Exponential gain in resonant four-wave mixing via dressed inversions

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High-efficiency coherent high-frequency generation via resonant four-wave mixing is investigated in a dressed states analysis of a bichromatically driven three-level system. We propose the existence of exponential gain due to population transfer at the anticrossings of closely spaced inverted dressed states. This occurs when the Rabi frequency of the weaker detuned driving field is much smaller than the leading resonant Rabi frequency and simultaneously comparable to the largest atomic relaxation rate. The dressed population inversion and thus the exponential gain is not affected until the generated field becomes comparable in intensity to one of the two applied fields, when saturation may take place. The analysis is applied to a krypton configuration of interest to current experiments. [S1050-2947(98)07302-8]

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The enhancement of coherent light amplification in the higher-frequency regime via the application of one or several external lower-frequency fields is by now a well established phenomenon [1–9]. In the area of lasing without inversion, such an additional applied field may lead via destructive pathway interference or population trapping to the inhibition of stimulated absorption and thus to enhanced coherent light amplification [1]. A particularly useful picture of understanding the underlying physics of these processes is in the basis of the eigenstates of the full Hamiltonian, which includes both the atom plus the applied strong laser fields (the so-called dressed states [2]). Here trapping of ground-state population in one of these dressed states and gain due to ''hidden inversions'' between dressed states has been identified as one of the main mechanisms of lasing without inversion [3]. Equally coherent field amplification via resonant four-wave mixing [4–7] owes its origin to the additional applied laser fields. The influence of three laser fields of which two often jointly couple to one dipole forbidden transition of a three-level atom makes the generation and amplification of a fourth usually high-frequency field possible.

One aim of this paper is to obtain additional insight into the process of resonant four-wave mixing via an analysis in the dressed states basis. As opposed to the common situation in lasing without inversion both the excited and the ground state of the transition with enhanced coherent field generation are coupled to each other via external driving fields. For this reason we find more similarities to the well understood process of light amplification in driven two-level systems [8,9]. Initial gain does not need to arise from a spontaneous photon or a probe field but arises due to the coherence established directly by the strong external driving laser fields. Then in the buildup stage of this field we find the possibility of a population inversion between dressed states at anticrossings and consequently exponential gain of resonant four-wave mixing in the light propagation process. This sheds new light on coherent light generation via four-wave mixing which so far has only been associated with bare states atomic coherences rather than population inversion in the dressed states basis. The other main aim is to point out the exact parameter regime when the inversion of dressed states occurs and also when it gives rise to strong population transfer between sufficiently closely spaced dressed states and thus exponential gain. Regarding the structure of the paper we first derive in succession the conditions for the existence of dressed anticrossings, for dressed population transfer, and for the maximum in dressed coherence necessary for the startup of the generated high-frequency field; then we proceed by evaluating the amount of dressed population inversion and finally its link to exponential gain.

We consider resonant four-wave mixing in a three-level system a-b-c as sketched in Fig. 1. The Hamiltonian in the interaction picture H describing the interaction of this three-level atom with three laser fields with frequencies ω_p, ω_d, and ω_l, Rabi frequencies Ω_p, Ω_d, and Ω_l, and detunings

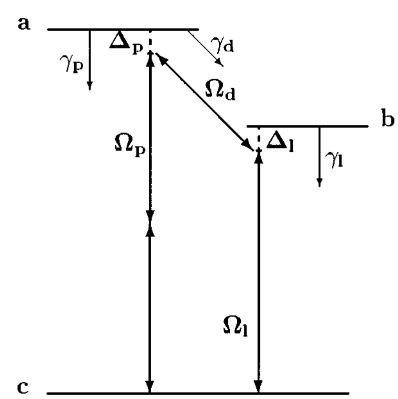

FIG. 1. The three-level atomic configuration of interest. The transitions a-c, a-b, and b-c interact with coherent fields with frequencies ω_p, ω_d, and ω_l, Rabi frequencies Ω_p, Ω_d, and Ω_l, and detunings $\Delta_p=\Delta_d+\Delta_l$, Δ_d, and Δ_l, respectively, with the corresponding spontaneous emission rates γ_p, γ_d, and γ_l. One of the transitions is of two-photon nature (chosen here a-c) and amplification via four-wave mixing is considered for one of the fields (chosen here Ω_l) as a function of all other fields and atomic parameters.

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$\Delta_p=\omega_{ac}-2\omega_p$, $\Delta_l=\omega_{bc}-\omega_l$, and $\Delta_d=\omega_{ab}-\omega_d$ can be cast in the form

$$H=\hbar[\Delta_p|a\rangle\langle a|+\Delta_l|b\rangle\langle b|+(\Omega_p|a\rangle\langle c|+\text{c.c.})+(\Omega_l|b\rangle\langle c|+\text{c.c.})+(\Omega_d|a\rangle\langle b|+\text{c.c.})]. \tag{1}$$

Here energy conservation $\Delta_p=\Delta_d+\Delta_l$ has been assumed so that the above Hamiltonian becomes time independent and the energy of state $|c\rangle$ has been set equal to zero as the reference energy. Since not all three transitions can be dipole allowed we here choose the a-c transition to be of two-photon nature, so that ω_p is selected such that two photons are necessary to be close to resonance with the corresponding transition. Therefore in spite of only three distinct fields present, including the generated field, one may speak of *four*-wave mixing. The a-b and a-c transitions are considered to be driven with fairly large one-photon and two-photon Rabi frequencies Ω_d and Ω_p, respectively, while the coherent field generation and resonant amplification of a weak field with Rabi frequency Ω_l via four-wave mixing shall be investigated on the b-c transition. This particular setup has been chosen to match the situation in krypton [7], of which the particular relaxation rates will be assumed later on, but for most of the paper the respective level position may be interchanged and also the choice of the two-photon transition will not influence the main conclusions here. The two strong fields give rise to a coherence ρ_{bc} which leads to coherent field generation around the b-c transition. Once a small amount of this field is generated, absorption due to the corresponding linear susceptibility $\chi^{(1)}$ may lead to saturation. If, e.g., quantum interference can render this absorption small, saturation at a high intensity is possible [4,7] and if we have a $\chi^{(1)}$ corresponding to gain, exponential gain via the four-wave mixing process appears feasible.

We here aim to obtain conditions for most efficient high-frequency generation and to gain a more intuitive understanding of four-wave mixing in the basis of the eigenstates of the Hamiltonian Eq. (1), the dressed states. The eigenvalues of the Hamiltonian Eq. (1) obey the third order polynomial equation

$$\lambda^3-\Delta\lambda^2-A\lambda+C=0, \tag{2}$$

with

$$\Delta=\Delta_p+\Delta_l,$$

$$A=|\Omega_d|^2+|\Omega_p|^2+|\Omega_l|^2-\Delta_p\Delta_l,$$

$$C=\Delta_l|\Omega_p|^2+\Delta_p|\Omega_l|^2-\Omega_l\Omega_d\Omega_p^*-\Omega_p\Omega_l^*\Omega_d^*, \tag{3}$$

with $\hbar$ set equal to unity. This equation has generally no simple exact analytic solution. However, a computational solution is straightforward. In Fig. 2 the eigenstates corresponding to the three solutions of the above Eq. (3) are displayed. Apparently the plotted dressed states strongly depend on the detuning Δ_l of the generated field to the bare transition b-c because the detuning of the pump field on the a-c transition is changed at the same time to ensure the above mentioned energy conservation $\Delta_p=\Delta_d+\Delta_l$ (while the field with the leading Rabi frequency Ω_d is kept on resonance).

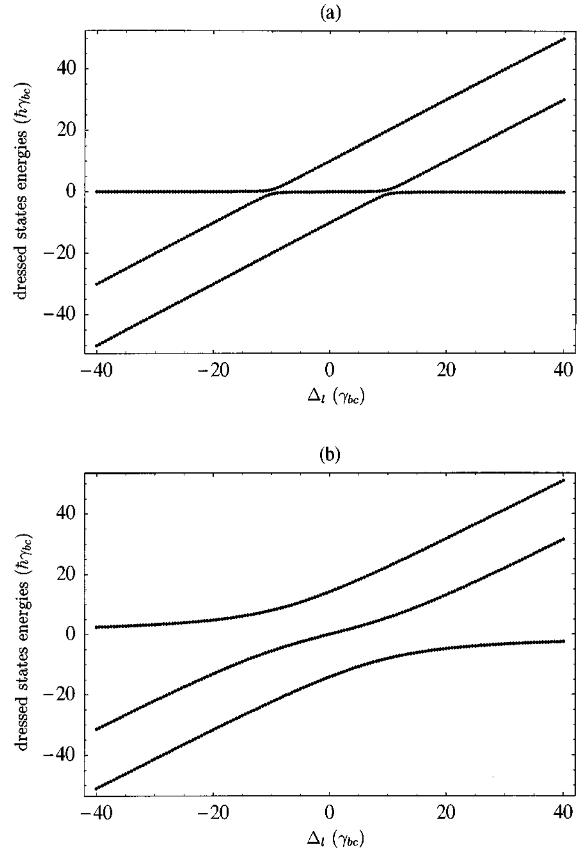

FIG. 2. The three dressed states energies of a driven three-level system (a-b-c from top to bottom) as a function of the generated field detuning $\Delta_l=\omega_{bc}-\omega_l$. The two strong laser fields on the driving and pump transitions a-b and a-c are assumed to be $\Omega_d=10\gamma_{bc}$ and $\Omega_p=1\gamma_{bc}$ (a) [$\Omega_p=10\gamma_{bc}$ (b)], respectively; $\Omega_l=0$. The relaxation rate of the coherence on the b-c transition γ_{bc} is assumed to be the arbitrary scaling parameter and relaxation rate with the largest order of magnitude. The strong driving laser on the a-b transition is assumed resonant and the detunings of the pump and generated fields are assumed equal to ensure energy conservation. We note that substantial population transfer at the dressed avoided crossing $\Delta_l\approx\pm10\gamma_{bc}$ is possible in (a) where the separation is not large as compared to the relaxation rate with the highest order of magnitude (here γ_{bc}).

We find the existence of avoided crossings at about $\pm$ the leading Rabi frequency [in Fig. 2(a) $\pm10\gamma_{bc}$]. This feature disappears, i.e., the closest distance between the dressed states increases strongly, when the weak field Ω_p becomes comparable to Ω_d as displayed in Fig. 2(b) or when the weak field is kept resonant and the strong field is detuned. In line with these findings, closely avoided crossings also occur, when the field associated with Ω_p is strong and kept on resonance while Ω_d is weak and the corresponding frequency detuned by at about $\pm\Omega_p$. We stress that the feedback of the small generated field on the eigenstates has been neglected compared to that of the two stronger fields with Rabi frequency Ω_p and Ω_d.

We can confirm the findings of Fig. 2 analytically and obtain the conditions for most efficient four-wave mixing within an approximate approach, which we will now perform for the situation displayed in Fig. 2(a) with the strong resonant field being on the a-b transition, i.e., $\Delta_d=0;\Delta_p=\Delta_l$;

$\Omega_d \gg \Omega_p \gg \Omega_l$. We note from Fig. 2(a) and Eq. (2) that there is a solution close to zero under those conditions and that this becomes most inaccurate when $A \approx 0$, i.e., we have the anticrossings positioned at $\Delta_l = \overline{\Delta_l}$ with

$$\overline{\Delta_l} = \pm \sqrt{|\Omega_d|^2 + |\Omega_p|^2 + |\Omega_l|^2}. \tag{4}$$

On neglecting the term λ^3 in Eq. (2) at $A \approx 0$, the two solutions $\lambda_\pm$ of the quadratic equation can easily be determined to give

$$\lambda_\pm \approx \pm \sqrt{\frac{C}{2\overline{\Delta_l}}} \approx \pm \frac{|\Omega_p|}{\sqrt{2}}. \tag{5}$$

Thus we find that the separation of the two dressed states at the avoided crossings is of the order of $|\Omega_p|$, which stands for the smaller of the two driving fields, where anywhere else it is of the order of the largest Rabi frequency.

This allows us also to determine the two eigenvectors $|\pm\rangle$ corresponding to the eigenenergies $\lambda_\pm$ at the two anticrossings $\overline{\Delta_l}$ [see Eq. (4)], which is positive or negative as to which of the two anticrossings is chosen. The remaining eigenvector associated with the state not involved in the anticrossing $|0\rangle$ must be orthonormal to $|+\rangle$ and $|-\rangle$, yielding

$$|\pm\rangle = \frac{1}{N_\pm}\left[(\Omega_p \mp \sqrt{2}\overline{\Delta_l})|c\rangle + \Omega_d |b\rangle + \left(\pm \frac{\Omega_p}{\sqrt{2}} - \overline{\Delta_l} \right) |a\rangle \right],$$

$$|0\rangle = \frac{1}{N_0}\left[\Omega_p |c\rangle + \frac{(2\overline{\Delta_l}^2 - \Omega_p^2)}{\Omega_d} |b\rangle + 2\overline{\Delta_l}|a\rangle \right], \tag{6}$$

with $N_\pm$ and N_0 being the appropriate normalization constants. We are interested in the case of close anticrossings, i.e., when Ω_p is small as compared both to Ω_d and to the largest relaxation rate, which is our scaling parameter. We note that in this case the state $|0\rangle$ not involved in the anticrossing is essentially a superposition of the two weakly populated excited bare states $|a\rangle$ and $|b\rangle$. Thus with $|0\rangle$ being virtually empty the essential physics is governed by the two anticrossing states. We confirm this later numerically.

We now would like to address the implications of avoided crossings for resonant four-wave mixing efficiency. In the first step we show the possibility of population transfer between inverted dressed states at anticrossings with sufficiently small separation of the participating eigenstates and in the second step its consequences for exponential gain. We are interested in coherent light generation around the frequency ω_l close to the b-c transition. In the chosen semiclassical dressed states picture, which is justified because of the strong driving fields, there are only three dressed states as in Fig. 2. In a fully quantum mechanical picture there would be an infinite set of three levels as those in Fig. 2 which are characterized by the photon numbers of the three involved coherent fields n_d, n_p, n_l, where the indices relate to those of the corresponding Rabi frequencies. We are interested in transitions of dressed states with photon number of the generated field n_l to those sets separated by the energy $\hbar\omega_l$ with photon numbers $n_l - 1$ and $n_l + 1$, corresponding to absorption and amplification. In the semiclassical approximation, one cannot distinguish between n_l and $n_l \pm 1$; therefore the transitions of interest are those of every dressed state to the same state. As a consequence the initial and final state of the transitions of interest are, within the semiclassical approximation, equally populated. We also recall that the coherence among such a dressed state and itself, being responsible for the light generation around the frequency ω_l, is a dressed state population. This cannot be vanishing for all three dressed states and may lead to initial gain. However, since there is naturally no population inversion among identical states, the newly generated field cannot be amplified exponentially (as discussed later in detail). The situation is only different at avoided crossings if their separation is not large compared to the largest relaxation rate Γ of the system. In order to fulfill this, the smaller driving Rabi frequency can be at most

$$\Omega_p \approx \Gamma. \tag{7}$$

Then there could be population transfer between differently populated dressed states, yielding a large coherence among the two involved dressed states and, as will be discussed later on, exponential gain. The largest possible Rabi frequency Ω_p is assumed for the benefit of strongest initial coupling of the two bare states $|b\rangle$ and $|c\rangle$.

The similarity of the system discussed here to a driven two-level system for the parameters at the closely avoided crossings may be surprising because rather than the direct coupling of the two states $|b\rangle$ and $|c\rangle$ we here have a coupling with two coherent driving fields via an intermediate state $|a\rangle$. The crucial similarity is that both excited and ground state ($|b\rangle$ and $|c\rangle$) are involved in the coupling by the strong external driving fields and thus the "dressing." Therefore any coherence created initially between $|b\rangle$ and $|c\rangle$ does not need to be self-generated as in most coherent amplification schemes, but can be induced directly by the external driving fields. This bare states coherence, as well as coherence between dressed states which is induced by the driving fields, may lead then to an initial coherent field generation. In the following we will evaluate precisely this coherence and the amount of population difference of the two relevant dressed states, because this will determine the stage after the initial field generation when exponential gain may occur.

From the given Hamiltonian Eq. (1) and the master equation which governs the dynamics of the density operator ρ of the system we can determine the various density matrix elements in steady state as done in [7]. Then via a unitary transformation

$$\rho' = U \rho U^T \tag{8}$$

we can calculate all dressed matrix elements, where the rows of the unitary 3×3 matrix U are given by the three eigenvectors arising from the eigenvalues in Eq. (2) and the Hamiltonian in Eq. (1). The density operators ρ and ρ' are here considered as 3×3 matrices, in the representation of the bare and dressed eigenstates, respectively.

We are interested in coherent field generation around the transition b-c, which is proportional to the imaginary part of the matrix element $\rho_{bc} = \langle b|\rho|c\rangle$. This is plotted in Fig. 3(a) at the avoided crossing at $\Delta_l = +\sqrt{|\Omega_d|^2 + |\Omega_p|^2 + |\Omega_l|^2}$ as a

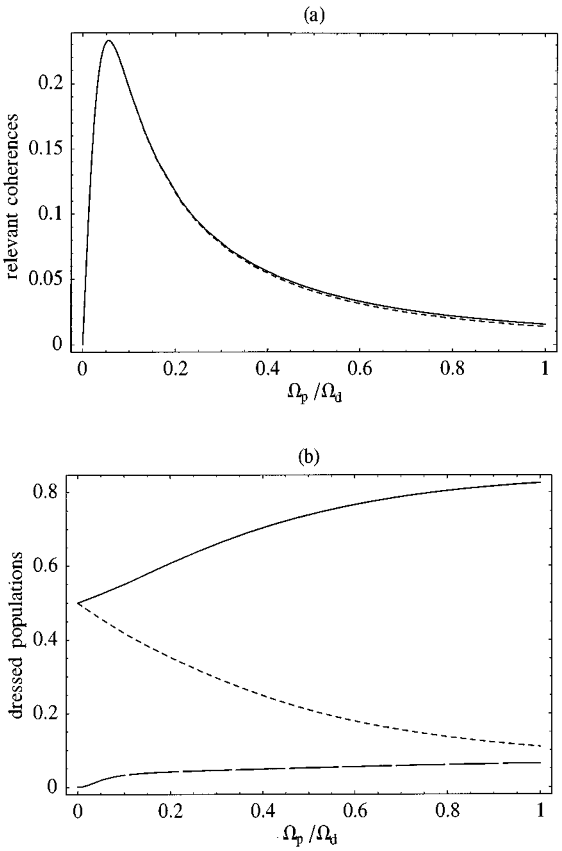

FIG. 3. (a) The imaginary part of the total coherence between the states b and c (dashed line) and its contribution that solely arises from the coherences between the dressed states involved in the anticrossing (solid line) at the resonance frequency of the anticrossing $\Delta_l=+\sqrt{\Omega_p^2+\Omega_d^2+\Omega_l^2}$ in Fig. 2(a). The plot is as a function of the ratio of the two Rabi frequencies Ω_p/Ω_d with $\Omega_d=10\gamma_{bc}$ and $\Omega_l=0$, so it displays the optimal conditions for the initial coherent field generation around the transition b-c. (b) The populations of the three dressed states which the initially generated field senses. The long dashed line represents the state not involved in the anticrossing. All parameters are scaled by the relaxation rate of the coherence on the b-c transition γ_{bc} with the highest order of magnitude and relaxation rates for krypton assumed as given in [7].

function of the ratio of the two applied Rabi frequencies Ω_p and Ω_d, i.e., the dressed state separation as Ω_d is kept constant. The matrix elements of ρ were here determined via the master equation in steady state utilizing the interaction Hamiltonian Eq. (1) and the relaxation rates of krypton as described in Petch *et al.* [7,10]. From the inverse transformation as given in Eq. (8) the bare states matrix elements can then be expanded into their contributions from the various dressed states matrix elements. The solid line in Fig. 3(a) shows the contribution of the dressed coherences between the two anticrossing states to the total imaginary part of ρ_{bc}. Because of the similarity of both curves, it becomes apparent that the essential physics is described by the coupling of the two anticrossing states. Figure 3(a) is evaluated for a vanishing Rabi frequency $\Omega_l=0$. As a consequence the maximum of the curves around $\Omega_p\approx 0.05\Omega_d\approx 0.5\gamma_{bc}$ indicates the optimal condition for the startup of the field generation. Thus Ω_p is not larger than the order of magnitude of the largest relaxation rate as required for population transfer at the anticrossings. In fact it is of the same order, confirming our earlier conjecture regarding optimal amplification to assume the largest possible Ω_p that still allows dressed population transfer to occur. Crucial now is the population difference sensed by the newly generated field. This is plotted in Fig. 3(b) (solid and short dashed lines for the two states involved in the anticrossing) and we note that the population difference is significant in the regime $\Omega_p\approx\gamma_{bc}$, when the involved dressed states are still close enough and there is efficient initial field generation.

We emphasize that the Rabi frequency of the generated field Ω_l has been set equal to zero both for the evaluation of the dressed states and the dressed matrix elements in both Figs. 2 and 3 because it has negligible influence in the first stages of buildup of this field. Once the generated field, however, becomes comparable to the two driving fields, the back action of the generated field on its own generation process is nonnegligible, yielding an effect on the dressed population difference, on the rate of exponential gain, and eventually saturation. Figure 3(b) also confirms our earlier statement, that the third dressed state which is not involved in the anticrossing is very weakly populated for a small Ω_p. We also note that for increased off-diagonal decay rates, such as those arising from laser fluctuations, the population difference between the anticrossing states becomes smaller, and the dressed state not involved in the anticrossing more populated. The large gain on the Rabi sidebands predicted in Fig. 13 in [7] can also be viewed as arising from an inversion of dressed states even though it is very small because of the large off-diagonal decay rates assumed in that analysis. Now we address the last step in which the connection between the dressed states population difference and exponential gain at close anticrossings is derived.

In order to consider the problem of exponential or linear gain with saturation we need to address the propagation equation of the generated field given by the Rabi frequency Ω_l as a function of the spatial coordinate x in the laser propagation direction:

$$\frac{\partial}{\partial x}\Omega_l+G\Omega_l=F, \tag{9}$$

with solution for the light intensity I,

$$I\propto\Omega_l^2=\left[\frac{F}{G}(1-e^{-Gx})\right]^2. \tag{10}$$

The particular form of G and F was derived in Petch *et al.* [7]; for the purpose of this article it is only of relevance that F is proportional to the polarization of the atom in zeroth order in the generated laser field and G is proportional to the coefficient of the linear contribution to this polarization ($G=-iK\chi^{(1)}$ with K being positive and real and $\chi^{(1)}$ being the linear susceptibility). As can be seen from Eq. (10), the essential parameter for exponential gain to occur is the real part of the linear contribution G. For the situation of interest here this parameter can be easily estimated: The two dressed states involved in the anticrossing, say $|1\rangle$ and $|2\rangle$ with energy difference $\hbar\omega_{21}$, interact with the field with Rabi frequency Ω_l as created initially due to the term F in Eq. (9). Then the coherence ρ_{21}, which is proportional to the polarization governing the response of the atom, and which may have the relaxation rate γ_{21}, follows the simple equation

$$\dot{\rho}_{21}=-[\gamma_{21}+i(\omega_{21}-\omega)]\rho_{21}+i\Omega_l(\rho_{22}-\rho_{11}), \quad (11)$$

with steady-state solution for the real part of G as a function of the angular frequency ω given by

$$\mathrm{Re}G(\omega)\propto \mathrm{Im}\rho_{21}(\omega)=\frac{\gamma_{21}\Omega_l}{[(\omega_{21}-\omega)^2+\gamma_{21}^2]}(\rho_{22}-\rho_{11}). \quad (12)$$

Thus the sign of ReG is governed by the sign of the population difference of the two essential dressed states and the absolute value depends linearly on the amount of the difference of the two populations. As opposed to a bare two-level system there is no excited and no ground state: transitions can take place from $1\rightarrow 2$ and from $2\rightarrow 1$; one being amplified and the other being damped. Let us assume Ω_l being positive and $|1\rangle$ denoting the higher populated state with consequently gain on $1\rightarrow 2$ and absorption on $2\rightarrow 1$ transition. According to Eq. (10) this means exponential gain on the $1\rightarrow 2$ and exponential damping on the $2\rightarrow 1$ transition. Thus with different dressed populations there is always an inverted dressed transition with the right sign of ReG in Eq. (10) on each of the two sidebands $\Delta_l=\pm\sqrt{|\Omega_d|^2+|\Omega_p|^2+|\Omega_l|^2}$ with the consequence of exponential gain. We emphasize that the gain will build up exponentially till the population difference of the dressed states and their separation becomes affected by the generated field, and this is when the generated field becomes comparable in strength to one of the two driving fields.

In conclusion, we pointed out the conditions and the underlying physics of exponential gain in resonant four-wave mixing. As the main condition the smaller Rabi frequency of the driving fields has to be at most comparable to the highest relaxation rate of the interacting atomic system, such that population transfer can occur at two closely spaced avoided crossings of dressed states. Then the coherence between these two dressed states leads to the generation of a field which senses a resonance with an inverted dressed transition and the field builds up exponentially till it becomes comparable in strength with one of the driving fields.

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[10] For comparison with Petch *et al.* we need to point out the following technical errors in [7] without consequence on the main claims of this paper: an all over factor $\frac{1}{2}$ is missing in the right-hand sides of Eqs. (3) and (4) of [7] to match with Eqs. (8)–(12) and a minus sign in the fifth entry of the lower row of the matrix M in Eq. (16). In Fig. 13 the detuning is scaled in terms of the off-diagonal element γ_{ac}.