|^2 + 2|\alpha_1|^2|\alpha_4|^2 - 2|\alpha_4|^2 - |\alpha_4|^4 - |\alpha_3|^2|\alpha_4|^2) + |h_2|^2|\alpha_1|^2|\alpha_4|^2, \tag{19}$$

$$\begin{aligned}\langle N_a N_c\rangle - |\langle a c^\dagger\rangle|^2 &= |f_2|^2(2|\alpha_1|^2 + |\alpha_1|^4 + |\alpha_1|^2|\alpha_3|^2 - 4|\alpha_2|^2 - 2|\alpha_1|^2|\alpha_2|^2 - 2|\alpha_2|^2|\alpha_3|^2) + |f_3|^2(|\alpha_4|^2 + 3|\alpha_3|^2|\alpha_4|^2 \\ &\quad + 3|\alpha_1|^2|\alpha_4|^2 - |\alpha_1|^2|\alpha_3|^2) + [(f_1^* f_3 \alpha_1^* \alpha_3^* \alpha_4 + h_2^* h_3 \alpha_1^{*2} \alpha_2 \alpha_4 + f_2^* f_3 \alpha_2^* \alpha_3^{*2} \alpha_4) + \text{c.c.}].\end{aligned} \tag{20}$$

The r.h.s.'s of Eqs. (16)–(20) are plotted in Figs. 2(b)–2(f). It is interesting to note that the presence of intermodal entanglement in the stimulated Raman process is observed between (i) the Stokes mode and the vibration (phonon) mode [Fig. 2(b)], (ii) the pump mode and the anti-Stokes mode [Fig. 2(c)], (iii) the Stokes mode and the anti-Stokes mode [Fig. 2(d)], and (iv) the pump mode and the vibration mode [Fig. 2(f)]. However, no signature of intermodal entanglement is observed in the other two cases [Figs. 2(a) and 2(e)]. Further, it does not show the presence of genuine entanglement among any three modes of the system. Interestingly, with a suitable choice of the complex eigenvalues α_i it is possible to observe the signature of intermodal entanglement using the HZ-1 criteria in a partially spontaneous Raman process in several ways but no such signature is observed for the completely spontaneous Raman process.

Since the HZ-1 criterion is only sufficient, we might have failed to detect some intermodal entanglement. In an attempt to detect such intermodal entanglement using the HZ-2 criterion (11), we have used Eqs. (3) and (4)–(8) to yield

$$\langle N_a\rangle\langle N_b\rangle - |\langle a b\rangle|^2 = |g_2|^2|\alpha_1|^4 + |f_3|^2|\alpha_2|^2|\alpha_4|^2 - [(g_1^* g_6 + f_1^* f_2 g_1^* g_2)|\alpha_1|^2|\alpha_2|^2 + \text{c.c.}] \tag{21}$$

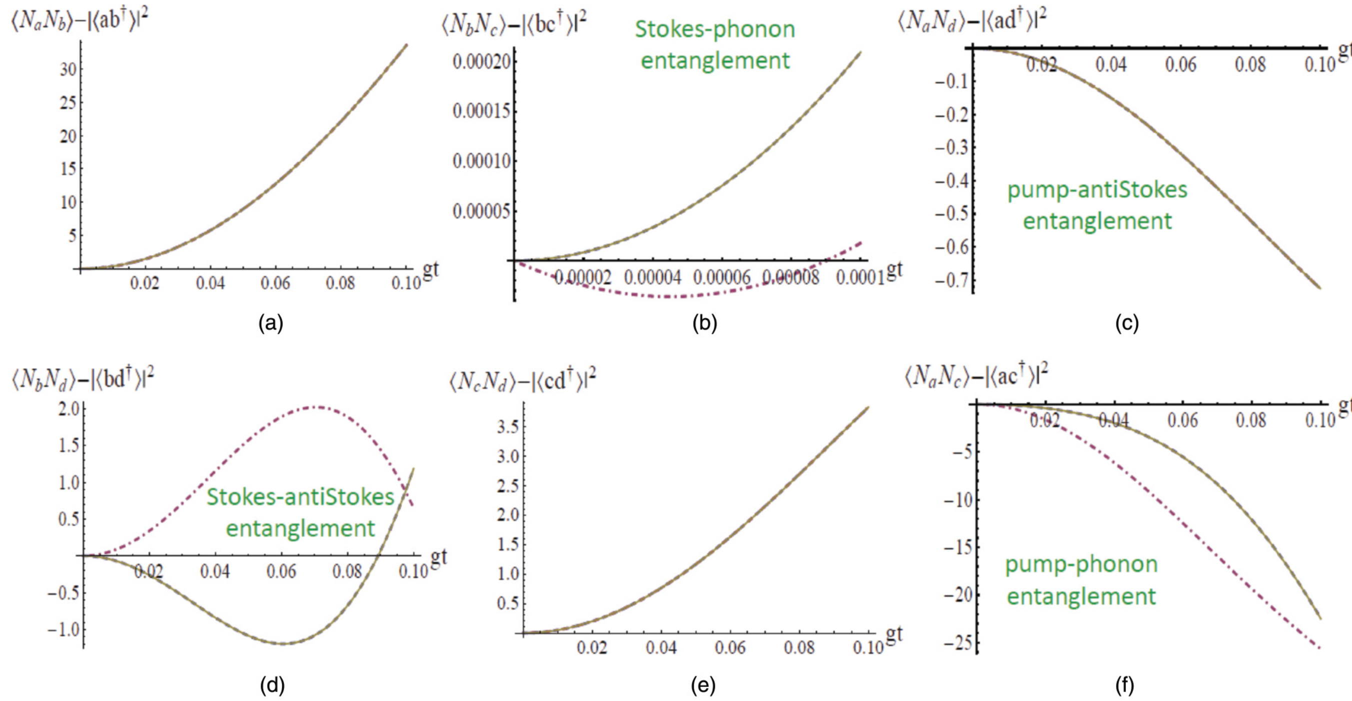

FIG. 2. (Color online) Intermodal entanglement in the stimulated Raman process using the HZ-1 criterion. The dotted line, dash-dotted line, and solid line represent the phase angle of the input complex amplitude α_1 for $\phi = 0$, π, and $\pi/2$, respectively. (a) Intermodal entanglement is not observed between pump mode and Stokes mode. (b) Intermodal entanglement is observed between Stokes mode and vibration-phonon mode for $\phi = \pi/2$. (c) Intermodal entanglement is observed between pump mode and anti-Stokes mode. (d) Intermodal entanglement is observed between Stokes mode and anti-Stokes mode for $\phi = 0$ and $\pi/2$. (e) The signature of intermodal entanglement is not observed between vibration-phonon mode and anti-Stokes mode. (f) Intermodal entanglement is observed between pump mode and vibration-phonon mode.

and

$$\langle N_b\rangle\langle N_c\rangle - |\langle bc\rangle|^2 = |g_2|^2|\alpha_1|^2|\alpha_3|^2 - |h_2|^2(1+|\alpha_2|^2)|\alpha_1|^2 + |h_3|^2|\alpha_2|^2|\alpha_4|^2 - [(h_1^* h_2 \alpha_1 \alpha_2^* \alpha_3^* + (h_1 h_4^* + g_1 g_2^* h_1 h_3^*)\alpha_2 \alpha_3^2 \alpha_4^* + h_2^* h_3 \alpha_1^{*2} \alpha_2 \alpha_4 + h_1^* h_6 |\alpha_2|^2|\alpha_3|^2 + g_1 g_2^* h_1^* h_2 |\alpha_1|^2|\alpha_3|^2) + \text{c.c.}], \tag{22}$$

$$\langle N_a\rangle\langle N_d\rangle - |\langle ad\rangle|^2 = |f_3|^2|\alpha_4|^4 - (l_1^* l_6 |\alpha_1|^2|\alpha_4|^2 + \text{c.c}), \tag{23}$$

$$\langle N_b\rangle\langle N_d\rangle - |\langle bd\rangle|^2 = |g_2|^2|\alpha_1|^2|\alpha_4|^2 - \left(l_1^* l_3 \alpha_1^2 \alpha_2^* \alpha_4^* + \text{c.c}\right), \tag{24}$$

$$\langle N_c\rangle\langle N_d\rangle - |\langle cd\rangle|^2 = |h_2|^2|\alpha_1|^2|\alpha_4|^2 + |h_3|^2|\alpha_4|^4 - (l_1^* l_5 |\alpha_3|^2|\alpha_4|^2 + \text{c.c}), \tag{25}$$

$$\langle N_a\rangle\langle N_c\rangle - |\langle ac\rangle|^2 = |h_2|^2|\alpha_1|^4 - |h_3|^2(|\alpha_4|^2 + |\alpha_1|^2|\alpha_4|^2) + |f_3|^2|\alpha_3|^2|\alpha_4|^2 - \big[\big(h_1^* h_3 \alpha_1^* \alpha_3^* \alpha_4 + h_1^* h_8 |\alpha_1|^2|\alpha_3|^2 + h_2^* h_3 \alpha_1^{*2} \alpha_2 \alpha_4 - h_1^* h_5 |\alpha_1|^2|\alpha_3|^2 + f_1^* f_2 h_3^* h_1 \alpha_2 \alpha_3^2 \alpha_4^* + f_1^* f_3 h_3^* h_1 |\alpha_3|^2|\alpha_4|^2\big) + \text{c.c.}\big]. \tag{26}$$

From the closed-form analytic expressions Eqs. (21)–(26), it is possible to obtain the signature of intermodal entanglement in various cases. Interestingly, we obtain intermodal entanglement between Stokes mode and vibration mode for the spontaneous Raman process. The intermodal entanglement Eqs. (21)–(26) for stimulated Raman processes are illustrated in Figs. 3(a)–3(f). In accordance with the HZ-2 criterion, negative values of the ordinates indicate the signature of entanglement. Therefore, the intermodal entanglement is observed in (i) Stokes mode and vibration mode, (ii) Stokes mode and anti-Stokes mode, and (iii) pump mode and vibration mode. However, there is no signature of intermodal entanglement in the remaining cases for stimulated Raman processes. It is possible to obtain intermodal entanglement for various partially spontaneous Raman processes. However, these results are not exhibited in the present text. It is interesting to note that the HZ-2 criterion failed to detect intermodal entanglement between the pump and the anti-Stokes modes. Thus the Raman process provides a very nice example of a physical system where it can be shown with a physical example that these inseparability criteria are only sufficient. Still there are two situations where we have not found the signature of intermodal entanglement. Let us see what happens when we apply another sufficient but not necessary criterion of inseparability.

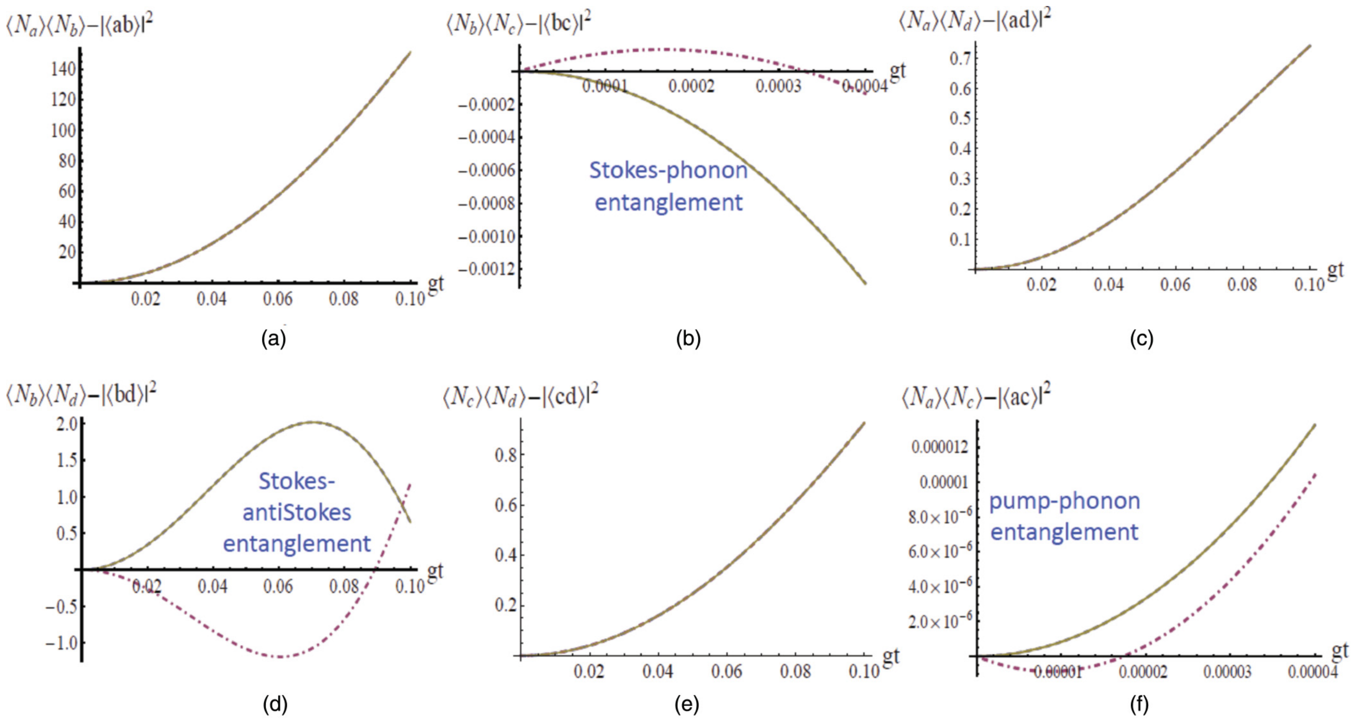

FIG. 3. (Color online) Intermodal entanglement in the stimulated Raman process using the HZ-2 criterion. The dotted line, dash-dotted line, and solid line are used for the phase angle of the input complex amplitude α_1 for $\phi = 0$, $\pi/2$, and π, respectively. (a) Intermodal entanglement is not observed between Stokes mode and pump mode. (b) Intermodal entanglement is observed between Stokes mode and vibration-phonon mode. (c) The signature of intermodal entanglement is not observed between pump mode and anti-Stokes mode. (d) Intermodal entanglement is observed between Stokes mode and anti-Stokes mode only for $\phi = \pi/2$. (e) Intermodal entanglement is not observed between vibration-phonon mode and anti-Stokes mode. (f) Intermodal entanglement (time dependent) is observed between pump mode and vibration-phonon mode for $\phi = \pi/2$.

Now the Duan criterion, Eq. (12), for the intermodal entanglement can also be written as [29]

$$D = \langle(\Delta u)^2\rangle + \langle(\Delta v)^2\rangle - 2 < 0, \tag{27}$$

where

$$\hat{u} = \frac{1}{\sqrt{2}}[(a + a^\dagger) + (b + b^\dagger)] \tag{28}$$

and

$$\hat{v} = \frac{1}{i\sqrt{2}}[(a - a^\dagger) + (b - b^\dagger)]. \tag{29}$$

Using Eqs. (3), (8), and (27)–(29) we can obtain an analytic expression for D on the left-hand side of the Duan *et al.* criterion, Eq. (27).

For modes a and b,

$$D_{ab} = 2\big\{|f_3||\alpha_4|^2 + |g_2|^2|\alpha_1|^2 + \tfrac{1}{2}[(f_1 g_6^* + f_5 g_1^*)\alpha_1\alpha_2^* + (2f_1 g_3^* + f_4 g_1^* + f_3 g_2^*)\alpha_1^*\alpha_4 + \text{c.c.}]\big\}; \tag{30}$$

for modes a and c,

$$D_{ac} = 2\big\{|f_3||\alpha_4|^2 + |h_2|^2|\alpha_1|^2 + |h_3|^2|\alpha_4|^2 + \tfrac{1}{2}[(f_1 h_5^* + f_6 h_1^* + f_3 h_3^* + f_7 h_1^* + f_1 h_8^*)\alpha_1\alpha_3^*] + \text{c.c.}\big\}; \tag{31}$$

for modes b and c,

$$D_{bc} = 2\big\{|g_2|^2|\alpha_1|^2 + |h_2|^2|\alpha_1|^2 + |h_3|^2|\alpha_4|^2 + \tfrac{1}{2}[(g_1 h_6^* + g_5 h_1^* + g_2 h_2^*)\alpha_3^*\alpha_2] + \text{c.c.}\big\}; \tag{32}$$

for modes a and d,

$$D_{ad} = 2\big\{|f_3||\alpha_4|^2 + \tfrac{1}{2}[(f_1 l_2^* + f_3 l_1^*)\alpha_3^* + (2f_1 l_3^* + f_4 l_1^* + f_2 l_2^*)\alpha_1^*\alpha_2 + f_8 l_1^*\alpha_1\alpha_4^* + \text{c.c.}]\big\}; \tag{33}$$

for modes c and d,

$$D_{cd} = 2\big\{|h_2|^2|\alpha_1|^2 + |h_3|^2|\alpha_4|^2 + \tfrac{1}{2}[(2l_4 h_1^* + l_2 h_2^* + l_1 h_4^*)\alpha_2\alpha_3 + (l_5 h_1^* + l_1 h_7^*)\alpha_3^*\alpha_4 + +\text{c.c.}]\big\}; \tag{34}$$

and for modes b and d,

$$D_{bd} = 2\big\{|g_2|^2|\alpha_1|^2 + \tfrac{1}{2}[(l_4 g_1^* + l_2 g_2^* + l_1 g_4^*)\alpha_3^2 + \text{c.c.}]\big\}. \tag{35}$$

The right-hand sides of Eqs. (30)–(35) are plotted in Figs. 4(a)–4(f). It is clear that intermodal entanglement is observed only between the pump mode and the anti-Stokes mode. The criterion is nonconclusive in all other cases. This is so because the Duan criterion is only sufficient.

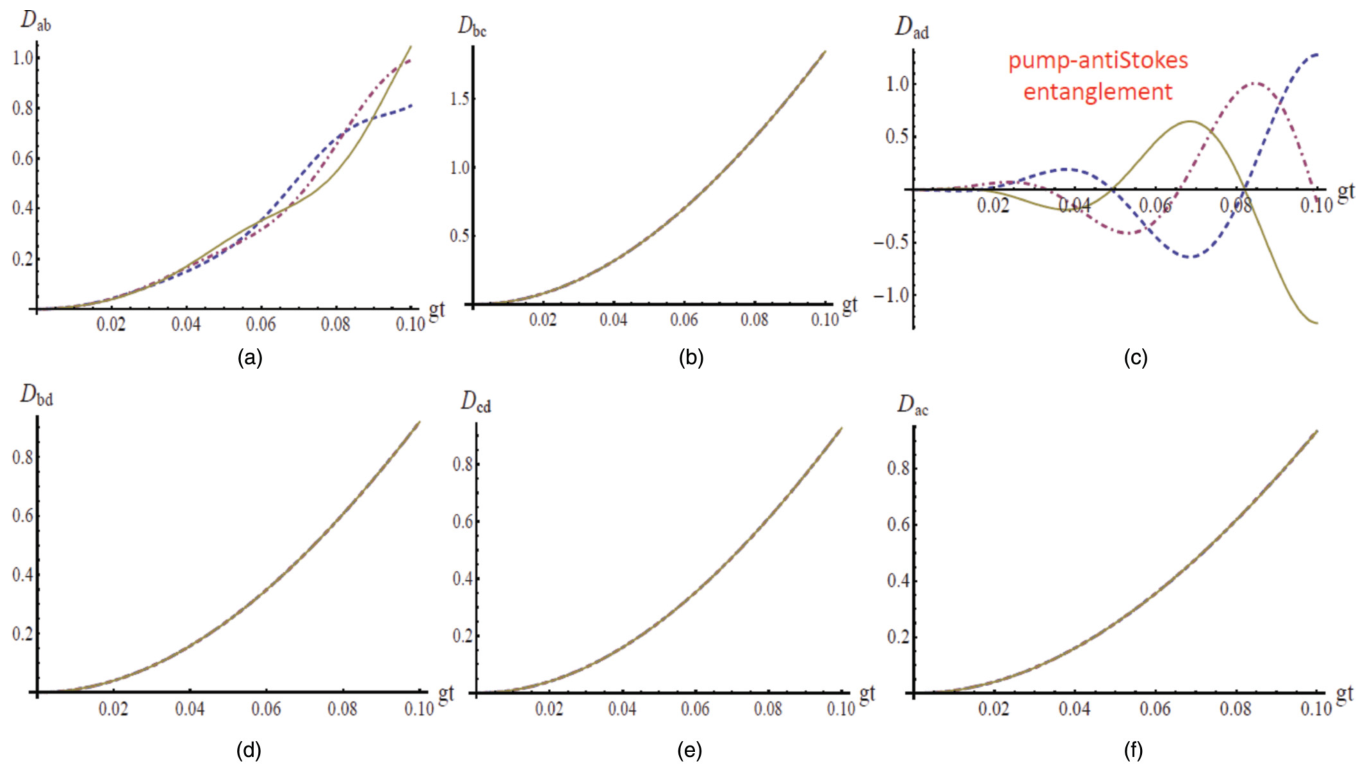

FIG. 4. (Color online) Intermodal entanglement in the stimulated Raman process using the Duan criterion. The dotted line, dash-dotted line, and solid line represent the phase angle of the input complex amplitude α_1 for $\phi = 0$, $\pi/2$, and π, respectively. Intermodal entanglement is only observed between pump mode and anti-Stokes mode (c).

IV. CONCLUSIONS

We have clearly established that the stimulated Raman process can produce intermodal entanglement. The observations are summarized in Table I. Here it would be apt to note that recently Pathak, Křepelka, and Peřina [26] have investigated the possibilities of observing intermodal entanglement in Raman processes using an approximated short-time solution. They have identified intermodal entanglement in the *pump-phonon ac* and *Stokes-phonon bc* modes only. However, in addition to those two modes we have observed intermodal entanglement in the *pump-anti-Stokes ad* and *Stokes-anti-Stokes bd* modes too. In addition to these, we explore the various possibilities of getting intermodal entanglement in partial spontaneous Raman processes. In this way, our solution is found to be more powerful compared to those of the solutions of Raman processes under short-time approximation. Further, the use of short-time solutions led to a monotonic nature of the entanglement parameter as seen in Eqs. (11) and (12) in Ref. [26]. Our solution is valid for all times and hence the entanglement parameters are free of this particular problem, which is generally a characteristic of short-time solutions. Another earlier effort to study intermodal entanglement in Raman processes, by Kuznetsov [25], was restricted to the study of intermodal entanglement between the Stokes mode and the vibration mode, as they considered a simplified two-mode Hamiltonian. Thus the use of a completely quantum mechanical description of the Raman process, our solution, and the strategy of using more than one inseparability criterion have helped us to obtain a relatively more complete picture

TABLE I. Relation among different nonclassical phenomena observed in the stimulated Raman process. nc, nonconclusive.

	HZ-1			HZ-2			Duan			Antibunching [19]	Squeezing [18]
Intermode	$\phi = 0$	$\pi/2$	π	$\phi = 0$	$\pi/2$	π	$\phi = 0$	$\pi/2$	π		
ab	nc	nc	nc	nc	nc	nc	nc	nc	nc	Possible	Possible
ac	Entangled	Entangled	Entangled	nc	Entangled (time dependent)	nc	nc	nc	nc	Bunching	Possible
ad	Entangled	Entangled	Entangled	nc	nc	nc	Entangled	Entangled	Entangled	Antibunching	Possible
bc	nc	Entangled (time dependent)	nc	Entangled	nc	Entangled	nc	nc	nc	Bunching	Possible
bd	Entangled	nc	Entangled	nc	Entangled	nc	nc	nc	nc	Antibunching	Not possible
cd	nc	nc	nc	nc	nc	nc	nc	nc	nc	Bunching	Possible

of the intermodal entanglement in Raman processes. Further, if we look at the possibilities of different kinds of nonclassicalities summarized in Table I (see the rows corresponding to the *ac*, *bc*, and *bd* modes), then we quickly recognize that the existence of any one of the nonclassical phenomena does not depend on the presence of the other. To be precise, entanglement, antibunching, and squeezing are nonclassical phenomena but they are independent of each other. However, the Duan criterion in the present form implies intermodal squeezing in one of the quadrature variables, but the converse is not true. This fact can be clearly seen in Table I, where we note that, except in the *bd* mode, intermodal squeezing is possible in all other coupled modes. However, the Duan criterion of intermodal entanglement is satisfied only for the *ad* mode.

In quantum optics, physical systems (matter-field interactions) are usually described by multimode bosonic Hamiltonians. The procedure followed in the present work may be applied directly to those systems to study intermodal entanglement. It is expected that most of these systems will show intermodal entanglement. This is so because most of the quantum states are entangled. Separability is a very special case. A natural question should arise at this point: If entanglement is so common, why are we looking for it? *The answer lies in the fact that entanglement is one of the most important resources for quantum information processing and quantum communication but still it is not very easy to produce useful multipartite entanglement.* As many of the quantum optical systems described by the bosonic Hamiltonian (including the system studied here) are experimentally achievable, useful intermodal entanglement may be produced by them. Entanglement obtained in such a system is expected to find application in controlled quantum teleportation, quantum information splitting, dense coding, direct secured quantum communication, etc. We conclude this paper with the optimistic view that the present work will motivate others to look for theoretical and experimental generation of useful multimode entanglement in other quantum optical systems.

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APPENDIX: PARAMETERS FOR THE SOLUTIONS IN EQ. (3)

$$
\begin{aligned}
f_1 &= \exp(-i\omega_a t), \quad f_2 = \frac{g e^{-i\omega_a t}}{\Delta\omega_1}(e^{-i\Delta\omega_1 t}-1),\\
f_3 &= -\frac{\chi e^{-i\omega_a t}}{\Delta\omega_2}(e^{i\Delta\omega_2 t}-1),\\
f_4 &= -\frac{\chi g e^{-i\omega_a t}}{\Delta\omega_1}\left[\frac{e^{-i(\Delta\omega_1-\Delta\omega_2)t}-1}{\Delta\omega_1-\Delta\omega_2}+\frac{e^{i\Delta\omega_2 t}}{\Delta\omega_2}\right]\\
&\quad -\frac{\chi g e^{-i\omega_a t}}{\Delta\omega_2}\left[\frac{e^{-i(\Delta\omega_1-\Delta\omega_2)t}-1}{\Delta\omega_1-\Delta\omega_2}-\frac{e^{-i\Delta\omega_1 t}}{\Delta\omega_1}\right],\\
f_5 &= \frac{g^2 e^{-i\omega_a t}}{\Delta\omega_1^2}(e^{-i\Delta\omega_1 t}-1)+\frac{ig^2 t e^{-i\omega_a t}}{\Delta\omega_1}, \quad f_6 = f_5,\\
f_7 &= \frac{\chi^2 e^{-i\omega_a t}}{\Delta\omega_2^2}(e^{i\Delta\omega_2 t}-1)-\frac{i\chi^2 t e^{-i\omega_a t}}{\Delta\omega_2}, \quad f_8 = -f_7.
\end{aligned}
\tag{A1}
$$

$$
\begin{aligned}
g_1 &= \exp(-i\omega_b t), \quad g_2 = -\frac{g e^{-i\omega_b t}}{\Delta\omega_1}(e^{i\Delta\omega_1 t}-1),\\
g_3 &= \frac{\chi g e^{-i\omega_b t}}{\Delta\omega_2(\Delta\omega_1-\Delta\omega_2)}[e^{i(\Delta\omega_1-\Delta\omega_2)t}-1]\\
&\quad -\frac{\chi g e^{-i\omega_b t}}{\Delta\omega_2\Delta\omega_1}(e^{i\Delta\omega_1 t}-1),\\
g_4 &= \frac{\chi g e^{-i\omega_b t}}{\Delta\omega_2(\Delta\omega_1+\Delta\omega_2)}[e^{i(\Delta\omega_1+\Delta\omega_2)t}-1]\\
&\quad -\frac{\chi g e^{-i\omega_b t}}{\Delta\omega_2\Delta\omega_1}(e^{i\Delta\omega_1 t}-1),\\
g_5 &= \frac{g^2 e^{-i\omega_b t}}{\Delta\omega_1^2}(e^{i\Delta\omega_1 t}-1)-\frac{ig^2 t e^{-i\omega_b t}}{\Delta\omega_1}, \quad g_6 = -g_5.
\end{aligned}
\tag{A2}
$$

$$
\begin{aligned}
h_1 &= \exp(-i\omega_c t), \quad h_2 = -\frac{g e^{-i\omega_c t}}{\Delta\omega_1}(e^{i\Delta\omega_1 t}-1),\\
h_3 &= -\frac{\chi e^{-i\omega_c t}}{\Delta\omega_2}(e^{i\Delta\omega_2 t}-1),\\
h_4 &= \frac{\chi g e^{-i\omega_c t}}{\Delta\omega_2}\left[\frac{e^{i(\Delta\omega_1+\Delta\omega_2)t}-1}{\Delta\omega_1+\Delta\omega_2}-\frac{e^{i\Delta\omega_1 t}}{\Delta\omega_1}\right]\\
&\quad -\frac{\chi g e^{-i\omega_c t}}{\Delta\omega_1}\left[\frac{e^{i(\Delta\omega_1+\Delta\omega_2)t}-1}{\Delta\omega_1+\Delta\omega_2}-\frac{e^{i\Delta\omega_2 t}}{\Delta\omega_2}\right],\\
h_5 &= -\frac{g^2 e^{-i\omega_c t}}{\Delta\omega_1^2}(e^{i\Delta\omega_1 t}-1)+\frac{ig^2 t e^{-i\omega_c t}}{\Delta\omega_1}, \quad h_6 = -h_5,\\
h_7 &= -\frac{\chi^2 e^{-i\omega_c t}}{\Delta\omega_2^2}(e^{i\Delta\omega_2 t}-1)+\frac{i\chi^2 t e^{-i\omega_c t}}{\Delta\omega_2},\\
h_8 &= \frac{\chi^2 e^{-i\omega_c t}}{\Delta\omega_2^2}(e^{i\Delta\omega_2 t}-1)-\frac{i\chi^2 t e^{-i\omega_c t}}{\Delta\omega_2}.
\end{aligned}
\tag{A3}
$$

$$
\begin{aligned}
l_1 &= \exp(-i\omega_d t), \quad l_2 = \frac{\chi e^{-i\omega_d t}}{\Delta\omega_2}(e^{-i\Delta\omega_2 t}-1),\\
l_3 &= \frac{\chi g e^{-i\omega_d t}}{\Delta\omega_1(\Delta\omega_1-\Delta\omega_2)}[e^{i(\Delta\omega_1-\Delta\omega_2)t}-1]\\
&\quad +\frac{\chi g e^{-i\omega_d t}}{\Delta\omega_2\Delta\omega_1}(e^{-i\Delta\omega_2 t}-1),\\
l_4 &= \frac{\chi g e^{-i\omega_d t}}{\Delta\omega_1(\Delta\omega_1+\Delta\omega_2)}[e^{-i(\Delta\omega_1+\Delta\omega_2)t}-1]\\
&\quad -\frac{\chi g e^{-i\omega_d t}}{\Delta\omega_2\Delta\omega_1}(e^{-i\Delta\omega_2 t}-1),\\
l_5 &= \frac{i\chi^2 t e^{-i\omega_d t}}{\Delta\omega_2}+\frac{\chi^2 e^{-i\omega_d t}}{\Delta\omega_2^2}(e^{-i\Delta\omega_2 t}-1), \quad l_6 = l_5.
\end{aligned}
\tag{A4}
$$

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