Anti-Stokes Raman lasers without inversion

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We describe the propagation dynamics and gain line shape of pulsed, tunable, anti-Stokes Raman lasers without inversion. Analytical expressions for the gain are obtained and evaluated for inhomogeneously broadened atomic media. The inversionless gain, which requires selective pumping of one of two lower atomic states, occurs regardless of the relative populations of these states and vanishes when the atomic populations are in equilibrium. [S1050-2947(97)08707-6]

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INTRODUCTION

The prospect of obtaining stimulated amplification without population inversion has fueled 20 years of intense theoretical and experimental study. In the past, lasers without population inversion (LWI) have been associated with the elimination of loss caused by lower laser-state atoms via quantum interference. The primary difference between the various LWI models is the manner in which the interference is obtained, whether by external injection of optical coherence [1], coherent population trapping (CPT) [2–4], or intrinsic atomic relaxation processes [5–7]. (Other references of general interest may be found in [8].)

In this paper, we work in a region far off resonance where there is no loss. Even so, we show that there is gain due to the difference in excitation rates between an upper state and a lower state, regardless of the relative populations of these states. As in other manifestations of LWI, a second, coherent field must be applied to mediate the two-photon process. Gain is possible when the sum or difference frequency of the two laser fields is nearly resonant with a Raman (dipole-forbidden) transition in the medium. Under these conditions, the primary loss mechanisms are two-photon absorption and Raman scattering, and so we discuss the possibility of coherent gain without inversion of the *two-photon* transition manifold. In this region of nominally lossless propagation, gain is made possible by atomic coherence, rather than quantum interference-based negation of lower laser-state atomic absorption. Still, our formalism may also be used to describe the usual class of LWI, in which lower-state absorption cannot be neglected. This gain mechanism, and the results of this paper, can, in principle, be extended to arbitrarily short wavelengths, subject to material constraints.

This paper consists of six sections. The first section derives the semiclassical equations of motion for the individual atoms of an inhomogeneously-broadened ensemble, and gives the propagation equations for the two laser fields. The second section derives an approximate solution for the ensemble atomic coherence, based on a series expansion, for the case when the atoms have a Maxwellian (‘‘thermal’’) velocity distribution and the response of the ensemble is Doppler broadened. From this series we may delineate the contributions of population inversion and population excitation to the ensemble coherence, as a function of the laser intensities and detunings. The terms in this series are closely related to the plasma dispersion function with complex argument. The third section uses the approximate solution to derive analytic formulas for the evolution of pulse energy and pulse area, and the fourth section evaluates the gain coefficient for a model atomic system. The fifth section relates the preparation of a Doppler-broadened medium by the applied laser field(s) to the ensemble coherence and to the form of the inversionless gain lineshape. In the sixth section, we discuss some of the experimental considerations that will influence the operation of this kind of inversionless laser.

Before proceeding, we cite important developments in the history of two-photon amplification. Coherent nonlinear energy extraction via two-photon amplification has been proposed in the past [9], and some of the special behavior of two-photon systems, e.g., pulse reshaping, has already been discussed. The idea is to store energy in an inverted metastable atomic level and then extract it coherently with a trigger pulse; this has the added benefit of effectively up converting or down converting the trigger laser wavelength with a higher saturation intensity than may be achieved in normal laser amplifiers. High-brightness, spontaneous, anti-Stokes Raman sources which do not require inversion have also been proposed [10]. Inversionless gain in the recent experimental observations of continuous-wave, resonance-line LWI in rubidium [11] and sodium [12], is due to a combination of atomic coherence and CPT. In other work, phase-dependent, inversionless coherent gain on picosecond time scales has been demonstrated [13].

I. THE SEMICLASSICAL EQUATIONS OF MOTION

We study a system that has many upper (or intermediate) states $|i\rangle$ which are connected by arbitrary matrix elements μ_{1i} and μ_{2i} to lower states $|1\rangle$ and $|2\rangle$, respectively. We consider the one-dimensional propagation of two linearly polarized quasimonochromatic electromagnetic fields with slowly varying spatial envelopes, a weak probe laser field $\vec{E}_p(z,t)=\mathrm{Re}[E_p(z,t)\hat{x}\exp i(\omega_p t-k_p z)]$, which may be applied or generated from noise, and a coupling laser field $\vec{E}_c(z,t)=\mathrm{Re}[E_c(z,t)\hat{x}\exp i(\omega_c t-k_c z)]$. The sum or difference frequency of these fields is nearly equal to a two-photon atomic transition frequency, as shown in Fig. 1. We are

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especially interested in the rate of change of the probe laser pulse energy, as this pulse propagates through a pumped atomic medium. The states $|i\rangle$ are adiabatically eliminated in favor of states $|1\rangle$ and $|2\rangle$ by assuming that the derivatives of the probability amplitudes of states $|i\rangle$ are much smaller than the detunings from these states. The atom-field interaction for the two-photon system may then be written in terms of a two-by-two Hamiltonian

$$H=-\frac{\hbar}{2}\begin{bmatrix} \sum_{q=p,c} a_q|E_q(z,t)|^2 & bE_p(z,t)E_c^*(z,t) \\ b^*E_p^*(z,t)E_c(z,t) & \sum_{q=p,c} d_q|E_q(z,t)|^2-2\Delta\omega_2 \end{bmatrix}. \tag{1}$$

The terms a_q, b, and d_q in the above Hamiltonian are material constants [14] and, for two fields applied in the Raman configuration of Fig. 1(a), are

$$a_q=\frac{1}{2\hbar^2}\sum_i |\mu_{i1}|^2\left[\frac{1}{\omega_{i1}-\omega_q}+\frac{1}{\omega_{i1}+\omega_q}\right],$$

$$d_q=\frac{1}{2\hbar^2}\sum_i |\mu_{i2}|^2\left[\frac{1}{\omega_{i2}-\omega_q}+\frac{1}{\omega_{i2}+\omega_q}\right],$$

$$b=\frac{1}{2\hbar^2}\sum_i \mu_{1i}\mu_{i2}\left[\frac{1}{\omega_{i1}-\omega_p}+\frac{1}{\omega_{i1}+\omega_c}\right]. \tag{2}$$

Here, the unperturbed atomic frequencies $\omega_{ij}\equiv(\omega_i-\omega_j)$. These constants include all rotating wave as well as cross-channel effects. (A general treatment should also include integration over the continuum.) The diagonal elements of the Hamiltonian are the dynamic Stark shifts of states $|1\rangle$ and $|2\rangle$ which would result from the application of either laser, if alone, and the off-diagonal terms are two-photon Rabi frequencies. The relative values of these coefficients obviously depend upon atomic structure and laser detunings, but we may identify two common experimental regimes, near-resonance and far-detuned, corresponding to the detuning of the laser fields from the intermediate state(s). In the near-resonance case, one Stark shift term will dominate for each laser and the counterrotating and cross-channel contributions may be dropped. If the interaction with all intermediate states is dominated by a single state, then $b^2=a_pd_c$ and we recover the ideal three-state results of Harris [15]. In the far-detuned case, the material constants are roughly equal and the effects of each laser field acting on each state must be considered. In either case, the procedure that follows is unchanged.

As a model of inhomogeneous broadening, we consider the interaction of the applied fields with an ensemble of atoms which have a Maxwellian velocity distribution. To our knowledge, an analytical solution of this problem, as it applies to LWI, has not been presented previously, although the problem has been studied numerically [16]. Because of the Doppler shift, different atomic velocity classes will interact more or less strongly with the applied fields, which may lead to selective velocity-class saturation (hole burning). We use the density-matrix formalism, which includes velocity-class-dependent rate and transverse relaxation terms, to describe the evolution of each of these velocity-class atoms. The density-matrix equations, for each particular velocity class of the inhomogeneously broadened ensemble, are generated from the Hamiltonian of Eq. (1) according to $(\partial\rho_{ab}/\partial t)=-i/\hbar[H,\rho]_{ab}$ plus phenomenological rate and relaxation terms

$$\frac{\partial\rho_{11}(z,v,t)}{\partial t}=\mathrm{Im}[b^*E_p^*(z,t)E_c(z,t)\rho_{12}(z,v,t)] +R_1^{\mathrm{in}}\rho_{rr}(z,v,t)-R_1^{\mathrm{out}}\rho_{11}(z,v,t)$$

$$\frac{\partial\rho_{22}(z,v,t)}{\partial t}=-\mathrm{Im}[b^*E_p^*(z,t)E_c(z,t)\rho_{12}(z,v,t)] +R_2^{\mathrm{in}}\rho_{rr}(z,v,t)-R_2^{\mathrm{out}}\rho_{22}(z,v,t), \tag{3a}$$

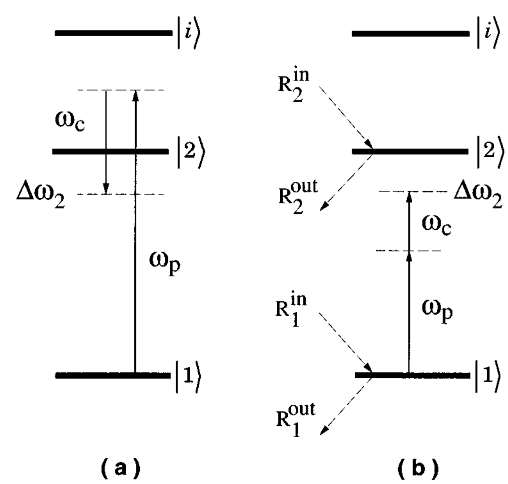

FIG. 1. Energy-level diagram for the (a) Raman laser (Λ field configuration) and (b) two-photon laser (ladder or cascade field configuration). The sum or difference frequency of the probe laser (ω_p) and the coupling laser (ω_c) nearly equals a two-photon atomic transition frequency; the small two-photon detuning parameter $\Delta\omega_2$ is defined by $\omega_{21}-(\omega_p\pm\omega_c)$ and is positive as shown. R_2^{in}, R_1^{out}, etc., are the rates at which population is pumped into or out of each state.

$$\frac{\partial\rho_{12}(z,v,t)}{\partial t}-i\Delta\rho_{12}(z,v,t)=h(z,v,t)$$

$$h(z,v,t)=\frac{i}{2}bE_p(z,t)E_c^*(z,t)[\rho_{22}(z,v,t)-\rho_{11}(z,v,t)]. \tag{3b}$$

The terms $R_1^{\rm in}$ and $R_2^{\rm in}$ represent arbitrary incoherent population transfer rates (in units of rad/s) to states $|1\rangle$ and $|2\rangle$ from a generalized ''reservoir'' state $|r\rangle$; this simple pumping provides no externally injected coherence and may, in general, be a function of the atomic velocity class as well as of position and time. Longitudinal relaxation terms $R_1^{\rm out}$ and $R_2^{\rm out}$ represent the rate at which population is removed from states $|1\rangle$ and $|2\rangle$ to other arbitrary states. The complex effective two-photon detuning parameter is defined as $\Delta=\frac{1}{2}[A(z,t)-D(z,t)]+\Delta\omega_2+i\gamma_{21}$, with $\Delta\omega_2$ defined as shown in Fig. 1. The real part accounts for light shifts, deliberate laser detunings, and two- photon inhomogeneous broadening; the quantities $A(z,t)=(a_p-d_p)|E_p(z,t)|^2$ and $D(z,t)=(d_c-a_c)|E_c(z,t)|^2$ are the net dynamic Stark shifts produced by each of the laser fields acting on either level in the absence of the other laser. As expected, these field-induced Stark shifts enter as two-photon detunings, and in a far-detuned system, $A(z,t)$ and $D(z,t)$ are both close to zero. The imaginary part γ_{21} is the net transverse relaxation rate of the $|1\rangle$-$|2\rangle$ transition coherence [17]. We write $\gamma_{21}=1/T_2+\frac{1}{2}(R_1^{\rm out}+R_2^{\rm out})$, where T_2 is the time scale set by collisional broadening and the longitudinal relaxation terms are present because removal of phased population constitutes a dephasing mechanism.

How is the propagation of the probe and coupling laser pulses influenced by material excitation? Assuming that the electric-field envelopes change slowly in space compared to their wavelengths, the propagation equations for the probe and coupling electric fields in a Raman configuration, in local time, are

$$\frac{\partial E_p(z,t)}{\partial z}=-i\eta\hbar\omega_pN\{[a_p\hat{\rho}_{11}(z,t)+d_p\hat{\rho}_{22}(z,t)]E_p(z,t)+b^*E_c(z,t)\hat{\rho}_{12}(z,t)\}, \tag{4a}$$

$$\frac{\partial E_c(z,t)}{\partial z}=-i\eta\hbar\omega_cN\{[a_c\hat{\rho}_{11}(z,t)+d_c\hat{\rho}_{22}(z,t)]E_c(z,t)+bE_p(z,t)\hat{\rho}_{12}^*(z,t)\}. \tag{4b}$$

Both fields are driven by the macroscopic polarization of the inhomogeneously broadened medium; $\hat{\rho}_{11}(z,t)$ and $\hat{\rho}_{22}(z,t)$ refer to the total fraction of atoms in these states at time t and position z, and $\hat{\rho}_{12}$ is the ensemble coherence. The macroscopic quantities are obtained by summation over atomic velocity classes. (The material constants a_q, etc., are valid for the entire ensemble when the detunings from the intermediate states are much greater than the Doppler widths of these states.) The excess phase shift per unit length of the probe laser alone, with all atoms in state $|1\rangle$, is given by $N\eta\hbar\omega_pa_p$; here, N is the atomic density, η the impedance of free space, and $\hbar\omega_p$the energy of probe-laser photons. The fields are coupled to each other via the ensemble coherence. [To describe a two-photon laser, when the fields are in the ladder or cascade configuration of Fig. 1(b), we make the following replacements: in the definition of the b coefficient ω_c becomes -ω_c, E_c and E_c^* terms are replaced by their respective conjugate variables in Eqs. (1), (3a), (3b), and (4a), and $bE_p\hat{\rho}_{12}^*$ in Eq. (4b) is replaced by its complex conjugate, $b^*E_p^*\hat{\rho}_{12}$.]

II. APPROXIMATE SOLUTION FOR THE ENSEMBLE ATOMIC COHERENCE

We first reformulate Eq. (3b), the differential equation for the coherence, as an integral equation. With variations of the quantity Δ small on a time scale $[\Delta]^{-1}$ over the duration of the probe-laser pulse, the coherence is

$$\rho_{12}(z,v,t)=\int_{-\infty}^{t}h(z,v,\tau)e^{i\Delta(t-\tau)}d\tau. \tag{5}$$

Equation (5) describes the coherence evolution of a particular velocity class of atoms, or of an ensemble of purely homogeneously broadened atoms. We have assumed ''natural'' initial conditions, so that before the laser fields are applied, the coherence of each velocity class is zero.

To proceed analytically in our description of the Doppler-broadened medium, we work in a small-signal, rate-equation regime. Thus, when computing the evolution of each atomic velocity-class population, $\rho_{22}(z,v,t)$ and $\rho_{11}(z,v,t)$, we neglect the coherent interaction term (a two-photon Rabi frequency proportional to the product of field strengths) in favor of the rate terms. This is equivalent to the first-order rate equation approximation theory of Sargent, Scully, and Lamb [18]. Strictly speaking, in a strong-signal regime, each atomic population term will evolve in a manner dependent upon the velocity class; retaining this dependence rigorously would allow us to describe nonadiabatic passage and macroscopic velocity-class saturation effects, but would render the problem analytically intractable. We assume, therefore, that the driving term may be factored as $h(z,v,t)=g(v)h(z,t)$, where v is the component of velocity along an axis defined by the wave vectors of the probe and coupling lasers, and $g(v)$ is a normalized Maxwellian thermal velocity distribution function. Note that this assumption requires that the population excitation mechanism also be factored with the same velocity distribution. This situation may be readily realized if the pumping is provided by, e.g., spontaneous emission from a distribution of thermal atoms.

Unlike the cw case considered in [18], if we postulate ns-length laser pulses, mm size beams, and typical thermal velocities, we may ignore the field averaging due to the motion of individual atoms and consider only the detuning effects. Because of the Doppler shift, an extra velocity-dependent detuning factor $\delta\omega[v]$ must be added to Δ. The velocity variable is converted to a two-photon detuning according to $\delta\omega[v]=(\vec{k}_p-\vec{k}_c)\cdot\vec{v}$, and the Maxwellian velocity distribution leads to a gaussian two-photon detuning distribution function with full width at half maximum $\Delta\omega_D$: $g(\delta\omega)=\sqrt{(4\ln 2)/(\pi\Delta\omega_D^2)}\exp[-4\ln 2(\delta\omega/\Delta\omega_D)^2]$. We note that in two-photon processes, the inhomogeneous linewidth is proportional to the wave vector *sum* (two-photon laser, in which fields are applied in a ladder or cascade configuration) or *difference* (Raman laser, Λ configuration); as is well known, for special ''Doppler-free'' configurations, this residual inhomogeneous broadening can be minimized. Carrying out the integration over the detuning variable $\delta\omega$, we find

$$\begin{aligned}\hat{\rho}_{12}(z,t) &= \int_{-\infty}^{\infty} \rho_{12}(z,v,t)dv \\ &= \int_{-\infty}^{t}\int_{-\infty}^{\infty} h(z,\tau)e^{i(\Delta+\delta\omega)(t-\tau)}g(\delta\omega)d(\delta\omega)d\tau \\ &= \int_{-\infty}^{t} h(z,\tau)e^{i\Delta(t-\tau)}e^{-\gamma_D^2(t-\tau)^2}d\tau, \end{aligned} \tag{6}$$

with the parameter $\gamma_D \equiv \Delta\omega_D/4\sqrt{\ln 2} \simeq 0.3\Delta\omega_D$. We now expand this Doppler- averaged integral in a series solution via integration by parts. This procedure is similar to the solution technique for homogeneously broadened multilevel systems described by Puell and Vidal [19] and references therein. Series expansions have also been used in the description of adiabatic following [20]. The time scales for variations of the different quantities in the integrand determine the rate of series convergence. Assuming that the time scale of variations of the driving term $h(z,t)$ are such that $[\partial^2 h(z,t)/\partial t^2]/[\partial h(z,t)/\partial t] \ll \gamma_D$, (note that this sets a restriction on the rate of change, rather than on the magnitude, of the population excitation rate) Eq. (6) is well approximated by the first two terms of an infinite series. As a function of the (complex) normalized two-photon detuning parameter $\zeta \equiv (\Delta/2\gamma_D)$, the approximate ensemble coherence solution is

$$\hat{\rho}_{12}(z,t,\zeta) \cong \frac{\sqrt{\pi}}{2\gamma_D}\left[h(z,t)G_1[\zeta] - \frac{1}{\gamma_D}\frac{\partial h(z,t)}{\partial t}G_2[\zeta]\right]. \tag{7}$$

This approximate solution is valid in all limits of γ_{21}/γ_D. The two functions in this equation, $G_1[\zeta]$ and $G_2[\zeta]$, are related to the plasma dispersion function $Z[\zeta]$ with a complex argument [18]

$$G_1[\zeta] = Z[\zeta]$$

$$G_2[\zeta] = i\zeta Z[\zeta] + \frac{1}{\sqrt{\pi}}$$

$$Z[\zeta] = e^{-\zeta^2}\mathrm{erfc}(-i\zeta) = \frac{i}{\pi}\int_{-\infty}^{\infty}\frac{e^{-y^2}dy}{\zeta - y} \tag{8}$$

and are shown in Fig. 2. We see that the ensemble coherence is determined by both the population difference term $h(z,t)$ and the rate of population excitation, described by $\partial h(z,t)/\partial t$. Spectral and laser intensity dependencies are contained within the functions $G_1[\zeta]$ and $G_2[\zeta]$.

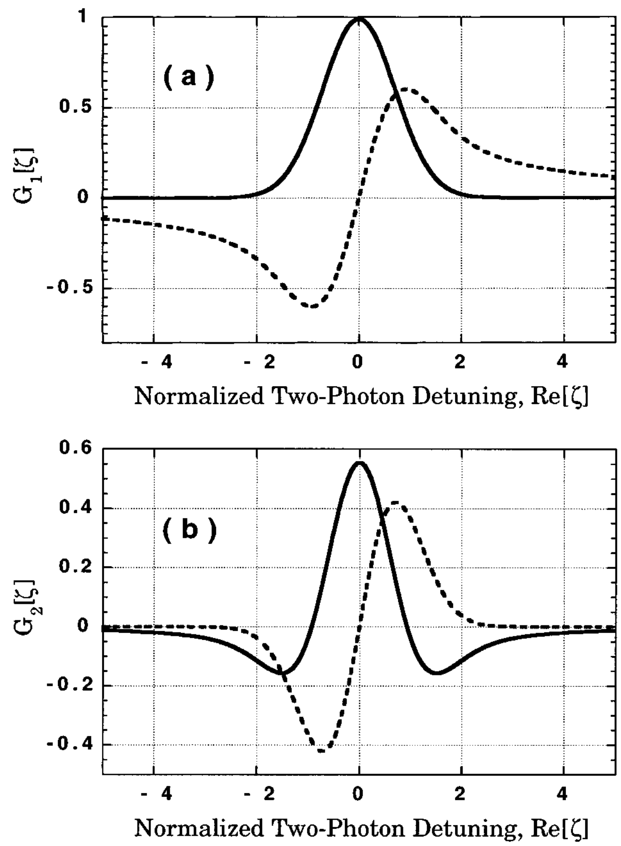

FIG. 2. Real parts (solid lines) and imaginary parts (dashed lines) of the functions (a) $G_1[\zeta]$, and (b) $G_2[\zeta]$. The abscissa is the normalized effective two-photon detuning Re[ζ], with $\zeta = \Delta/2\gamma_D$. For these curves, the normalized coherence relaxation rate $(\gamma_{21}/2\gamma_D) = 0.01$.

III. THE EVOLUTION OF PULSE ENERGY AND AREA

We consider the variation of the probe-laser pulse energy density $\mathcal{E}$ and pulse area $\mathcal{A}$ caused by propagation through an arbitrarily pumped atomic system. The pulse energy density at any point in the medium is the integral of the intensity. The change of this quantity as the pulse propagates through the medium is

$$\begin{aligned}\frac{\partial\mathcal{E}(z)}{\partial z} &= \frac{\partial}{\partial z}\left[\int_{-\infty}^{\infty}\frac{1}{2\eta}|E_p(z,t)|^2 dt\right] \\ &= \frac{1}{2\eta}\int_{-\infty}^{\infty} 2\,\mathrm{Re}\left[E_p^*(z,t)\frac{\partial E_p(z,t)}{\partial z}\right]dt. \end{aligned} \tag{9}$$

When we substitute $\partial E_p/\partial z$ from the probe propagation equation and use the approximate solution for the ensemble coherence $\hat{\rho}_{12}$, the gain and loss of probe-laser energy is found to be

$$\begin{aligned}\frac{\partial\mathcal{E}(z,\zeta)}{\partial z} &= \sigma_0\int_{-\infty}^{\infty} I_c(z,t)I_P(z,t) \\ &\quad\times\left[\mathrm{Re}[G_1[\zeta]] - \frac{1}{2\gamma_D}\mathrm{Re}[G_2[\zeta]]\frac{\partial}{\partial t}\right] \\ &\quad\times(\hat{\rho}_{22} - \hat{\rho}_{11})dt, \end{aligned} \tag{10}$$

with a two-photon absorption parameter $\sigma_0 = N\eta^2\hbar\omega_p|b|^2\sqrt{\pi}/\gamma_D$ [21]. I_c is the coupling laser intensity. Since the coherent interaction terms have been neglected in the derivation of Eq. (6), the energy loss and gain coefficients are independent of pulse area, and due strictly to coherence relaxation and population excitation mechanisms. There are two terms in this equation that modify the probe laser energy density during propagation. (We have neglected a term proportional to the rate of change of the phase difference of the laser fields, which arises from the bandwidth or chirp of the lasers, and frequency pulling due to interaction with the medium. Its value, averaged over the spectral content of the laser fields, is assumed to be much smaller than the Doppler width γ_D, a condition generally satisfied when using transform- limited, several ns-long, laser pulses.)

The first term is due to two-photon absorption and stimulated Raman scattering. We note that this term causes a loss at the probe field unless a two-photon inversion is maintained since the direction of the two- photon absorption is driven by the population difference. Application of a strong probe (pump) laser field to the input of an noninverted Raman-active medium will generate a coherent coupling (Stokes) laser field with a line shape proportional to $\mathrm{Re}[G_1[\zeta]]$ with maximum gain obtained at the position of the Stark-shifted two-photon resonance. If the lower manifold is inverted, then Eq. (10) can describe a stimulated anti-Stokes Raman device. Due to its importance in atomic and molecular spectroscopy, this Raman line shape has been studied extensively in the past; it is also the line shape of coherent anti-Stokes Raman scattering (CARS) [22].

The second term, which depends on the temporal evolution of the atomic population and the function $G_2[\zeta]$, is the primary result of this paper. This term describes changes to probe laser energy due to the coherent trapping of atoms which have been incoherently pumped into the system. Energy is extracted from both the atomic medium and the coupling laser field. *The key to the realization of inversionless lasers is the fact that gain may be provided by the rate of change of a population difference, and not just the absolute value of that difference.* Probe-laser gain is possible regardless of the relative lower manifold atomic populations, is independent of the pulse area, and depends only on the pumping mechanism and the laser intensities and detunings. This result is borne out in numerical simulations, but is in contradiction with the results of earlier theoretical work on Raman LWI by Agarwal, Ravi, and Cooper [23], who predicted that gain will be terminated when the two-photon transition becomes inverted, i.e., when $\hat{\rho}_{22}$ exceeds $\hat{\rho}_{11}$. This gain cannot be regarded simply as a Raman-scattering mechanism since it does not require the existence of a nonallowed transition homogeneous linewidth. In addition, it does not require phase matching. The complete line shape is plotted in Fig. 3 for several normalized pumping rates; note how it blends smoothly with the unsaturated Raman line shape when the pumping rate becomes very small.

Similarly, the pulse area $\mathcal{A}$ is defined as the integral of the magnitude of the electric field and may change as the pulses propagate. We determine its evolution in a manner similar to Eq. (10); if the coupling laser is taken as constant over the duration of the probe pulse (which is the case if the probe-laser intensity is much weaker than that of the coupling laser [24]) then

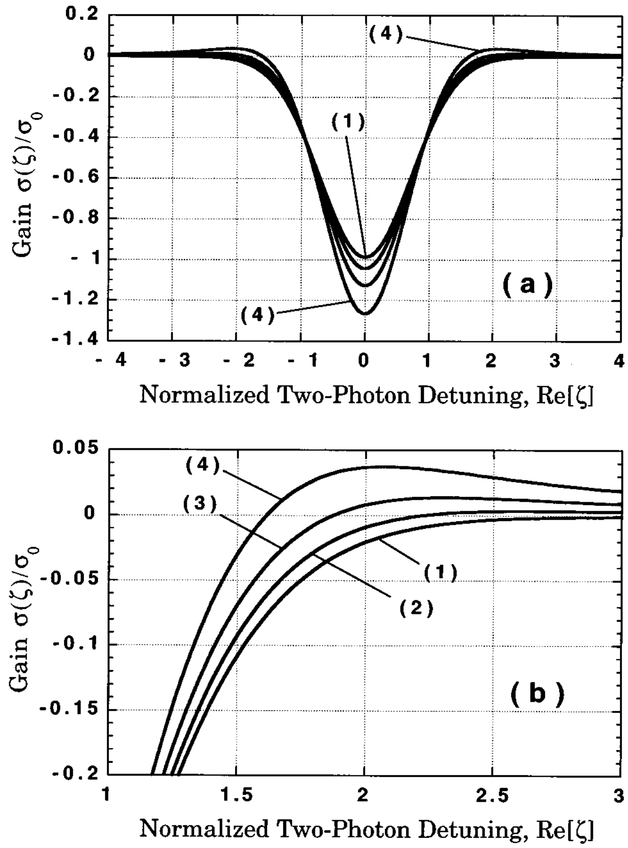

FIG. 3. Anti-Stokes Raman gain line shape. (a) Small-signal plane-wave gain $\sigma(\zeta)/\sigma_0$, (with $\mathcal{E}(z)=\mathcal{E}_0\exp[\sigma(\zeta)(I_c/2\pi)z]$) is plotted vs normalized effective two-photon detuning. The normalized pumping parameter $\mathcal{P}=(1/2\gamma_D)(\partial/\partial t)[\hat{\rho}_{22}(z,t)-\hat{\rho}_{11}(z,t)]$ is increased from 0.0 (curve 1), to 0.10 (curve 2), to 0.25 (curve 3), to 0.5 (curve 4). In each curve, the normalized coherence relaxation rate $(\gamma_{21}/2\gamma_D)=0.01$ and the factor $[\hat{\rho}_{22}(z,t)-\hat{\rho}_{11}(z,t)]=-1.0$, i.e., all atoms in state $|1\rangle$. (b) Expansion of the positive region of these curves. The normal two- photon absorption line shape is obtained (curve 1) in the absence of pumping; a large pumping rate (curve 4) permits significant inversionless gains over a wide range of effective detuning, to either side of the two-photon resonance.

$$\frac{\partial\mathcal{A}(z,\zeta)}{\partial z}=\frac{\sigma_0}{2}\int_{-\infty}^{\infty}I_c(z,t)|E_p(z,t)|\mathrm{Re}[G_1[\zeta]](\hat{\rho}_{22}-\hat{\rho}_{11})dt. \tag{11}$$

This indicates that evolution of the pulse area is independent of the pumping rate. In general, therefore, as the probe-laser pulse is amplified without inversion, it is also temporally compressed to keep the total pulse area constant, and thus the Raman and two-photon laser amplifiers are also nonlinear pulse compressors, as predicted by Narducci *et al.* [9]. Dephasing mechanisms also act to modify the pulse area, regardless of amplification.

Following the rate equation approximation, the difference of the state populations in Eq. (10) is determined by rates only, i.e., $\partial/\partial t[\hat{\rho}_{22}-\hat{\rho}_{11}]=R_2^{\mathrm{in}}\hat{\rho}_{22}+R_1^{\mathrm{out}}\hat{\rho}_{11}+\cdots$. However, pulse propagation itself will cause the populations to change slightly in a characteristic group velocity signature [24], independently of the pumping mechanism. Energy is absorbed from the front part of the probe pulse to prepare the superposition eigenstate (increasing the upper-state population) and then coherently added to the back of the pulse. If the fields are applied adiabatically, the integral of this process is nearly zero; changes in probe energy are a result of preparing fresh and dephased atoms. Comparison with Eq. (10) for net energy gain indicates that if the difference of atomic populations is in a steady state, i.e., $\partial/\partial t[\hat{\rho}_{22}-\hat{\rho}_{11}]=0$, inversionless gain is impossible. This is a major difference between the far-detuned, two-photon system considered here and continuous-wave, resonant-intermediate experiments: no gain is possible in steady state in the far-detuned system. Far-detuned, coherent, inversionless gain effects are thus a transient phenomena and will lend themselves to pulsed laser experiments. In resonant experiments [11,12], it is the coupling of the intermediate state $|i\rangle$ to state $|2\rangle$ which recycles part of the atomic population and makes small-signal cw LWI possible even while the atomic populations are in steady state. We note that the dynamics described above are characteristic to both Raman and two-photon LWI.

IV. SMALL-SIGNAL GAIN

We now estimate the gain coefficients for a model atomic system. For the sake of continuity with existing experiments, we calculate numbers for a low-pressure atomic vapor of isotopically pure ^{208}Pb [24]. This is a Raman (Λ) system with a large 10 650 cm^{-1} shift (1 cm^{-1}=30 GHz) with ns-duration probe and coupling lasers detuned 1000 cm^{-1} from the intermediate state. Probe-laser photon energy is 4.3 eV and matrix elements are each taken as 1 a.u., so that the value of b is approximately 1.7×10^{-5} (mks units). At a temperature of 900 °C, the two-photon transition is primarily Doppler broadened: the density is about 10^{15} cm^{-3}, and the Doppler width is 0.02 cm^{-1}, 100 times greater than the 2×10^{-4} cm^{-1} collisionally broadened linewidth. (This corresponds to a 25 ns dephasing time T_2). Higher-order corrections to the gain formula of Eq. (10) may be ignored, therefore, when the time scale for pulse evolution, etc., is much longer than about 800 ps. The small-signal, plane-wave gain coefficient under these conditions is given by

$$\sigma(\zeta)=\sigma_0\left[\mathrm{Re}[G_1[\zeta]]-\frac{1}{2\gamma_D}\mathrm{Re}[G_2[\zeta]]\frac{\partial}{\partial t}\right](\hat{\rho}_{22}-\hat{\rho}_{11})$$

$$\mathcal{E}(z)=\mathcal{E}_0\exp\left[\sigma(\zeta)\frac{I_c}{2\pi}z\right], \tag{12}$$

where all frequencies are radian frequencies, and I_c is the coupling laser intensity. The following numerical estimates take I_c in units of [MW/cm^2]. The value of σ_0 for these parameters is 4.7×10^{-3} cm/MW. For comparison purposes, if this system were completely inverted and a strong-coupling laser were applied, the probe field would be generated by normal stimulated anti-Stokes Raman scatter with a gain coefficient of $7.5\times10^{-4}\times I_c$ cm^{-1}. The magnitude of the inversionless gain depends upon the rate at which we pump the system. In particular, it is desirable to maximize the dimensionless pumping parameter $\mathcal{P}$ $=(1/2\gamma_D)\partial/\partial t[(\hat{\rho}_{22}-\hat{\rho}_{11})]$. Assuming that 10% of the population is pumped into state $|2\rangle$ in 0.5 ns (a pumping rate 5 times the coherence relaxation rate) then $\mathcal{P}=0.10$. At this pumping rate, the gain factor $(\sigma(\zeta)/\sigma_0)$ reaches a maximum value of 2.7×10^{-3} when the difference of Stark shifts and laser detuning is approximately two Doppler widths. (See Fig. 3, curve 2) In the most severe case, with all atoms in the ground state ($|1\rangle$), the maximum inversionless anti-Stokes gain is found to be $2\times10^{-6}\times I_c$ cm^{-1}. Similar to other resonantly enhanced two-photon processes, this gain is a strong function of detuning from the intermediate level. Decreasing the detuning to 5 cm^{-1}, with other parameters held constant, brings the inversionless anti-Stokes gain up by a factor $(200)^2$ to $0.08\times I_c$ cm^{-1}. However, increasing the coupling laser intensity (especially near resonance) modifies the effective two-photon detuning Δ through the stark shift parameter $D(z,t)$, and reduces the available gain; in order to maintain an optimum value of Δ, the laser two-photon detuning may also be adjusted accordingly.

V. PREPARATION OF AN INHOMOGENEOUSLY BROADENED ATOMIC MEDIUM

Although the Raman loss maximizes at the position of the Stark-shifted two-photon resonance as expected, it is not immediately obvious why the gain due to population excitation does not also maximize at this point. We see from Fig. 2 that the function $G_2[\zeta]$ actually maximizes at values of the effective detuning roughly equal to the inhomogenous linewidth. *These detunings determine the maximum gain that may be achieved in a laser amplifer configuration, or the frequency of a probe-laser oscillator which builds from noise.* We posit that these optimum detunings maximize the real part of the ensemble atomic coherence for given probe and coupling laser intensities, and that this maximally coherent condition is the basis for tunable inversionless lasers.

Our understanding of the inversionless gain line shape will be aided considerably by a knowledge of what is happening on a single-atom level. Under the influence of the applied fields, atoms in $|1\rangle$ are adiabatically evolved into a phased or an anti-phased superposition of states $|1\rangle$ and $|2\rangle$. The coherence of a given velocity-class atom in this superposition state, is given (with z and t dependence omitted but understood) by [14]

$$\rho_{12}(v)=\pm\frac{bE_pE_c}{2\sqrt{\Delta[v]^2+|b|^2|E_p|^2|E_c|^2}}. \tag{13}$$

The evolution of a particular velocity-class ground-state atom is determined by the sign of the effective two-photon detuning Δ. If the effective detuning is negative, then an atom initially in $|1\rangle$ is evolved into the antiphased superposition state (so that $\mathrm{Re}[\rho_{12}(z,v,t)]$ is negative), and vice versa. In order to maximize the time-averaged ensemble coherence, the net difference in Stark shifts and laser detunings must be of one sign for each atom in the ensemble, i.e., $\frac{1}{2}[A(z,t)-D(z,t)]+\Delta\omega_2\ll-\Delta\omega_D$. With this detuning, a large fraction of the atomic velocity classes are ''prepared'' into the antiphased superposition state. As the magnitude of the effective detuning is increased, the magnitude of the ensemble coherence first increases, as more and more velocity classes are prepared; however, increasing the two-photon detuning actually reduces the single velocity-class interaction strength and so the ensemble coherence will begin to decrease. The maximum value occurs for effective detunings of approximately one inhomogeneous linewidth to either side of the resonance. This preparation condition is common to many electromagnetically induced transparency (EIT) experiments.

In addition to the requirement on the magnitude of the effective two-photon detuning, the field strengths must be adiabatically changed to ensure that each atom remains in a given eigenstate; the time scale for field variations must therefore be small compared to the inverse of the separation between eigenstates of the system, which, at any point, is $[2\sqrt{\mathrm{Re}[\Delta]^2+|b|^2|E_p(z,t)|^2|E_c(z,t)|^2}]^{-1}$.

It is evident that preparation may be accomplished either with laser intensity (through the difference in Stark shift factors) or with a deliberate two-photon detuning, depending on the detunings from the intermediate state(s) and the available laser power. Preparation into the antiphased eigenstate with minimum $\Delta\omega_2/D(z,t)$ is advisable, whenever possible, to take advantage of the inherent beam propagation protection that occurs in EIT [25]. In the near-resonant intermediate case, optimal preparation may be realized by zero detuning and counterintuitive pulse propagation, whence a strong-coupling laser [such that $D(z,t)>2\Delta\omega_D$] is sent into the medium prior to the probe laser. (If the probe laser builds up from noise, we expect that the medium will ''self-prepare'' by generating matching coupling and probe-laser envelopes [26].) The recent development of high-power, narrow-linewidth, laser systems like those described in [27] has enabled experimenters to adiabatically prepare atoms of a strongly absorbing, Doppler-broadened media with laser fields alone at two-photon resonance. Note that in the far-detuned case, where the Stark shift coefficients are roughly equal, preparation is accomplished by deliberate detuning to either side of the two-photon resonance.

VI. EXPERIMENTAL CONSIDERATIONS

We now make some general remarks on the type and preparation of atomic media suitable for the demonstration of these concepts. The requirement on the coupling laser is not terribly restrictive, because the gain is independent of pulse shape. The gain does, however, depend on the dephasing rate, which, in turn, depends upon the laser phase-diffusion linewidth, which recommends the use of transform-limited lasers. The atomic medium may be chosen to match available or desired laser wavelengths; while the gain may be polarization-dependent, there are no phase-matching considerations and thus material dispersion is irrelevant.

The choice of states $|1\rangle$ and $|2\rangle$ is quite important. In both normal and inversionless lasers, a certain state must be pumped faster than the relevant relaxation mechanism. In normal lasers, an inversion condition must be maintained; in inversionless lasers, the coherence must be maintained. Gain is not possible in either case if the medium is in local thermodynamic equilibrium (LTE). In LTE, there will, of course, be no inversion, but the more subtle point, which precludes inversionless gain under these conditions, is that the gain due to the forward rate into state $|2\rangle$ is exactly cancelled by the backward rate-induced dephasing loss. The ability to maintain ''one-way'' rates (e.g., via spontaneous emission pumping) is key to the success of inversionless lasers. To minimize the coherence relaxation rate, a metastable state $|2\rangle$ is the obvious choice. It may be possible to develop a unique kind of recombination laser from these concepts, but electron pumping is unavoidably accompanied by some measure of Stark broadening (collisional broadening by charged particles) of the $|1\rangle$-$|2\rangle$ transition. This effect is relatively benign to standard recombination laser systems, but deadly to the coherence-based laser described here.

We note that the transient nature of the gain leads to a natural choice of long, single-pass cells. This is different from Raman cells, in which a single, multipass cell configuration may be optimized for maximal generation of a given sideband [28]; further increase in path length actually reduces the conversion efficiency to this sideband, an indication of the parametric nature of the energy transfer in Raman scattering. Moreover, narrow beams or experimental cells will generally be required in order to minimize optical trapping effects which lead to reverse rates. If permitted by material constraints, both two-photon and Raman processes will, in general, occur simultaneously, resulting in the amplification of probe frequencies $\omega_{21}\pm\omega_c$.

There is a subtle aspect to the nature of the pumping mechanism that will drive an inversionless laser. Thermodynamically, the gain of any laser, with or without inversion, is derived from an irreversible process. In an inversionless laser, gain is the result of a nonadiabatic interaction between the laser fields and those atoms pumped into the lower manifold. In a sense, we have replaced nature's ready-made irreversibility (the elastic and inelastic dephasing rate of the lower manifold, which constitutes the linewidth of the two-photon transition) with our own. The single-atom pumping process must therefore occur on a time scale which is short compared to the reaction time of the system. Otherwise, the system will have time to evolve in a reversible manner during the course of the pumping and any energy gained by the radiation fields will be returned to the medium at the end of the pulse. The relevant reaction time scale for atomic media described in this paper is the inverse of the effective two-photon detuning, which, for a weak probe laser may be written $|\Delta\omega_2-D(z,t)/2|^{-1}$. Following the prescription for maximum gain, the magnitude of this detuning will be roughly equal to the two-photon Doppler width, which for typical atomic vapors implies a reaction time of several hundred ps. Not all pumping mechanisms satisfy this requirement, e.g., slow collisions between atoms and low-energy electrons. Equation (3) implicitly assumes instantaneous single-atom pumping events, a valid approximation for, e.g., spontaneous emission.

Intermediate state resonant enhancement of the coherent two-photon gain is quite significant. Traditionally, the cost of tuning lasers ever closer to an intermediate state is drastic beam quality degradation due to one- photon absorption and a self-phase and cross-phase modulation-induced nonlinear index of refraction. Under these circumstances, however, if the medium is prepared in the manner described in the previous sections into the antiphased superposition (dark) state, the index of refraction is zeroed, and the lasers can utilize the intermediate resonance with impunity. In a manner identical to resonant-intermediate LWI, the generated probe beam will be protected from reabsorption by quantum interference. Experimentally, we expect to tune both probe and coupling lasers to resonance with the intermediate state and observe significantly enhanced gains. This is the mechanism for the enhanced nonlinear optical conversion efficiency of [29]. Our formalism may in fact be used to describe resonant-intermediate interactions, when the resonant intermediate state can be adiabatically eliminated. This is possible when its homogeneous linewidth is very large, or when the medium is counterintuitively prepared with a strong-coupling laser [30].

However, it will not be desirable to tune to *exact* resonance with the intermediate state. High-gain, resonant-intermediate experiments are severely limited by coupling laser absorption [31]. At a fixed pumping rate, the symmetry of the line shape in Fig. 3 leads us to expect gain regardless of the sign of the effective detuning from two-photon resonance. As discussed previously, the sign of the net detuning determines the superposition state into which state $|1\rangle$ atoms are evolved; hence the behavior of the superposition states in the regime far-detuned from the intermediate state(s) is identical. However, when the lasers are tuned within the absorption profile of the intermediate resonance, this symmetry is broken, and the fraction of atoms which are not stimulated to the antiphased superposition state will couple strongly to the intermediate state, and lead to an absorption coefficient of the coupling laser nearly equal to the gain coefficient of the probe laser. Hence, after a characteristic propagation distance, resonant-intermediate, inversionless lasers will self-terminate. On the other hand, far-detuned experiments obey a photon conservation relation; for every probe photon generated, a coupling laser photon is absorbed (Raman laser) or generated (two-photon laser) [32]. In far-detuned experiments, therefore, the ratio of the coupling laser loss coefficient to the probe-laser gain coefficient is nearly equal to the ratio of the intensities, and for weak-probe experiments, the coupling laser absorption (or gain) is quite small. We therefore expect that inversionless lasers in opaque media will exhibit maximum gain when both lasers are detuned just outside the absorption profile of the intermediate resonance, and when the ensemble is prepared in the antiphased (dark) eigenstate.

VII. SUMMARY

We have discussed the propagation dynamics and spectral dependence of pulsed, widely tunable, inversionless anti-Stokes Raman laser amplifiers in inhomogeneously broadened atomic media, in regions of loss-free probe laser propagation. Our formalism also describes normal Raman and two-photon absorption phenomena. The approximate solution for the ensemble coherence may be employed, without higher-order terms, when higher-order temporal derivatives of the atomic variable $h(z,t)$ are small compared to the two-photon doppler width, i.e., $[\partial^2 h(z,t)/\partial t^2]/[\partial h(z,t)/\partial t] \ll \gamma_D$. The prescription for observing inversionless amplification is simple: one of two lower atomic states is selectively pumped and the differential pumping rate of these states must exceed the total dephasing rate of the two-photon transition coherence. Because inversionless gain requires a nonadiabatic interaction between the applied fields and the pumped atom, the single-atom pumping event must occur on a time scale short compared to the inverse of the effective two-photon detuning. For a given atomic system and laser frequencies, maximum gain is obtained when the fields are detuned roughly one inhomogeneous linewidth to either side of a two-photon resonance. Unlike the resonant-intermediate case, in the off-resonance region, the behavior of the phased and antiphased superposition eigenstates is identical; it is the magnitude of the real part of the ensemble coherence, not the sign (which is closely linked to quantum interference effects), that matters most. The gain in such lasers will vanish when the atoms are in local thermodynamic equilibrium. Consistent with existing experimental EIT results, it should be possible to significantly resonantly enhance the inversionless gain without propagation distortion if the atoms are prepared in the antiphased superposition (dark) state by appropriate negative effective detuning. We note that a typical Raman laser (and parametric four-wave-mixing processes, such as CARS) operates without inversion, but the energy in this case is not obtained from the atomic medium: it is parametrically transferred from one laser field to another. Anti-Stokes gain, i.e., probe-laser gain, is not normally possible unless the two-photon transition is inverted. In the inversionless laser scheme described here, energy is extracted both from the atomic medium and from a separate coherent field. The major difficulty in the realization of inversionless lasers continues to be the selection of an optimal pumping mechanism. Once this problem is overcome, however, it may be possible to extend the principles of this paper to the design of short-wavelength laser amplifiers.

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