Coherence-controlled transparency and far-from-degenerate parametric gain in a strongly-absorbing Doppler-broadened medium

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An inversionless amplification of anti-Stokes radiation above the oscillation threshold in an optically-dense far-from-degenerate double-Λ Doppler-broadened medium accompanied by Stokes gain is predicted. The outcomes are illustrated with numerical simulations applied to sodium dimer vapor. Optical switching from absorption to gain via transparency controlled by a small variation of the medium and of the driving radiation parameters which are at a level less than one photon per molecule is shown. Related video/audio clips see in: A.K. Popov, S.A. Myslivets, and T.F. George, Optics Express 7, No 3, 148 (2000). http://www.optics-express.org/abstract.cfm?URI=express-07-03-148

The concept of quantum coherence and interference in multilevel schemes as an origin of difference in probabilities of absorption and induced emission plays an important role in optical physics. A mechanism for achieving amplification without bare-state population inversion (AWI) in an optical transition based on this concept was explicitly proposed, numerically illustrated for the V-type scheme of Ne transitions, and experimentally proved. Coherence and interference effects have been extensively explored to manipulate energy level populations, nonlinear-optical response, refraction, absorption and amplification of optical radiation in resonant media over the past decade (for a review, see Ref. 4). Recent theoretical and experimental investigations have demonstrated a slowing down of the light group speed to a few meters per second, highly efficient frequency conversion, optical switches, and potential sources of squeezed quantum state light for quantum information processing on this basis. In this Letter we propose a scheme for coherent quantum control (CQC) that enables one to form upconverted AWI without incoherent excitation in an initially strongly absorbing optically dense medium as well as to accomplish optical switching. All this is shown to be possible by application of driving fields at the level of one photon per several molecules. Unlike other major approaches, the proposed scheme does not require coherent population trapping. In contrast to the research reported in Ref. 7, for which the frequencies of all the coupled fields are almost equal, we consider far-from-degenerate CQC, for which the features of an optically dense Doppler-broadened medium play a crucial role. The interplay of absorption, gain, four-wave mixing (FWM) and their interference at inhomogeneously broadened transitions is shown to result in substantial enhancement of the gain by proper resonance detuning, which was not realized in a recent experiment.

Let us consider four-level scheme [Fig. 1(a)]. The fields $E_1(\omega_1)$ and $E_3(\omega_3)$ are driving, whereas $E_4(\omega_4)$ and $E_2(\omega_2)$ are weak probes. The problem under consideration reduces to the solution of a set of the coupled equations for four waves, $(E_i/2) \exp[i(\tilde{k}_i z - \omega_i t)] + c.c.\ (i = 1\ldots4)$, copropagating in an optically thick medium:

$$dE_{1,2}(z)/dz = i\sigma_{1,2}E_{1,2} - i\tilde{\sigma}_{1,2}E_3E_2, \quad (1)$$

$$dE_{1,3}(z)/dz = i\sigma_{1,3}E_{1,3} + i\tilde{\sigma}_{1,3}E_4E_3. \quad (2)$$

Here $\omega_4 + \omega_2 = \omega_1 + \omega_3$, $k_j$ are wave numbers in vacuum; $\sigma_j = -2\pi k_j \tilde{\chi}_j$ and $\tilde{\sigma}_j = -2\pi k_j \chi_j = \delta k_j + i\alpha_j/2$; and $\chi_j$, $\alpha_j$, and $\delta k_j$ are intensity-dependent cross-coupling susceptibilities, absorption indices, and dispersion parts of $k_j$. If $E_{1,3}$ are homogeneous along $z$ (e.g., at the expense

![FIG. 1: (a) Energy levels and (b) AWI, where $\Omega_4 = \omega_4 - \omega_{nl}$, $L_4 = \alpha_4^0$, $\alpha_4 = \alpha_4(G_1 = 0, \Omega_4 = 0)$, $G_{10} = 100$, $G_{40} = 40$ MHz, $\Omega_1 = \omega_1 - \omega_{pl} = 0$ and $\Omega_3 = \omega_3 - \omega_{mn} = 100$ MHz. of saturation), the system of Eqs. (1) and (2) reduces to two coupled equations for $E_4$ and $E_2$, where the medium parameters are homogeneous as well. Their solution is:

$$E_2 = \exp\left(-\frac{\alpha_2}{2} z - \beta z\right) \left\{ -i\frac{\gamma_2}{R} E_{40} \sinh(Rz) \right\}$$

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220
170
2
120
70
20
10
0
0
500
750
1000
250
200
150
100
50
0

FIG. 1: (a) Energy levels and (b) AWI, where $\Omega_4 = \omega_4 - \omega_{nl}$, $L_4 = \alpha_4^0$, $\alpha_4 = \alpha_4(G_1 = 0, \Omega_4 = 0)$, $G_{10} = 100$, $G_{40} = 40$ MHz, $\Omega_1 = \omega_1 - \omega_{pl} = 0$ and $\Omega_3 = \omega_3 - \omega_{mn} = 100$ MHz.
\[ + E_{20} \left\{ \cosh(Rz) + \frac{\beta}{R} \sinh(Rz) \right\} , \]
\[ E_4 = \exp \left\{ -\frac{\alpha_4}{2} z + \beta z \right\} \left\{ \frac{1}{R} E_{20}^4 \sinh(Rz) \right\} , \]
\[ + E_{10} \left\{ \cosh(Rz) - \frac{\beta}{R} \sinh(Rz) \right\} . \]

Here \( R = \sqrt{\beta^2 + \gamma^2} \), \( \beta = \left( (\alpha_4 - \alpha_3)/2 + i \Delta k \right) / 2 \), \( \Delta k = \delta k_1 + \delta k_3 - \delta k_2 - \delta k_4 \), \( \gamma^2 = \gamma_4^2 \gamma_4 \), \( \gamma_{4,2} = \chi_{4,2} E_1 E_3 \), and \( E_{20} \) and \( E_{10} \) are input values (at \( z = 0 \)). The first terms in braces indicate FWM and second – optical parametric amplification (OPA) processes. If either of the driving fields is switched off, \( \gamma_{4,2} = 0 \), and the weak radiations are described as \( |I_{4,2}|^2 = |I_{10,20}|^2 \exp(-\alpha_{4,2} z^2) \) \((I_{4,2} = |E_{4,2}|^2)\). Owing to FWM coupling, probe radiation \( E_4 \) generates a wave \( E_2 \) close to \( \omega_m \), which in turn contributes to \( E_3 \) because of FWM. This process results in correlated propagation of two waves along the medium. A gain or absorption of any of them influences the propagation features of the other. If an absorption (gain) exceeds the rate of the FWM conversion \((|\gamma|^2/|\beta|^2 < 1)\), \( \Delta k = 0; \) if \( E_{20} = 0, \) and \( E_{40} \neq 0, \) we obtain at \( z = L \) the result that \( I_{4}/I_{10} = \left| \exp(-\alpha_4 L/2) + \left( |\gamma|^2/|\beta|^2 \right)^2 \exp(g_2 L/2) - \exp(-\alpha_4 L/2) \right|^2 \), where \( g_2 = -\alpha_2 \). Alternatively, if \( E_{40} = 0, \) and \( E_{20} \neq 0, \) \( \eta_{4} = I_{4}/I_{10} = \left( |\gamma|^2/|\beta|^2 \right)^2 \exp(g_2 L/2) - \exp(-\alpha_4 L/2) \). One can see that achieving gain requires large optical lengths \( L \) and significant Stokes gain on the transition \( g n \) \((|\exp(g_2 L/2)|^2 \approx |(2\beta/|\gamma|^2)|^2)\), as well as effective FWM at both \( \omega_2 \) and \( \omega_4 \). The dependence \( I_{4}(L) \) is predetermined by the sign of \( Im\gamma_{4,2} \) and \( Re\gamma_{4,2} \).

The important feature of the far-from-degenerate interaction is that the magnitude and the sign of the multiphoton resonance detunings and, consequently, of the amplitude and phase of the lower-state coherence \( \rho_{nl} \) differ for molecules at different velocities because of the Doppler shifts. Such is not the case in near-degenerate schemes. The interference of elementary quantum pathways, with Maxwell’s velocity distribution and saturation effects taken into account, results in a nontrivial dependence of the macroscopic parameters on the intensities of the driving fields and on the frequency detunings from the resonances \( \boxed{0} \). A density matrix solution was found exactly with respect to \( E_{1,3} \) and to a first approximation for \( E_{4,2} \). We use these formulas here for numerical averaging over velocities, for analysis of the quantities \( \alpha_{4,2} \) and \( \gamma_{4,2} \), and also to obtain a numerical solution of the system of Eqs. \( \boxed{1}-\boxed{4} \), with the inhomogeneity of the coefficients taken into account. We stress that the effect under consideration is AWI rather than conventional OPA accompanied by absorption and Stokes gain, because the quantum interference involved in resonant schemes is so crucial that thinking in terms of the Manley–Rowe conservation law would be misleading. \[ \boxed{5} \]

The main outcomes of the simulations the conditions of the experiment are illustrated in Figs. 1(b), 2 and 3. The transitions of Fig. 1(a) and relaxation parameters are attributed to those of \( Na_2 \): \( \lambda_{1-4} = 655, 756, 532, 480 \text{ nm}, \gamma_{m, g, n} = 230, 200, 30, \gamma_{mn, ml, gn} = 24, 10, 40, \Gamma_{mn, ml} = 110, \Gamma_{gm} = 130, \) and \( \Gamma_{ng}, \Omega_0 = 140 \) (all in \( 10^6 \text{ s}^{-1} \)). Here \( \Gamma_i \) is the population, \( \Gamma_{ij} \) are the coherence, and \( \gamma_{ij} \) are the spontaneous interlevel relaxation rates. At \( T = 450 \text{ °C} \), the Doppler FWHM of the transition at \( \lambda_4 \) is 1.7 GHz, and the Boltzmann population of level \( n \) is 2% of that of level \( l \). The Rabi frequencies \( G_1 = E_1 d_{ij}/2h \) and \( G_3 = E_3 d_{nnm}/2h \) of \( \sim 100 \text{ MHz} \) correspond to 100-mW beams focused on a spot with sizes of a few parts of a millimeter, i.e. one photon per several molecules. However, the presence of such fields give

![FIG. 2: (a) Macroscopic Stokes gain, (b) absorption, and (c-f) cross-coupling indices: \( \Omega_2 \equiv \omega_2 - \omega_m = \Omega_3 + \Omega_4 - \Omega_4 \). Solid curves, same parameters as in Fig. 1(b); dashed curves, \( G_1 = 43 \text{ MHz}, G_3 = 39 \text{ MHz} \) (corresponding to \( L/L_4 = 20 \), where the gain reaches its maximum value).](image)

![FIG. 3: (a,b) Coupled fields versus optical length (\( L_2 \) is reduced by \( 10^4 \)). (c) CQC optical switching. For (a) and (b) and for the inset in (c), \( \Omega_4 = 160 \text{ MHz} \), and for the inset in (b), \( \Omega_1 = \Omega_2 = \Omega_3 = \Omega_4 = 0 \). The remaining parameters are the same as in Fig. 1(b). \([(c) I_4/I_{10} = 5 \times 10^{-4} \text{ at } \Omega_4 = 75 \text{ MHz} \text{ and } I_4/I_{10} = 10^{-2} \text{ at } \Omega_4 = 69 \text{ MHz} \text{ inset in (c), } I_4/I_{10} = 10^{-4} \text{ at } G_{10} = 80 \text{ MHz and } I_4/I_{10} = 10^{-2} \text{ at } G_{10} = 89 \text{ MHz} \).](image)
imaginary parts take on even different signs [as in Fig. 2(c)-2(f)]. This behavior is in marked contrast to that of solid-state and off-resonant nonlinear optics.

The inhomogeneity of the driving fields [Fig. 3(a)] gives rise to a significant change of the material parameters along the medium (dashed curves in Fig. 2), so $\alpha_4$ may even increase above its value in the weak-field limit. The interplay of these effects determines the spatial dynamics, optimum parameters, and achievable gain [Fig. 1(b)]. Along a substantial medium length, the probe field is only being depleted [Fig. 3(b)]. Its growth begins at the length where the generated and enhanced field $E_2$ (dashed curves in Fig. 3(b)) becomes comparable with $E_{40}$. The simulations explicitly reveal that the fully resonant conditions explored in Ref. 8 are far from optimal [inset in Fig. 3(b)], and most probably the gain reported in Ref. 8 is a misinterpretation of the experiment. The maximum gain in Fig. 1(b) is 1050, which is well above the characteristic threshold for self-oscillation to be established inside the optical cavity from the spontaneous radiation. This gain can readily be increased further to the mirrorless oscillation level. Both linear and laser-induced nonlinear dispersion inhomogeneous along the medium are taken into account in Figs. 1(b) and 3. Our results also demonstrate that the problem of AWI in similar schemes may not be reduced to the condition of a sign change of $\alpha_4$, as was done in the research reported in Ref. 11. The solid curve in Fig. 3(b) shows that there is an optical thickness controlled by the driving radiations whose small variation results in a switching from the absorption regime to transparency and further to amplification. Figure 3(c) presents the possibility of controlling this switching with a small change of either the frequency of the probe radiation or the intensity of the driving radiation (inset, Fig. 3). Obviously, the same processes can be employed for generating and manipulating large dispersion without the accompanying depletion of radiation. The required intensity can be further decreased in identical but more favorable atomic schemes. Owing to the generated molecular coherence, fields $E_4$ and $E_2$ may possess nearly perfect quantum correlations that yield almost complete squeezing.

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