Forward Gain Suppression of Optically Pumped Stimulated Emissions Due to Self-Induced Wave-Mixing Interference during a Pump Pulse

W. R. Garrett

Chemical Physics Section, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

(Received 22 March 1993)

Theoretical and experimental presentations are given of a drastic suppression, during the interval of the excitation pulse, of forward-directed optically pumped stimulated emission from a two-photon resonantly excited state to a lower state which is also coupled to the ground state. The effect is caused by a self-induced interference from the difference-frequency mixing field that is generated during the time that the excitation fields are present. Several other implications of this effect are given and some puzzling observations in other studies are noted and clarified.

PACS numbers: 32.70.Fw, 32.80.Wr, 42.50.Md

Though detailed considerations of spontaneous and induced optical transitions in atoms and molecules constitute the oldest and perhaps historically the most cited subjects in quantum physics, there are nevertheless new features of such problems being discovered even now. Much attention has been given recently to strong effects on such transitions that can result from placing excited atoms between mirrors, in optical cavities, or in dielectric media [1]. At higher number densities effects described much earlier including super-radiance and superfluorescence drastically change radiation rates. In the present study a new and very dramatic feature of a very old subject, optically pumped stimulated emission (OPSE), is revealed. Under well defined circumstances, a destructive interference involving the OPSE, also called amplified spontaneous emission (ASE), photons can suppress gain for forward emission and shift line profiles for backward emission. The interference is self-induced in the sense that the incoherent OPSE is itself one component in the interfering loop. Experimental verifications of some of the OPSE effects predicted here are very readily achieved.

The present study fits into the context of well known features of several strongly resonant multiphoton-driven excitation processes where results can become drastically altered by destructive interference produced by wave-mixing fields that are generated along with direct excitation [2], when propagation effects must be included in the problem.

If resonant excitation of an optically allowed transition of frequency ω_r is driven by an odd-order process, i.e., $(2n+1)$-photon pumping, then the driving fields also generate a $(2n+2)$-wave-mixing field ω_m at the sum or difference frequency which corresponds to odd-photon resonance, (i.e., $\omega_m \simeq \omega_r$). A destructive interference can be established between pumping of the resonant transition by the wave-mixing field at ω_m and by direct odd-photon-excitation [3,4]. Consequentially excitation probabilities can become severely suppressed or strongly shifted [5,6]. Similarly, stimulated hyper-Raman (SHR) scattering can experience gain suppression for forward SHR emission [4,7] and pressure-dependent frequency shifts for emissions in other directions [8]. These interference effects are independent of the intensity of the driving fields, and manifest themselves in a number of ways [2–4].

For two-photon excitations a second type of destructive interference can be established when a second pathway from the two-photon state back to the initial state is created by phase-matched parametric four-wave mixing (PFWM) fields (as opposed to strongly absorbed, badly phase-matched wave-mixing fields in the odd-photon excitation case). Thereby coherent and incoherent processes that are driven by two-photon pumping can become limited (independent of their directions) at elevated laser intensities [4,9]. There are no line shifts associated with suppression of resonant excitations in this two-photon-resonant category, and unlike the odd-photon case, the suppression *is not* independent of pump intensity.

In the present study we show theoretically and confirm experimentally the unexpected and counterintuitive result that a gain-suppressing interference can be produced even when one or more of the photons directly involved in the interfering loop is generated by optically pumped stimulated emission. Specifically, when resonant laser excitation of a state leads to an inversion and subsequent OPSE where the stimulated transition terminates on a state which is optically coupled back to the ground state, then the combined resonant laser and ASE fields produce a polarization and corresponding wave-mixing field with a destructive odd-photon interference which *reduces the gain for forward OPSE during the interval of the laser pulse to a tiny fraction of its expected value.* Backward gain is unaffected, though the ASE is predicted to show a pressure-dependent and intensity-independent shift for emission occurring during the pump pulse. Note immediately that this prediction is fundamentally different from suppression of ASE (in all directions) that results from PFWM interference which limits the two-photon pumping of the population from which ASE is produced [4,9].

By closely following previously established results by Payne and Garrett [3], it is relatively easy to sketch a

0031-9007/93/70(26)/4059(4)$06.00

proof of the claimed result. Thus a three-state model describes resonant two-photon excitation from $|0\rangle$ to $|2\rangle$ by laser photons ω_L accompanied by stimulated emission, ω_{SE}, from $|2\rangle$ to $|3\rangle$. (See inset of Fig. 1). The time-dependent wave function for an atom at z,t with energy states $\hbar\omega_i$ is written as

$$|\Psi(z,t)\rangle = a_0(z,t)e^{-i\omega_0 t}|0\rangle + a_2(z,t)e^{-i(\omega_2 t-2k_L z)}|2\rangle + a_3(z,t)e^{-i(\omega_3 t-2k_L z)}|3\rangle . \quad (1)$$

An atom at z,t is subjected to a plane-polarized plane-wave laser field $E_L = E_{L0}(t-z/v_L)\cos(\omega_L t - k_L z)$. In a semiclassical picture, wherein the initial start-up of ASE is ignored, a weak field E_{SE} is also present due to stimulated emission from $|2\rangle$ to lower lying vacant state $|3\rangle$. From the population created along the laser beam this field has gain for a forward-directed component E_{SE}^+ ($+z$ direction, parallel to the laser field), and backward (antiparallel) component E_{SE}^-. These are also treated as plane waves. Thus $E_{SE} = E_{SE}^+ + E_{SE}^-$ and $E_{SE}^{\pm} = \frac{1}{2}E_{SE0}^{\pm} \times \exp[i(\omega_{SE}t \mp k_{SE}z)] + \text{c.c.}$ (It is the gains for these fields which are of primary interest here.) Couplings between the three relevant states are written in terms of reduced (j)-photon Rabi frequencies, $\Omega_{n,m}^{(j)}$. For one-photon coupling $\Omega_{n,m}^{(1)}(z,t) = D_{n,m}E_0/2\hbar$, where $D_{n,m}$ is a matrix element of the electric dipole operator $\hat{D}$ between states n and m and a plane-wave field of frequency ω is written as $E(z,t) = E_0\cos(\omega t - k_\omega z)$. Thus states $|2\rangle$ and $|3\rangle$ are coupled by $\Omega_{2,3}^{(1)+} = D_{2,3}E_{SE0}^+/2\hbar$ and by $\Omega_{2,3}^{(1)-}$ similarly defined. States $|0\rangle$ and $|2\rangle$ are coupled by a two-photon Rabi rate $2\Omega_{02}^{(2)}$ due to the laser field.

Now consider a critical step that is always omitted in treating the stimulated emission problem. In spite of the fact that stimulated emission is incoherent with respect to the pump field, nevertheless, during the interval of time when E_L and E_{SE} are both present, the combination of the laser field and the field E_{SE} creates a nonlinear polarization P_{ω_m} at frequency $\omega_m = 2\omega_L - \omega_{SE}$, which is a source for generation of a four-wave mixing field E_m at ω_m. (This statement applies only in cases where $|3\rangle$ is coupled to the ground state.) The resonant contribution to the polarization contains a nonlinear component, $P_{\omega_m}^+$, driven by the laser field combined with forward-stimulated emission and a component, $P_{\omega_m}^-$, associated with the laser field plus backward-stimulated emission. In order to separate the component $P_{\omega_m}^+$ from $P_{\omega_m}^-$, the part of a_3 which is phased with $+z$ propagation is separated from that associated with $-z$ propagation by writing $a_3(z,t) = a_3^+(z,t)e^{-ik_{SE}z} + a_3^-(z,t)e^{ik_{SE}z}$ in Eq. (1) and in the definition of the polarization. In terms of the matrix elements and amplitudes, the components of the polarization at ω_m are

$$P_{\omega_m}^+ = ND_{0,3}a_0^*(z,t)a_3^+(z,t)e^{-ik_{SE}z} \times e^{2ik_L z}e^{-i(2\omega_L-\omega_{SE})t} + \text{c.c.} \equiv ND_{0,3}Y^+(z,t) + \text{c.c.}, \quad (2a)$$

$$P_{\omega_m}^- = ND_{0,3}a_0^*(z,t)a_3^-(z,t)e^{+ik_{SE}z} \times e^{2ik_L z}e^{-i(2\omega_L-\omega_{SE})t} + \text{c.c.} \equiv ND_{0,3}Y^-(z,t) + \text{c.c.}, \quad (2b)$$

where for convenience the number density N and dipole matrix element $D_{0,3}$ are separated out to define a reduced polarization $Y^{\pm}(z,t)$. There is no question that a nonlinear polarization of the medium is created by the combined laser and ASE fields. It is a matter of determining its consequences. As it turns out the consequences are dramatic.

With the laser, ASE, and FWM fields included in the problem, equations of motion for the amplitudes are obtained in a truncation appropriate to low excitation probabilities, i.e., $a_0 \simeq 1$ and $|a_3| \ll 1$. With $a_0 = 1$ and the usual approximations [3], equations for a_2 and a_3 take the form

$$\partial a_2/\partial t = -(\gamma_2/2)a_2 + i\Omega_{2,0}^{(2)}a_0 + i\Omega_{2,3}^{(1)+}a_3^+ + i\Omega_{2,3}^{(1)-}a_3^- + i\Omega_{2,3}^{(1)+}e^{i2k_{SE}z}a_3^- + i\Omega_{2,3}^{(1)+}e^{-i2k_{SE}z}a_3^+ , \quad (3a)$$

$$\partial(a_3^+e^{-ik_{SE}z} + a_3^-e^{ik_{SE}z})/\partial t = -\tfrac{1}{2}\gamma_3(a_3^+e^{-ik_{SE}z} + a_3^-e^{ik_{SE}z}) + i[\Omega_{3,0}^{(1)+} + \Omega_{3,0}^{(1)-}]e^{-2ik_L z}a_0 + i[\Omega_{3,2}^{(1)+}e^{-ik_{SE}z} + \Omega_{3,2}^{(1)-}e^{ik_{SE}z}]a_2 . \quad (3b)$$

Here γ_2 and γ_3 are the total spontaneous decay rates for states $|2\rangle$ and $|3\rangle$, which are included to simulate the natural widths of the lines. Finally, the quantity $\Omega_{3,0}^{(1)\pm}$ is the reduced one-photon Rabi frequency associated with the difference-frequency mixing field $E_m^{\pm}$ at ω_m, i.e., $\Omega_{3,0}^{(1)\pm} = D_{3,0}E_{M0}^{\mp}/2\hbar$. This field, which must be determined as part of the problem, resonantly couples $|3\rangle$ with $|0\rangle$ (shown dotted in Fig. 1, indicating that it is strongly absorbed). The gain for the fields $E_{SE}^{\pm}$ (Rabi frequencies $\Omega_{3,2}^{(1)\pm}$) must also be determined.

It is not necessary to completely solve the set (3a),(3b) in order to extract the result of interest here. If we consider a simple square pulse, Eq. (3a) can be solved in the approximation $|a_3| \ll 1$ (certainly valid at early time) to give $a_2 \simeq i(2/\gamma_2)\Omega_{20}^{(2)}(1-e^{-\gamma_2 t/2})$. This result can be substituted into Eq. (3b), and the resulting equation can be separated into one equation for a_3^+ and another for a_3^-. (In this procedure a negligible coupling between forward and backward components is omitted.) The equation for a_3^+ becomes

$$\partial a_3^+/\partial t = -(\gamma_3/2)a_3^+ + i\Omega_{3,0}^{(1)+}e^{-i(2k_L-k_{SE})z}a_0 - (2/\gamma_2)\Omega_{3,2}^{(1)+}\Omega_{20}^{(2)}(1-e^{-\gamma_2 t/2}) . \quad (4)$$

Equation (4) can be expressed as an equation for the polarization $P_{\omega_m}^+$ or equivalently for Y^+. The same can be done for a_3^-. Note that $\Omega_{3,0}^{(1)+}$ and $\Omega_{3,2}^{(1)+}$ must yet be obtained as part of the problem. But the fields $E_m^{\pm}(\omega_m)$ satisfy Maxwell's equation with $P_{\omega_m}^{\pm}$ as a source term. Without repeating the steps here we note that a solution of Maxwell's equation for $E_m^{\pm}$ can be obtained in terms of an integral over $\partial P_{\omega_m}^{\pm}/\partial t$ [8]. The expressions for $E_m^{\pm}$, or equivalently $\Omega_{3,0}^{(1)\pm}$, can be substituted back into Eq. (4) to yield separate coupled Bloch-Maxwell equations for the reduced polarizations $Y^{\pm}(z,t)$. The component $Y^+(z,t)$, propagating in the $+z$ direction, satisfies

$$\frac{\partial Y^+(z,t)}{\partial t} \simeq -\kappa_{0,3}\int_0^z dz' Y^+(z',t-(z-z')/v) - \frac{\gamma_3}{2}Y^+(z,t) + i\Omega_{3,0}^{(3)+} . \quad (5)$$

$\Omega_{0,3}^{(3)+} = (i/\gamma_2)2\Omega_{2,0}^{(2)}\Omega_{3,2}^{+}e^{i\mu^+ z}(1-e^{-\gamma_2 t/2})$ is the three-photon Rabi rate for excitation of $|3\rangle$, phased with the forward ASE field, where $\mu^{\pm} = (2k_L \mp k_{\rm SE}) - \omega_m/v)$, $\Omega_{3,2}^{+} = D_{3,2}E_{\rm SE0}^{+}$, and $\kappa_{0,3} = 2\pi N|D_{0,3}|^2\omega_m/\hbar c$. A similar equation can be written for $Y^-(z,t)$.

We now examine the form of Eq. (5) which describes the forward component of the electric polarization at ω_m. It is immediately apparent that this equation is of exactly the same form as that developed and studied extensively by Payne and Garrett [3,4,6] in describing odd-photon interference phenomena. This equation differs from the earlier result only in the functional form of $\Omega_{3,0}^{(3)}$. But an interesting feature of the solution to Eq. (5) is that the result $Y^+(z,t) \simeq 0$ obtains, independent of the form of the pumping term $\Omega_{3,0}^{(3)+}$ [4,6]. Thus by reducing this part of the problem to the above form *we can say immediately that the polarization $P_{\omega_m}^+(z,t)$ vanishes and thus the components $a_3^+(z,t)$, driven by forward beams, vanish shortly beyond the entrance into the medium* [3]. It follows from Eq. (5) that pumping of $|3\rangle$ by the combination of laser-driven forward-stimulated emission and the generated FWM field yields $Y^+ \sim 0$. Thus no stimulated excitation of $|3\rangle$ is produced by the copropagating laser and ASE fields. This is explicitly manifest in another way. The gain for the stimulated emission can be written in terms of a source term, the SE polarization, $P_{\rm SE}^+(\omega_{\rm SE}) = ND_{2,3}a_2^* a_3^+ e^{i\omega_{\rm SE}t} + \text{c.c.}$ Since this forward polarization is proportional to a_3^+, which remains vanishingly small, then $P_{\rm SE}^+$ also remains small. Thus the forward ASE gain is effectively reduced to zero after building up to only a tiny fraction of its expected value. Finally, because of the great difference in phase considerations, the interfering FWM field associated with the backward ASE will only produce a shift [5,6] in $\omega_{\rm SE}$ *during the laser pulse*; otherwise the ASE emission in the backward direction should be normal.

One can now predict several new features of optically pumped stimulated emissions in the circumstances posited. To test some of these, experiments were performed in Na vapor in a laser–heat-pipe–spectrometer arrangement identical to that described in [4]. In a manner similar to [4], backward- and forward-stimulated emissions were collected with equal collection efficiencies and injected into an optical spectrometer under exact two-photon resonant excitation of $3S \rightarrow 4D$ and $3S \rightarrow 3D$ transitions over pressure ranges from 0.05 to 4.0 Torr of Na.

In Fig. 1 traces of $4D_{3/2,5/2}$ to $3P_{1/2}$ and $3P_{3/2}$ OPSE profiles at 568.4 and 569.0 nm are shown for forward (upper trace) and backward (lower trace) emissions. These data result from unfocused $\simeq 4$ ns pulses of 5 mJ/pulse with 2 mm beam diameter and $\simeq 0.1$ cm^{-1} bandwidth. In this "high" pressure case the forward component is not visible above the noise (2× gain), but strong backward emissions are readily observed. The start-up time for OPSE under the present experimental conditions is much shorter than the laser pulse length; thus almost all of the observed emissions "occur during the laser pulse." Under this condition the forward component is strongly suppressed by the interference mechanism under discussion. In Fig. 2 similar traces are shown for the 818.5 and 819.7 nm emissions from resonant two-photon pumping of the unresolved $3D_{3/2,5/2}$ state. Here weak ASE and/or parametric four-wave mixing emissions to the $3P_{3/2}$ and $3P_{1/2}$ levels are visible along with an axial parametric four-wave mixing component near 819.2 nm [9]. (Only a small fraction, if any, of the forward

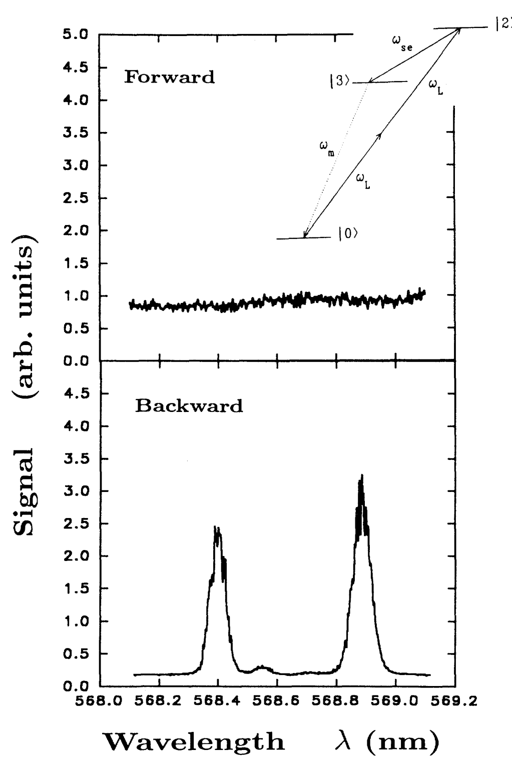

FIG. 1. Optically pumped stimulated emission (OPSE) profiles from resonant two-photon excitation of Na $4D_{3/2,5/2}$ as depicted in the inset. Upper trace, forward $4D \rightarrow 3P_{1/2,3/2}$ components; bottom trace, backward components. $P_{\rm Na} = 1.8$ Torr. Pump energy, 5 mJ/pulse.

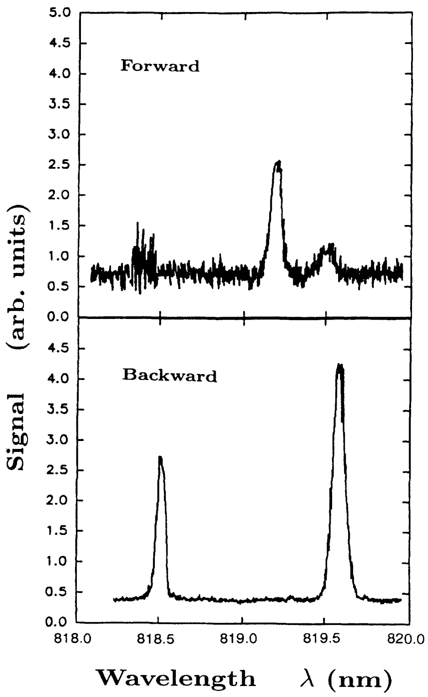

FIG. 2. OPSE ($3D_{3/2,5/2} \rightarrow 3P_{1/2,3/2}$) from two-photon pumping of the $3D$ state in Na at 3.6 Torr of Na. Upper trace, forward emission, where largest peak is axially phase-matched parametric four-wave mixing. Lower trace, backward emissions. $P_{\mathrm{Na}}=3.6$ Torr. Pump energy, 1.8 mJ.

conical emissions produced by angle-phase-matched parametric four-wave mixing was injected into the spectrometer.) Very strong backward emissions at the OPSE wavelengths are again evident in the lower trace. These data were taken at a laser energy of 1.5 mJ/pulse. At much lower number densities where population dumping during the laser field is less complete, more fractional forward versus backward emission is produced at much lower number densities, though always suppressed to a considerable degree. Note the easy prediction that any forward emission should be time delayed relative to the pump beam. However, forward emissions were suppressed to such an extent that, though visible with a photomultiplier, their time profiles could not be traced with an available but relatively insensitive fast photodiode. Note also that the predicted linearly pressure-dependent blueshift in the backward emissions that occurs during the laser pulse is not discernible (<0.05 cm^{-1}) at the low pressures achievable in the heat pipe.

Many other data similar to Figs. 1 and 2 have been recorded in separate experiments over a range of pressures and laser powers, including a manifestation of the interference in a different mode involving one-photon pumping of a high lying state and an unequivocal example involving a five-photon interference. In these accompanying studies ten different pump-transition combinations have been observed to show the predicted effects, all completely consistent with the expected interference-based behavior. Indeed, though surprising and counterintuitive, the suppression effects are so prevalent that independent experimental verification of this phenomenon requires only the crudest of measurements.

Additionally, note that the present result explains very abnormal branching ratios that were observed by Miller [10] for forward ASE in Xe in cases where the final state could and could not couple back to the ground state through FWM, and abnormal forward-backward ASE profiles observed in the course of stimulated hyper-Raman studies [4,7].

Finally note that in addition to changing the perspective on an old and well studied subject, the effects described here have important implications for other problems including a subset of two-photon laser schemes, for certain remote sensing applications, and for some device applications.

This research was sponsored by U.S. DOE Office of Health and Environmental Research, under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

[1] S. Haroche and D. Kleppner, Phys. Today **42**, (1), 24 (1989); S. E. Morin, Q. Wu, and T. W. Mossberg, Opt. Photonics News **3**, 8 (1992), and references therein.

[2] E. A. Manykin and A. M. Afanas'ev, Zh. Eksp. Teor. Fiz. **48**, 931 (1965); **52**, 1246 (1967) [Sov. Phys. JETP **21**, 619 (1965); **25**, 828 (1967)]; J. C. Miller, R. N. Compton, M. G. Payne, and W. R. Garrett, Phys. Rev. Lett. **45**, 114 (1980); M. G. Payne, W. R. Garrett, and H. C. Baker, Chem. Phys. Lett. **75**, 468 (1980); D. J. Jackson and J. J. Wynne, Phys. Rev. Lett. **49**, 543 (1982).

[3] M. G. Payne and W. R. Garrett, Phys. Rev. A **26**, 356 (1982); M. G. Payne, W. R. Garrett, and W. R. Ferrell, Phys. Rev. A **34**, 1143 (1986); W. R. Garrett, S. D. Henderson, and M. G. Payne, Phys. Rev. A **34**, 3463 (1986); **35**, 5032 (1987).

[4] W. R. Garrett, M. A. Moore, R. C. Hart, M. G. Payne, and R. Wunderlich, Phys. Rev. A **45**, 6687 (1992).

[5] R. Friedberg, S. R. Hartman, and J. T. Manassah, Phys. Rev. A **39**, 93 (1989); M. G. Payne and W. R. Garrett, Phys. Rev. A **42**, 1434 (1990).

[6] W. R. Garrett, R. C. Hart, J. E. Wray, I. Datskou, and M. G. Payne, Phys. Rev. Lett. **64**, 1717 (1990).

[7] M. A. Moore, W. R. Garrett, and M. G. Payne, Opt. Commun. **68**, 310 (1988).

[8] W. R. Garrett, R. C. Hart, J. C. Miller, M. G. Payne, and J. E. Wray, Opt. Commun. **86**, 205 (1991).

[9] M. S. Malcuit, D. J. Gauthier, and R. W. Boyd, Phys. Rev. Lett. **55**, 1086 (1985).

[10] J. C. Miller, Phys. Rev. A **40**, 6969 (1989).