Intrinsically Irreversible Multiphoton Laser Gain Mechanisms

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Single-photon stimulated radiative interactions are universally symmetric relative to emission and absorption. We point out that certain multiphoton stimulated processes, whose completion is dependent on the spontaneous emission of at least one photon, do not behave analogously. The broken symmetry found in such cases invalidates simple correlations between population inversion and laser gain. [S0031-9007(97)02451-4]

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Essential to the creation of laser gain is a discrimination which favors stimulated photon production over photon loss by absorption. This is typically achieved by creating a population inversion. However, irreversibility offers a natural but unrecognized mechanism for producing laser gain. We identify here a fundamental family of multiphoton laser gain processes, uniquely characterized by their composite stimulated and spontaneous nature, that provide gain independent of relative atomic state populations. This mechanism may be operating at a fundamental level in many instances of lasing without inversion.

In Figs. 1(a)–1(c), respectively, we depict exemplary one-, two-, and three-photon processes that may lead to changes in atomic and field states. We refer to the field modes involved as G, C, and S, where the C mode always represents an applied driving field. We are concerned with the conditions necessary for stimulated emission to dominate G-mode interactions.

Each process in Fig. 1 may be categorized [1] into elemental subprocesses according to direction and mechanism (spontaneous or stimulated) of population flow into the G mode. In Fig. 1(a), the single-photon process is divided into the three subprocesses of [1(a)-i] spontaneous emission, [1(a)-ii] stimulated emission, and [1(a)-iii] stimulated absorption. The stimulated subprocess [1(a)-ii] produces G-mode laser gain which in this instance is at the atomic transition frequency. The respective subprocesses of Fig. 1(a) have transition rates proportional to $A_1 N_e$, $B_1 I_G N_e$, and $B_1 I_G N_g$, where A_1 and B_1 are the Einstein coefficients, I_G is the G-mode intensity, and N_g (N_e) is the ground (excited)-state population. Importantly, the coefficient, B_1, is the same for both the emissive and absorptive stimulated subprocesses. It follows that stimulated emission [1(a)-ii] dominates over absorption [1(a)-iii] only if $N_e > N_g$.

In Fig. 1(b), we depict a two-photon Raman process enabled through application of driving field C. Three subprocesses are identified: [1(b)-i] a subprocess wherein a C photon is absorbed and a G photon is spontaneously emitted, [1(b)-ii] a similar subprocess wherein G photons are created by stimulated emission, and [1(b)-iii] a subprocess wherein a G photon is absorbed and a C photon is emitted. As in the one-photon case, the three Raman subprocesses can be described by rate coefficients $A_R(I_C, \Delta)$ and $B_R(I_C, \Delta)$, where I_C is the C-field intensity and Δ is an appropriate intermediate-state detuning. It is again true that a single coefficient, B_R, characterizes both the emissive [1(b)-ii] and absorptive [1(b)-iii] stimulated processes. Consequently, the G-mode field can only experience a net positive Raman gain when a Raman-type population inversion, i.e., $N_g > N_e$, exists.

Positive gain, derived from the one- and two-photon processes reviewed above, is coupled through the fundamental symmetry of stimulated emissive and absorptive

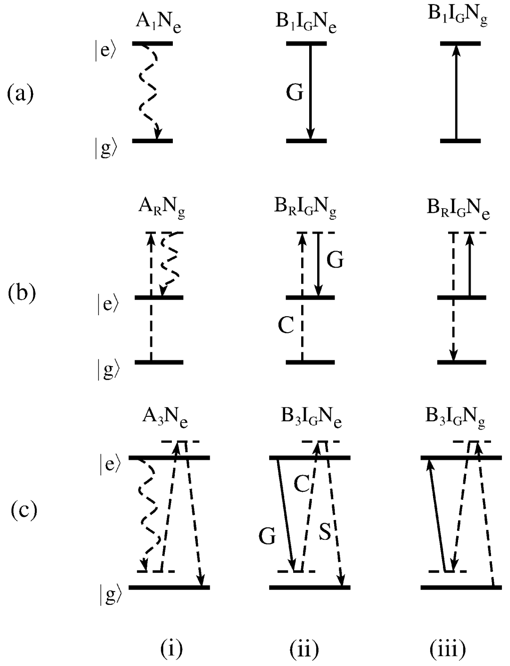

FIG. 1. Reversible (a) one-photon, (b) two-photon, and (c) three-photon transition processes. Categories (i–iii) represent spontaneous G-mode emission, stimulated G-mode emission, and stimulated G-mode absorption, respectively. Solid arrows (G field) represent an applied field experiencing gain or loss. Dashed arrows (C and S fields) are applied driving fields. Wavy arrows represent spontaneous radiative emission.

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subprocesses to normal ($N_e > N_g$) or Raman-type ($N_g > N_e$) population inversion. We now turn our attention to higher order multiphoton processes and find that the preceding results do not generalize.

One exemplary process, involving two driving fields C and S, is shown in Fig. 1(c). A cursory analysis of this process, assuming classical C and S fields, leads as above to a coupling of G-mode gain and population inversion, i.e., since the same rate coefficient B_3 describes both the G-mode emissive [1(c)-ii] and absorptive [1(c)-iii] stimulated subprocesses, the condition $N_e > N_g$ must be satisfied for the G mode to experience net positive gain. As we point out below, however, accounting for the quantum nature of the S-mode driving field, particularly its vacuum state, leads to a different conclusion regarding the coupling of G-mode gain and population inversion.

For simplicity, we assume that the three-photon transition of Fig. 1(c) is driven by atomic coupling to three distinct field modes corresponding, respectively, to transition segments G, C, and S. We assume that each mode is initially occupied by n_G, n_C, and n_S photons, respectively. We assume that the S mode is damped, at a rate fast compared to relevant atomic transition rates, as in the bad cavity limit of cavity QED [2]. It is also assumed that the atom has only two active levels, that the three-photon process is overall resonant, and that intermediate detunings are large compared to the widths of the atomic states.

In terms of this model, the beginning and final states for the stimulated emissive subprocess [1(c)-ii] are $|b\rangle_e = |n_S, n_C, n_G, e\rangle$ and $|f\rangle_e = |n_S + 1, n_C - 1, n_G + 1, g\rangle$, respectively. For the absorptive subprocess [1(c)-iii], the corresponding states are $|b\rangle_a = | n_S, n_C, n_G, g\rangle$ and $|f\rangle_a = | n_S - 1, n_C + 1, n_G - 1, e\rangle$, respectively. The population of the G-field mode is increased (decreased) in the emissive (absorptive) subprocess.

At the lowest order of perturbation theory [3], the relative rates of the G-mode stimulated emissive [1(c)-ii] and absorptive [1(c)-iii] subprocesses, respectively, obey the following relations:

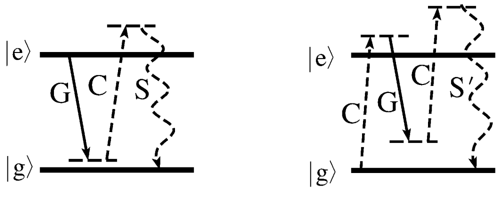
$$\gamma_e \propto N_e \left(\frac{2\pi |\mu_{ge}|^6}{\hbar^6 \Delta_G^2 \Delta_S^2} \right) (n_S + 1) n_C n_G \,, \tag{1}$$

$$\gamma_a \propto N_g \left(\frac{2\pi |\mu_{ge}|^6}{\hbar^6 \Delta_G^2 \Delta_S^2} \right) n_S (n_C + 1) n_G \,, \tag{2}$$

where $\Delta_G \equiv \omega_{\mathrm{eg}} - \omega_G$ and $\Delta_S \equiv \omega_{\mathrm{eg}} - \omega_S = \omega_G - \omega_C$, ω_{eg} is the atomic transition frequency, ω_G, ω_C, and ω_S are the respective field frequencies, and μ_{ge} is the relevant atomic transition matrix element. The presence of the n_G factor in the expression for γ_e is characteristic of a laser (i.e., phase-insensitive) gain process. Photons initially in the G mode stimulate the emission of additional photons into that mode.

From relations (1) and (2), it is apparent that setting $n_S = 0$ results in $\gamma_a = 0$ but $\gamma_e \neq 0$. In other words, when one allows the S segment of the three-photon process to proceed only via coupling to the electromagnetic vacuum (i.e., spontaneously), the G-mode stimulated absorptive subprocess is nonexistent. This can be understood in terms of the intrinsic irreversibility of spontaneous processes. Thus when $n_S = 0$, the stimulated emission subprocess [1(c)-ii] is intrinsically free of competition from its twin absorptive subprocess [1(c)-iii]. It follows that G-mode gain produced by the particular three-photon process shown in Fig. 1(c) is proportional, when the S mode is in its vacuum state, to N_e and not $N_e - N_g$. The damped S mode is dissipative and photons emitted into it during G-mode stimulated emission events propagate away from the physical region of interest. S-mode photons created during stimulated three-photon G-mode emission [1(c)-ii] thus does not necessarily "turn-on" the absorptive process [1(c)-iii]. These comments also apply when the S segment of the three-photon transition is driven by coupling to the ambient electromagnetic vacuum rather than a specific damped mode as may be the case in typical physical realizations of the system.

We conclude that the essential symmetry between stimulated emission and absorption subprocesses, originally introduced [1] in the description of single-photon transitions, is fundamentally absent in the case of multiphoton transitions wherein spontaneous emission is integrally involved [see Fig. 2(a)]. The absence of an inviolable symmetry between emission and absorption for each and every multiphoton process eliminates the often assumed basis for identifying positive population inversion (normal or Raman) with positive gain. However, breaking the path by path symmetry between stimulated emission and absorption does not imply that one should routinely see positive gain in inversionless driven systems. To determine the total gain exhibited by a given system, one must sum over emissive and absorptive multiphoton processes of all kinds and orders. Generally, a system will support a multitude of irreversible multiphoton processes, some of which are emissive (Fig. 2) and some absorptive (Fig. 3). In some situations, the sum over distinct multiphoton pathways may lead to inversionless gain, in other cases not. Spontaneous-emission-activated multiphoton



(a) (b)

FIG. 2. Intrinsically irreversible multiphoton laser gain mechanisms for producing stimulated emission into mode G. Spontaneous emission into dissipative mode S is irreversible. (a) Raman-type and (b) Rayleigh-type processes that contribute to stimulated emission into a specific G-field mode.

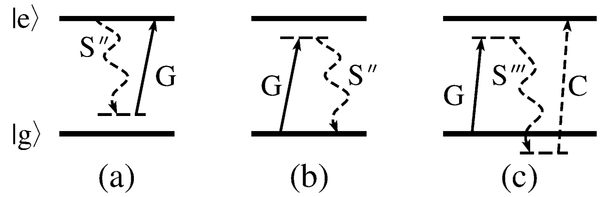


FIG. 3. Competing, irreversible but nonequivalent, absorption processes for G mode photons.

gain provides an alternative motivation for the phenomenon of lasing without inversion, one quite distinct from the quantum interference picture often cited [4].

The irreversible multiphoton G-mode gain process of Fig. 2(b) warrants comment. In this instance, the process begins and ends in the same atomic state and therefore belongs to the set of processes normally associated with parametric (i.e., phase-sensitive) gain. In the present context, however, the presence of the spontaneous emission step, S', enables the process to provide true (phase-insensitive) gain into the G mode. Note that in process 2(b), G-mode energy may accrue only at the expense of the other field modes, while in the case of process 2(a), stored material energy may also be extracted. The process of Fig. 2(b) is related to that operative in the singly resonant parametric oscillator [5].

A characteristic physical observable associated with irreversible multiphoton gain processes is the appearance of fluorescence photons of specific frequency. We have performed an experiment involving driven two-level atoms that demonstrates fluorescence characteristic of the processes shown in Fig. 2.

Barium provides a simple two-level-like atomic system whose predominant isotope ^{138}Ba (72%) is free of nuclear spin and hence complicating hyperfine structure. Barium atoms in a beam and passing through the center of a confocal fluorescence analyzer cavity [6] are driven by a C field resonant with the 554 nm $(6s^2)\ ^1S_0$- $(6s6p)\ ^1P_1$ transition. A second field (the G field) can be applied with a detuning of $\Delta_G \equiv \omega_{\text{eg}} - \omega_G = 160$ MHz. The driving fields are collinear and orthogonal to the barium atomic beam and the analyzer cavity. The cavity and atomic beam are also orthogonal. The analyzer cavity is piezoelectrically scannable with a free spectral range of 1.5 GHz and has a finesse of 500. The S (S') segment of the transitions shown in Fig. 2 is driven via coupling to the entire free-space vacuum reservoir. The analyzer cavity employed perturbs this reservoir insignificantly.

In general, bichromatically driven atomic spectra [7] contain various spectral peaks, some of whose frequencies are dependent on the strengths (Rabi frequencies) of the driving fields. The frequency of the spontaneously emitted S field is, however, expected to be entirely determined by the bare atomic, C- and G-field frequencies. In our experiment, with $\omega_C = \omega_{\text{eg}}$, the fluorescence frequency characteristic of either process of Fig. 2 is $\omega_{\text{eg}} + \Delta_G$.

Figure 4(a) shows the optical power emitted out the end of the analyzer cavity as a function of cavity resonance frequency when the atoms are driven by the C field alone. The emitted spectrum has the standard Mollow format with a central peak at the atomic transition frequency and two sidebands symmetrically displaced by the C-field Rabi frequency of 190 MHz. Figure 4(b) shows an analogous spectrum obtained while applying both C and G fields. The applied G field has a Rabi frequency of 60 MHz. In the presence of the G field, two new spectral features appear. One, indicated by the "G" in Fig. 4(b), corresponds to G-field elastic scattering while

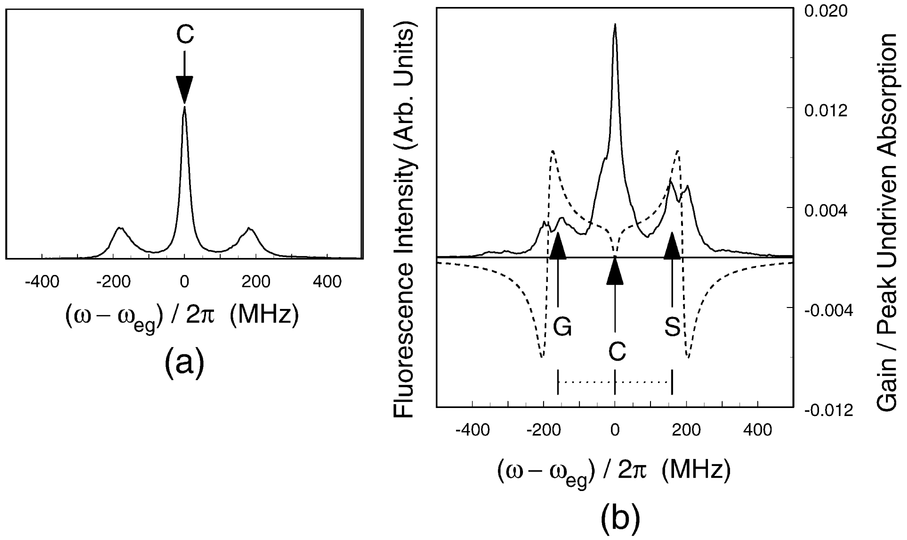
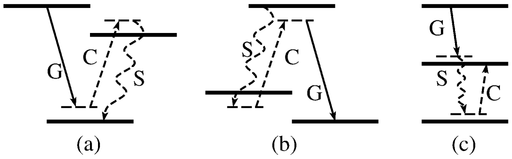


FIG. 4. Observed atomic fluorescence intensity versus frequency. (a) Monochromatic resonant excitation. The central Mollow peak and two sidebands are observed. (b) Bichromatic excitation with one resonant component C and a weaker detuned component G. The spontaneously emitted S peak has the properties expected for S-mode emission as in Fig. 2. Overlayed dotted line indicates the calculated normalized gain profile (right vertical axis).



(a) (b) (c)

FIG. 5. Representative irreversible multiphoton gain processes in three-level, inversionless lasing systems of various structure: (a) V, (b) Λ, and (c) cascade systems.

the second, indicated by the "S," occurs at the frequency $\omega_{\mathrm{eg}} + \Delta_G$ and is thus consistent with S-mode emission of either process shown in Fig. 2. The S peak is observed to tune with ω_G as expected. We note that the observed linewidth of the emitted fluorescence is consistent with the instrumental resolution, the processes of Fig. 2, and laser and atomic linewidths.

Earlier theoretical and experimental work concerned with resonant ($\Delta_C \equiv \omega_{\mathrm{eg}} - \omega_C = 0$) excitation of two-level atoms [8–10] has shown that a probe field experiences inversionless gain for most detunings within the Mollow sidebands. The overlayed dashed line in Fig. 4(b) shows the gain profile calculated [9] according to our experimental parameters. It is clear that the G field employed here is tuned to a spectral region exhibiting gain. Observation of the S fluorescence peak suggests that intrinsically irreversible multiphoton processes play an important role in generation of inversionless laser gain in the driven two-level-atom system. Future experiments may demonstrate explicit correlation between intrinsically irreversible multiphoton gain processes using cavity QED techniques [2] in as much as a "bad" cavity of finite bandwidth tuned across the S-segment frequency may through modification of spontaneous emission rates modify system gain.

We have shown that irreversible multiphoton gain processes like those shown in Figs. 2 (and by generalization the three-level analogs shown in Fig. 5) do not have equivalent absorptive processes. This does not mean that competing processes are entirely absent. Several processes that compete with the G-field gain derived from the processes of Fig. 2 are shown in Fig. 3. While there is still competition between emissive and absorptive processes, the competing processes are not generally equivalent, and it is not surprising that in some instances gain mechanisms can outweigh absorptive mechanisms in the absence of population inversion. It is no longer necessary to view inversionless laser gain as arising from the destructive interference of absorptive processes. Irreversible (spontaneous-emission-activated) multiphoton transitions are intrinsically imbalanced relative to emission and absorption and a single elemental process may provide gain that is independent of the relative populations of the initial and final states. Relative atomic populations enter only through competition with nonequivalent absorptive processes. Thus, in systems supportive of intrinsically irreversible multiphoton stimulated emission, the conditions governing the appearance of positive gain will differ from the simple inversion condition traditionally invoked. The historic gain condition is implicitly dependent on the assumption of equivalent emissive and absorptive pathways that can now be seen to lack generality.

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[1] A. Einstein, Phys. Z. **18**, 121 (1917) [English translation by D. ter Haar, in *The Old Quantum Theory* (Pergamon, Oxford, 1967)].

[2] *Cavity Quantum Electrodynamics,* edited by P. R. Berman (Academic Press, Boston, 1994); P. R. Rice and H. J. Carmichael, IEEE J. Quantum Electron. **24**, 1351 (1988).

[3] R. Loudon, *The Quantum Theory of Light* (Clarendon Press, Oxford, 1983), 2nd ed., p. 196.

[4] G. Grynberg and C. Cohen-Tannoudji, Opt. Commun. **96**, 150 (1993); A. S. Zibrov, M. D. Lukin, D. E. Nikonov, L. Hollberg, M. O. Scully, V. L. Velichansky, and H. G. Robinson, Phys. Rev. Lett. **75**, 1499 (1995); G. Grynberg, M. Pinard, and P. Mandel, Phys. Rev. A **54**, 776 (1996). For reviews see O. Kocharovskaya, Phys. Rep. **219**, 175 (1992); M. O. Scully, *ibid.* **219**, 191 (1992); P. Mandel, Contemp. Phys. **34**, 235 (1993).

[5] S. E. Harris, Proc. IEEE **57**, 2096 (1969); S. T. Yang, R. C. Eckardt, and R. L. Byer, J. Opt. Soc. Am. B **10**, 1684 (1993).

[6] J. D. Cresser, J. Häger, G. Leuchs, M. Rateike, and H. Walther, in *Dissipative Systems in Quantum Optics: Resonance Fluorescence, Optical Bistability, Superfluorescence,* edited by R. Bonifacio *et al.* (Springer-Verlag, Berlin, 1982).

[7] M. G. Rozman, Proc. Acad. Sci. Estonian SSR **33**, 206 (1984); Y. Zhu, Q. Wu, A. Lezama, D. J. Gauthier, and T. W. Mossberg, Phys. Rev. A **41**, R6574 (1990); Z. Ficek and H. S. Freedhoff, Phys. Rev. A **53**, 4275 (1996), and references therein.

[8] S. Haroche and F. Hartmann, Phys. Rev. A **6**, 1280 (1972); B. R. Mollow, Phys. Rev. A **5**, 2217 (1972); S. L. McCall, Phys. Rev. A **9**, 1515 (1974).

[9] F. Y. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow, Phys. Rev. Lett. **38**, 1077 (1977); A. M. Bonch-Bruevich, S. G. Przhibel'skii, and N. A. Chigir, Vestn. Mosk. Univ. Fiz. **33**, 35 (1978).

[10] D. Grandclément, G. Grynberg, and M. Pinard, Phys. Rev. Lett. **59**, 40 (1987); G. Khitrova, J. F. Valley, and H. M. Gibbs, Phys. Rev. Lett. **60**, 1126 (1988); A. Lezama, Y. Zhu, M. Kanskar, and T. W. Mossberg, Phys. Rev. A **41**, 1576 (1990); D. J. Gauthier, Q. Wu, S. E. Morin, and T. W. Mossberg, Phys. Rev. Lett. **68**, 464 (1992).