Highly efficient inelastic four-wave mixing using dual induced transparency and coherently prepared states

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Abstract

We investigate a lifetime broadened and coherently prepared five-state system for multi-wave mixing processes. We show that very efficient wave mixing occurs, producing an unconventional mixing wave that has the characteristics of both conventional four-wave mixing (FWM) and stimulated hyper-Raman (SHR) emission. In addition, we show interesting multiple simultaneous multi-photon interference effects at large propagation distances and demonstrate more than 10 orders of magnitude suppression of populations of the probe wave terminal state and the near three-photon resonance mixing wave generating state. These new type of multi-photon interference based induced transparency effects, which are critically dependent on two distinctive relaxation processes involving both an external supplied and an internally generated fields, are fundamentally different from the conventional three-state electromagnetically induced transparency effect which does not depend on propagation. As a consequence, both the probe and the wave-mixing field to propagate nearly free of absorption and distortions in a highly dispersive medium.

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1. Introduction

Electromagnetic induced transparency (EIT) has been shown in many recent studies to be a valuable technique in facilitating highly efficient nonlinear processes in optically thick media [1–13]. For instance, it has been predicted that significant enhancements can occur for certain nonlinear optical processes where an EIT type of excitation is a part of the interaction between optical fields and the atomic media. These enhancement effects include, but are not limited to, nonlinear optics at low light intensity [6]. Kerr nonlinearity at the single photon level [10], efficient multi-wave mixing [11], single photon switching [12], and four-wave mixing (FWM) channel opening techniques [13]. The key feature in all these studies is an EIT process in which the one-photon terminal state for an on-resonance optical wave is driven transparent. This feature is particularly important for multi-wave mixing processes since an efficient one-photon resonance enhanced excitation channel is necessary for highly efficient wave mixing processes using optically thick media.

In this paper, we present a comprehensive study of a new type of FWM process and a new type of induced transparency using a five-state ladder scheme. Unlike the conventional FWM scheme, this new type of FWM starts from and terminates at different atomic states that form an initially prepared long-lived superposition of two atomic states. This gives the highly directional FWM field the characteristics of stimulated hyper-Raman (SHR) emission. The second feature of the present work is simultaneous multi-photon destructive interference based induced transparency effects which introduces several previously unnoticed features and effects. We emphasize that the multi-photon interference based induced
transparency effects discussed here is fundamentally different from the conventional three-state $A$ scheme widely used in EIT related works. The latter does not rely on any internally generated field or any differential relaxation processes. Our work is organized as follows. In Section 2, we first present the problem of wave mixing using coherently prepared states. With a generic five-state system, which is capable of encompassing both ladder and double-$A$ type of excitations for efficient wave mixing, we show that a new type of mixing wave can be generated very efficiently. We further discuss several new effects associated with the dual-EIT scheme and give several useful approximations. Section 3 is devoted to the discussion of conditions for effective destructive interferences [14] and simple relations that can accurately predict the degree of suppression of populations of the probe wave terminal state and the mixing wave generating state. In Section 4, we present numerical calculations aimed at validating, and testing the analytical results obtained in the previous sections. Here, we compare full numerical solutions and analytical solutions for both ladder and double-$A$ systems. Finally, in Section 5, a conclusion for our study is presented.

2. Solution of problem and new effects

We consider a life time broadened five-state system that interacts with a pulsed (pulse length $\tau$) probe laser, $E_p$, and two continuous wave (cw) control lasers, $E_{c1}$ and $E_{c2}$, see Fig. 1. We assume that the probe laser is tuned near the $|1\rangle \rightarrow |2\rangle$ resonance, and two cw control fields are sufficiently intense to strongly saturate the $|2\rangle \rightarrow |3\rangle$ and $|3\rangle \rightarrow |4\rangle$ transitions. Therefore, we have $|\Omega_{23}\tau| \gg 1$, $|\Omega_{43}\tau| \gg 1$, $|\Omega_{42}\tau|^2 \gg \max(\gamma_{31}\tau, \gamma_{32}\tau)$, and $|\Omega_{42}\tau|^2 \gg \max(\gamma_{21}\delta_{42}\tau, \gamma_{41}\delta_{42}\tau, |\delta_{42}|)$. Here, $2\Omega_{23} \times 4 = p_{23(34)}E_{c1(2)}/\hbar$ is the Rabi frequency of the transitions with a dipole moment of $p_{23(34)}$. Our objective is to seek the time dependent response of the system under the excitations described above, and to investigate the propagation dynamics of both the probe and generated fields. With appropriately selected laser polarizations and atomic ion core configurations, a new wave at $\omega_m$ will be generated as a result of the allowed transition from state $|4\rangle$ to $|5\rangle$. In the case of an alkali atom, one may start with $F = 2, M_F = -2$ for state $|1\rangle$ and $F = 2, M_F = +2$ for state $|5\rangle$. If the polarizations of the probe and control lasers have the same handedness, we will have $M_F = +1$ for state $|4\rangle$ and $M_F = -1$ for state $|2\rangle$. In this way the ordinary FWM coupling the transition $|4\rangle \rightarrow |1\rangle$ cannot occur and the generated wave will have the opposite sense of circular polarization as all of the lasers. In addition, two-wave mixing coupling the transition $|2\rangle \rightarrow |5\rangle$ cannot occur. This scheme can work for an $s \rightarrow p \rightarrow d \rightarrow p \rightarrow s$ sequence of transitions in an alkali atom. Note that state $|5\rangle$ usually is a different magnetic sub-state of the $F = 2$ hyper fine level of the ground state manifold, therefore without a magnetic field states $|1\rangle$ and $|5\rangle$ are degenerate. The initial condition of the system is one of the key features that distinguishes our scheme from all other EIT related FWM schemes. We assume that the system is in a coherent superposition of long-lived states $|\Psi_0\rangle = A_1e^{-i\omega_1t}|1\rangle + A_2e^{-i\omega_2t+\Delta\kappa t}|5\rangle$ prior to the arrival of the probe and control fields.

The $\Delta\kappa$ appearing in $|\Psi_0\rangle$ depends on the laser scheme used to make the coherent superposition state. With parallel laser beams one would have $\Delta\kappa = (\omega_5 - \omega_1)/c$. This complex $z$ dependent phase factor represents a grating written to the medium because of the difference in energy of the initial and final states in the mixing process. This initial preparation of the superposition state can be achieved with coherent population transfer techniques [15]. It is this initial state preparation and the choice of the FWM ending state $|5\rangle$ that gives the generated field the features of a combination of conventional FWM and conventional stimulated hyper-Raman (SHR) emission. However, unlike the conventional SHR generation, the process here is produced by beams in uni-directional (co-propagating) configuration whereas the conventional SHR is usually bi-directional (sometimes only counter-propagating) and can be produced by pump beams of arbitrary orientation. In addition, after appropriate propagation distance, unlike a conventional SHR, no additional population transfer to the final state is produced. Thus the present generation process has inelastic wave mixing character and will be designated here as inelastic four-wave mixing (IFWM). We will use this term through out the rest of the paper.

To be able to encompass different excitation schemes, we define the atomic state vector as follows:

$$|\Psi(z,t)\rangle = A_1(z,t)e^{-i\omega_1t}|1\rangle + \sum_{k=2}^{5} A_k(z,t)e^{-i(\omega_0+\delta_k)t}e^{i(\omega_0+\delta_k)z/c}|k\rangle,$$

(1)

where $\delta_2 = \omega_2 - \omega_1$, $\delta_3 = \omega_3 - \omega_1$, $\delta_4 = -2\omega_1$, $\delta_5 = 0$, with the signs of the laser angular frequencies in the definition of the $\delta_j$.

Fig. 1. Energy level diagram showing relevant laser couplings used in a ladder system. With appropriate definition of detunings, the resulting equations also hold for other orderings of the five states in energy.
depending on whether the light in question is absorbed (+) or stimulates an emission (−). This definition of the atomic wave function allows a general treatment so that both ladder and double $A$ schemes can be contained naturally in a single framework. We note, however, that a ladder scheme is more useful for frequency up conversion. A double-$A$ scheme usually can only generate a side band to one of the pump laser frequencies.

Substituting Eq. (1) into the time dependent Schrödinger equation we obtain equations of motion for $A_j (j = 2, 3, 4)$

$$\frac{\partial A_2}{\partial t} = i(\delta_2 + i\gamma_2/2)A_2 + i\Omega_{23}A_3 + i\Omega_{21}(0),$$  
$$(2a)$$

$$\frac{\partial A_3}{\partial t} = i(\delta_3 + i\gamma_3/2)A_3 + i\Omega_{32}A_2 + i\Omega_{34}A_4,$$  
$$(2b)$$

$$\frac{\partial A_4}{\partial t} = i(\delta_4 + i\gamma_4/2)A_4 + i\Omega_{45}A_5(0) + i\Omega_{43}A_3,$$  
$$(2c)$$

where $\gamma_j$ is the decay rate of the relevant state. We note that since states $|1\rangle$ and $|5\rangle$ are prepared prior to the arrival of the probe and control fields and because the probe laser is assumed to be very weak, we have assume that $A_1$ and $A_5$ do not change appreciably during the period when the probe and control fields are present. This requires that $|\Omega_{12}| \ll |\Omega_{23}|$ so that most of the population remains in the prepared states $|1\rangle$ and $|5\rangle$ [16]. Therefore, we have taken $A_1 \simeq A_1(0)$ and $A_5 \simeq A_5(0)$ [17]. In addition, we neglect any propagation effects of the two control lasers since there is never any appreciable population in states $|2\rangle$, $|3\rangle$, and $|4\rangle$ to cause significant polarizations at the frequencies of the control lasers.

We now show that the generation of IFWM can be very efficient. Taking parallel beam configuration and introducing $\kappa_{ij} = 2\pi n_0 N |p_j A_i(0)|^2/\hbar c$ where $N$ is the concentration in cm$^{-3}$, the Maxwell’s equations for the probe and IFWM fields in the slowly varying amplitude approximation can be expressed as

$$\frac{\partial \Omega_{(m)}}{\partial z} = i{\kappa_{12}A_2A_3A_3(0)},$$  
$$(3)$$

We solve Eqs. (1) and (2) using the standard Fourier transform method, and obtain

$$A_m(z, \eta) = \frac{S_3A_p(0, \eta)}{2L} e^{i}\Omega_{23}/\hbar c (e^{i}\Omega_{23}/\hbar c - e^{-i}\Omega_{23}/\hbar c),$$  
$$(4a)$$

$$A_p(z, \eta) = A_p(0, \eta) e^{i}\Omega_{23}\hbar c (L - \beta) 2L e^{i}\Omega_{23}/\hbar c + L - \beta 2L e^{-i}\Omega_{23}/\hbar c,$$  
$$(4b)$$

where $L = \sqrt{\beta^2 + S_3S_3}$, $\beta = (K_2 - K_3)/2$, $D = (K_2 + K_3)/2$, $K_2 = -K_{12}A_1(0)$, $W_{c2}$, $K_3 = -K_{34}A_4(0)^2 W_{c1}$, $S_2 = K_{12}A_0(0)A_4(0)\Omega_{24}^2\Omega_{45}^2\Omega_{23}$, $S_3 = K_{54}A_5(0)A_4(0)\Omega_{24}^2\Omega_{45}^2\Omega_{23}$, $W_{c1} = \Omega_{23}^2/\hbar c - D_2A_4$, $W_{c2} = \Omega_{23}^2/\hbar c - D_4A_4$, $D_j = \delta_j + \eta + i\gamma_j/2$ ($j = 2, 3, 4$, $\Delta_0 = H - D_2D_4$, and $H = D_0\Omega_{23}^2 + D_2\Omega_{45}^2$.

In Eqs. (4a) and (4b), $A_m(0, \eta)$ is the Fourier transform of the probe (IFWM) field, $\eta = c\tau$ is dimensionless transform variable, and we have taken that $A_p(0, \eta)$ is known and $A_m(0, \eta) = 0$.

It is interesting to note that if the states $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$ were chosen so that an allowed transition existed between $|1\rangle$ and $|4\rangle$, and there was no state preparation, then ordinary FWM would occur, as treated in Ref. [11]. If one sets $\kappa_{54} \equiv \kappa_{14}$ and in general identifies $|5\rangle$ with $|1\rangle$, then the equations describing IFWM are identical to those for ordinary FWM. Thus, all of the effects that will be described here have their counterparts in ordinary FWM. In the next section we will use Eqs. (4a) and (4b) to demonstrate new effects and introduce useful approximations that provide substantial insight to the physical processes under investigation. As in Ref. [11], the IFWM process starts with a weak probe laser. This is quite different from the pulsed FWM processes given in Ref. [7] where the FWM process starts with two powerful long pulsed lasers. Such a strong pulsed scheme should be viewed as two-wave mixing with prepared states rather than FWM.

We now examine new effects enabled by the dual EIT scheme using Eqs. (4a) and (4b). We first focus on the complex exponents in Eqs. (4a) and (4b) under the conditions where $|\Omega_{23}|^2 \gg |D_3D_4|$ and $|\Omega_{23}|^2 \gg |D_1D_2|$. In this limit, the atomic response exhibits adiabatic behavior, therefore we can expand $(D \pm L)/\Delta_0$ in a power series and keep terms up to $\eta^2$. This is a first-order adiabatic approximation method that includes correctly the first order correction to the usual adiabatic approximation. We take (see Appendix A for expressions of these coefficients)

$$\frac{D + L}{\Delta_0} = a_0 + a_1 \eta + a_2 \eta^2,$$  
$$(5a)$$

$$\frac{D - L}{\Delta_0} = a_0 + a_1 \eta + a_2 \eta^2.$$  
$$(5b)$$

Next, we investigate the $\eta$ dependence of coefficients in Eqs. (4a) and (4b). We note that under the conditions specified the coefficients of the exponential functions in Eqs. (4a) and (4b), i.e., $S_3$, $L$ and $\beta$, are nearly independent of $\eta$. That is,

$$L - \beta \approx \kappa_{12}^2 |\Omega_{23}|^2 |A_1(0)|^2 = M_p,$$  
$$L + \beta \approx \kappa_{34}^2 |\Omega_{23}|^2 |A_5(0)|^2 = M_m,$$  
$$2L \approx M_p + M_m,$$  
$$S_3 = \kappa_{34}^2 \Omega_{45}^2 \Omega_{23}^2 A_4(0)A_1(0).$$

With these approximations, it is readily shown that the second exponential terms in Eqs. (4a) and (4b) always decay faster than the first exponential terms so that at sufficiently large $z$ only the first exponential terms remain, resulting in $A_0/A_p = S_3(L + \beta) \approx A_1(0)\Omega_{23}(S_3A_5(0))\Omega_{23}$ and

$$A_m(z, t_0) = \frac{S_3 e^{i\Delta_0 \eta}}{2L} \Omega_p(0, t - z V_g^{-1}),$$  
$$(6a)$$

$$A_m(z, t_0) = \frac{\kappa_{54} |\Omega_{23}|^2 |A_1(0)|^2 e^{i\Delta_0 \eta}}{2L} \Omega_p(0, t - z V_g^{-1}).$$  
$$(6b)$$

We emphasize that Eqs. (6a) and (6b) are the key results that will lead to multiple simultaneous destructive inter-
ference. Indeed, if we solve Eqs. 2a and 2b, 2c together with Eq. (6a), (6b) using the Fourier transform method, we immediately conclude that within the adiabatic approximation we have $A_2 \approx 0$ and $A_4 \approx 0$. This implies that two three-photon destructive interferences have been simultaneously established deep inside the medium, i.e., $\Omega_p \rightarrow \Omega_m \Omega_2 \Omega_2$ and $\Omega_p \rightarrow \Omega_m \Omega_2 \Omega_2 \rightarrow \Omega_m$, resulting in strong suppression of the population in states $|2\rangle$ and $|4\rangle$. Consequently, the production of the IFWM field saturates.

The accuracy involved in this conclusion depends strongly on $|\Omega_{23}|^2 \gg |D_2 D_3|$ and $|\Omega_{45}|^2 \gg |D_4 D_5|$. However, extensive numerics have shown that even when these inequalities and other requirements for adiabatic behavior are only modestly satisfied, these destructive interferences indeed occur at large $z$ and both $|A_d(z, t)|^2$ and $|A_d(z, t)|^2$ are suppressed by as much as twelve orders of magnitude once the fast decaying terms in Eqs. (4a) and (4b) become negligible. These multi-photon destructive interference based induced transparency effects allow the probe and IFWM fields to propagate with identical group velocity and with negligible absorption and distortion by maintaining a proper ratio of their amplitudes.

We now consider the case with a Gaussian input probe pulse, i.e., $\Omega_p(0, t) = \Omega_p(0, 0) \exp(-2(t/\tau)^2)$, and derive a useful relation that holds reasonably accurately from small to relatively large $z$, even with both exponential terms in Eqs. (4a) and (4b) are kept and all $\eta^2$ terms in Eq. (5) are retained. Substituting Eq. (5) into Eqs. (4a) and (4b) we obtain (defining $B_{\pm} = (L\pm b)/(2L)$ and $C = S_f/(2L)$)

$$\Omega_m(z, t) = C \Omega_p(0, 0) e^{i \omega_p z} \left[ e^{\frac{-\eta^2 B_{\pm} z^2}{1 - 8 s_{\pm} Z}} + e^{\frac{-\eta^2 B_{\pm} z^2}{1 - 8 s_{\pm} Z}} \right] \tag{7a}$$

$$\Omega_p(z, t) = \Omega_p(0, 0) e^{i \omega_p z} \left[ B_+ e^{\frac{-\eta^2 B_{\pm} z^2}{1 - 8 s_{\pm} Z}} + B_- e^{i \omega_p z} e^{\frac{-\eta^2 B_{\pm} z^2}{1 - 8 s_{\pm} Z}} \right]. \tag{7b}$$

The physical interpretation of $a_m$ and $a_p$ can be deduced from Eqs. (7a) and (7b). We first note that $a_{m0}$ and $a_{p0}$ are related to phase factors and absorption coefficients for the two eigenfunctions involved in solving Maxwell’s equations. The expansion coefficients $a_{m1}$ and $a_{p1}$, on the other hand, are related to the reciprocal of the group velocity for the two parts of the IFWM and probe fields. Finally, $a_{m2}$ and $a_{p2}$ are related to the rate at which the two pulse terms in the solutions for the probe and IFWM fields broaden as a function of distance.

Eqs. (7a) and (7b) will now serve as the basis for a discussion of how the ultimate efficiency can be achieved in generating the IFWM field. In this investigation it will be useful to note that if $F_p(0, 0)$ is the initial photon flux of the probe laser and $F_m(z, t)$ is the photon flux of the IFWM field at $(z, t)$, we find

$$F_m(z, t) = \frac{\kappa_{12}}{\kappa_{54}} \left| \Omega_m(z, t) \right|^2$$

$$F_p(0, 0) = \frac{\kappa_{12}}{\kappa_{54}} \left| \Omega_p(0, 0) \right|^2,$$

where $F_m = c E_m^2/(8\pi n_0 w_0)$ with $E_m$ being the IFWM field amplitude. It is readily shown that the amplitudes of the two terms in $A_m$ are largest in absolute value if we choose $\kappa_{54} |\Omega_{23}|^2 |A_2(0)|^2 = \kappa_{12} |\Omega_{45}|^2 |A_4(0)|^2$ (which is equivalent to $M_m = M_p$). This is the condition for getting the largest conversion efficiency. This choice makes $|S_f/(2L)| = \sqrt{\kappa_{54}/\kappa_{12}}/2$ which leads to a smaller $\beta$. Consequently, $L$ dominates the coefficients of the complex exponentials, resulting $B_{\pm} \approx 0$. Using these results it is clearly seen that the two terms in Eq. (7b) can destructively interfere to produce zero probe field. When this occurs, the same flux of photons exists at the IFWM frequency. That is, all probe photons have been converted to IFWM photons, corresponding a 100% photon flux conversion efficiency. To understand this, we define $1/V_p = 1/c + \tau \text{Re}[(\omega_{m0}(\omega_{p0}))]$, where $V_p$ is the group velocity of the first (second) part in the above expressions of the probe and IFWM fields. Under our approximations $\text{Im}(\omega_{m0}z)$, $\text{Im}(\omega_{p0}z)$, as well as $|\delta_{p0}z|$ and $|\delta_{m0}z|$ are all very small, and the relative phase of the two terms is determined by $\text{Re}[(\delta_{m0} - \delta_{p0})z] = (2n + 1)\pi$ ($n$ is integer), constructive interference between the two terms in Eq. (8a) $\Omega_m(z, t)$ is achieved. If $\text{Im}(\delta_{m0}z) \ll 1$, and $\text{Im}(\delta_{p0}z) \ll 1$ for the $z$ value with $n = 0$, then this carefully chosen thickness leads to a near 100% conversion of probe photons to IFWM photons. The key point is to choose the parameters such that in the distance required to accumulate the $\pi$ phase difference the rapidly decaying terms have not decayed appreciably. This requires $\text{Im}(\delta_{m0}) \ll \text{Re}(\delta_{p0})$, a condition that is not always satisfied.

3. Effectiveness of the destructive interference

The purpose of this section is to reveal what determines the effective suppression of the amplitudes of $A_2(z, t)$ and $A_4(z, t)$ by the destructive interference shown in Section 2. To achieve this we derive accurate approximate limits on the completeness of the cancellation effect.

We start with a general case of the solution for the probe and IFWM fields given approximately by (see Eqs. (4a) and (4b), (5), (7a), (7b))

$$\Omega_m(z, t) = C e^{i \omega_{m0} t} \left[ \Omega_p \left( 0, \frac{t - z/V_p}{\tau} \right) - e^{i \omega_{p0} t} \Omega_p \left( 0, \frac{t - z/V_p}{\tau} \right) \right]. \tag{8a}$$

$$\Omega_p(z, t) = B e^{i \omega_p t} \left[ \Omega_p \left( 0, \frac{t - z/V_p}{\tau} \right) + B e^{i \omega_p t} \Omega_p \left( 0, \frac{t - z/V_p}{\tau} \right) \right]. \tag{8b}$$

where $\lambda_{\pm} = (D \pm L)/\lambda_0$, and group velocities, phase, and damping factors can be taken from the above approximations. In obtaining Eqs. (8a) and (8b), we have assumed that the convergence is rapid due to the satisfaction of
the conditions for accurate adiabatic behavior. Therefore, we have neglected the spreading of the pulses due to \(a_{m2}\) and \(a_{p2}\).

At large \(z\), where destructive interference occurs and the probe and IFWM pulses propagate with very little absorption and distortion, it is clear that the only reason that the polarizations at \(\omega_0\) and \(\omega_1\) (these are proportional to \(A_2(z, t)\) and \(A_4(z, t)\), respectively) are not zero is due to any residual absorption and the difference between the actual group velocity and the vacuum speed \(c\). From Eqs. (7a) and (7b) (neglecting \(\delta \mu_{z'z}\) and \(\delta \mu_{mz}\)) we see that at large \(z\) where \(\text{Im}(a_{m0}z) < 1\), \(\Omega_{m}(z, t)\) satisfies

\[
\frac{\partial \Omega_m(z\tau)^{\tau}}{\partial z} + \frac{1}{V_{g\tau}} \frac{\partial \Omega_m(z\tau)^{\tau}}{\partial (t/\tau)} = ia_{p0} \Omega_m(z\tau)^{\tau}.
\]

However, \(\Omega_m(z, t)\) must also satisfy Eq. (3), which we obtain for large \(z\)

\[
A_4(z, t) = \frac{ia_{p1}}{A_{s1}(0)} \frac{\partial \Omega_m(z\tau)^{\tau}}{\partial (t/\tau)} + a_{p0} \Omega_m(z\tau)^{\tau}.
\]

If we evaluate \(A_2\) and \(A_4\) at the time, \(t_0\), at which \(\Omega_m(0, t_0/\tau) - t/v_{g\tau}\) is a maximum, we obtain \(\Omega_m(z, t_0) = C \Omega_m(0, 0)\) and \(A_4(z, t) = B \Omega_m(0, 0)\). Since the partial derivatives of \(\Omega_m\) and \(\Omega_p\) at large \(z\) and time \(t_0 = z/v_{g\tau}\) are zero, we have

\[
A_2(z, t_0) = \frac{\kappa_{s1}\tau A_{s1}(0) A_{s1}(0) C}{\kappa_{s1}\tau A_{s1}(0) C} = \frac{\kappa_{s1}\tau A_{s1}(0) A_{s1}(0) C}{\kappa_{s1}\tau A_{s1}(0) C}.
\]

4. Numerical tests of the approximate equations

In this section we compare analytical results obtained in Eqs. (7a) and (7b) with direct numerical evaluation of inverse transforms of Eq. (4a), (4b). We further compare these results with full numerical solutions of 7 simultaneous differential equations for 5 amplitudes \(A_j\) and two fields \(\Omega_m\) without the assumptions of constant \(A_1\) and \(A_3\). This last test is particularly important in validating our analytical treatment. We have carried out such tests for all of the examples given in this section using realistic decay rates \(\gamma_1\) and \(\gamma_3\). As we will show that numerical solutions for the probe and IFWM fields differed by only a few percent from the solutions obtained analytically.

4.1. High photon flux conversion efficiency with a ladder system

Here, we assume that the first condition required to optimize the conversion from probe to IFWM photons, i.e., \(M_p = M_m\), is fulfilled. In particular, we choose the following experimentally achievable parameters appropriate to cold alkali vapors: \(|\Omega_{2s\tau}| = 1000\), \(\tau = 10\ \mu s\), \(\Omega_{4z\tau} = 100\), \(\kappa_{12\tau} = 100,000/cm\), \(\kappa_{54\tau} = 1000/cm\), \(\gamma_2 = 625\), \(\gamma_4 = 4\), \(\gamma_1 = 4\), \(\gamma_3 = 0\), and \(A_1(0) = A_5(0) = 1/\sqrt{2}\). In Appendix B, Table B1 shows the dependence of \(a_{mi}\) on \(\delta\tau\). Table B2 shows how well the approximate formulas for these quantities work by comparing results obtained from Eqs. (A.1) and (A.2) with numerically evaluated coefficients based on the method described in Section 2.

To achieve high efficiency we choose the thickness of the sample so that the two terms in Eq. (7a) interfere constructively. This requires that \(\text{Re}(\delta a_{m0} - \delta a_{p0}) = \pm \pi\) (for the smallest \(z\)) to avoid the rapid attenuation of the second term and the growing peak separation of the two terms due to differences in group velocities. Even after taking these points into consideration, one must choose \(\delta\tau\) large enough so that the real part of \(a_{m0}\) is much larger than the imaginary part. This means that when \(z\) is large enough to give \(\text{Re}(\delta a_{m0}) = \pi\), we still have \(\text{Im}(a_{m0}) < 1\), and the attenuation of this part of the IFWM field is not substantial. For \(\delta\tau < 50\) there is significant attenuation by the time \(z\) is large enough to produce the first constructive interference. This means that the conversion efficiency will be considerably less than unity. However, if we choose \(\delta\tau = 150\) we get an efficiency greater than 80% for a medium length of \(z = 0.4704\) cm (6.678z = \(\pi\)). Fig. 2 shows a surface plot of \(\sqrt{\kappa_{12}/\kappa_{54}}|\Omega_m(z, t)|/\Omega_{p0}\) (left panel) and \(|\Omega_m(z, t)|/\Omega_{p0}\) (right panel) versus \(t/\tau\) and \(z/d_0\) (scaling constant \(d_0 = 10\) cm) using the parameters described above with \(\delta\tau = 150\). Notice that the probe starts out at unity, while the IFWM field starts at zero but reaches a value of 0.92 at \(z = 0.4704\) cm. At this depth the probe field is close to zero, and almost all probe photons have been converted to IFWM photons. We have generated similar figures using the full numerical solutions of 7 simultaneous equations with the assumption that \(\gamma_1 = \gamma_5 = 350\) Hz. These decay rates is what would be expected for a concentration of \(N = 10^{13}/cm^3\). We have assumed that the prepared state was made about 30 \(\mu s\) before the peak of the probe laser arrived at the entrance to the cell. There is no visible difference between the surface plots based on the full numerical solutions of 7 equations and those shown in Fig. 2. Examination of the actual numbers shows that the largest difference in this plot is less than 1%, demonstrating the accuracy and validity of the analytical solutions obtained.

4.2. Prepared state IFWM in a double \(A\) system

The disadvantage of the ladder arrangement of the first four energy levels is that state \(3\) has a much shorter lifetime. This problem is avoided with a double-\(A\) system \(\{7, 11\}\), where \(3\) is a member of the ground state hyperfine manifold. With a double-\(A\) system, where states \(3\) and \(5\) are two different magnetic sub states of the ground hyperfine manifold, \(\gamma_3 = 10^{-2}\) is more typical. This leads to very small \(\delta a_{p0}\) and a much better reduction to absorption. In addition, smaller \(\gamma_3\) will greatly improve the adiabatic behavior of \(A_2\) and \(A_4\) (see Eq. (9)), and hence more efficient conversion of probe beam to IFWM can be obtained. In fact, the cancellation effects that sup-
press $A_2$ and $A_4$ at large $z$ can be truly spectacular. In the following example we show that the slowly decaying parts of the probe and IFWM fields can penetrate hundreds of cm without appreciable absorption and with almost no pulse spreading.

We first consider an example where the conversion between probe photons and IFWM photons is very efficient. Let $|\Omega_{23}| = 1000$, $|\Omega_{34}| = 100$, $\kappa_{12} = 10^3$ cm$^{-1}$, $\kappa_{54} = 1000$ cm$^{-1}$, $\delta_2 = \delta_3 = 0$, $\delta_{42} = 300$, $\gamma_{24} = 625$, $\tau = 10\mu$s, $\gamma_{34} = 0.02$, and $\gamma_{43} = 4$. With these parameters we obtain $a_{p0} = 0.000044i$, $a_{p1} = 0.0250$, $a_{p2} = 0.0000$, $a_{m0} = -3.3314 + 0.056954i$, $a_{m1} = 0.03617 - 0.001424i$, and $a_{m2} = -0.0002$, respectively. Using Eqs. (7a) and (7b) we find that the absorption of the slowly decaying part is negligible for $z < 100$ cm, and the more rapidly decaying part decays by a factor of $e^{-0.569/2} = e^{-0.0575}$. Thus, after $z = d_0 = 10$ cm the peak value of the rapidly decaying term has decreased to $e^{-0.569/2} = 0.283$, while the slowly decaying term is still 0.5. That is, at $z = 10$ cm the constructive interference should give a peak height of 0.78, while the destructive interferences should give about 0.217. Fig. 3 shows $\sqrt{\kappa_{12}/\kappa_{54}}|\Omega_m(z, z/V_g)/\Omega_{p0}|$ and $|\Omega_p(z, z/V_g)/\Omega_{p0}|$ as functions of $z$ ($V_g$ is the average of the group velocities of the two terms in the solutions).

Our theory predicts that the first constructive interference in the IFWM should occur at $z = \pi/3 \approx 0.943$ cm. In Fig. 3 we see this peak height is about 0.97. If this were the total thickness of the medium, the conversion efficiency from probe to IFWM photons would be 94%. The second constructive interference for IFWM occurs at $z = 2.82$ cm. These predictions are very close to what can be observed in Fig. 3.

In order to obtain a spectacular example of the destructive interference and demonstrate the exceptional multi-photon induced transparency we take $|\Omega_{34}| = 200$ and $\delta_{42} = 10$ with all other parameters being the same as in Fig. 3. With these parameters, $B_1 = 0.2$, $B_2 = 0.8$, $C = 0.04$, $a_{m0} = -80.56 + 116.81i$, $a_{m1} = -2.9843 - 7.8419i$, $a_{m2} = 0.48080 + 0.1155i$, $a_{p0} = 0.000100i$, $a_{p1} = 0.0100$, and $a_{p2} = 0$.> From the values of $a_m$ we already see that the slowly decaying part propagates relatively fast and it can penetrate more than 100 cm without appreciable attenuation or change in pulse width. From the imaginary part of $a_{m0}$ we see that after $z = 0.05$ cm the rapidly
Parameters are the same as in Fig. 3 except the surviving components of the probe and the IFWM fields are largest. Negligible we see that \( |A_2| \) is further suppressed by approximately a factor of \( 10^5 \), while \( |A_4| \) is further suppressed by a factor of approximately \( 10^7 \). The suppression of the populations of these levels, and hence the induced transparency at \( z = 0.1 \) cm is spectacular. We emphasize that it is important to distinguish this new type of induced transparency from the conventional three-state EIT. The latter involves only two single photon channels with externally supplied fields whereas in our case it is the destructive interference between one- and three-photon channels that leads to such a remarkable induced transparency. We point out that in the conventional three-state EIT, a dark state is established immediately after the probe field enters the medium (assuming that the cw control field is already present). This is, however, not the case of the system studied here. In fact, there is no dark state in the medium prior to the establishment of the three-photon destructive interference deep inside the medium. A characteristic propagation distance is required so that the generated wave is strong enough to open the back coupling channel, leading to the three-photon destructive interference. In other words, unlike the conventional EIT scheme, the dark state has a root in the nonlinear wave mixing process. In fact, it is established by the interplay of the one- and three-photon coupling after a characteristic propagation distance. This characteristic propagation distance can be short or long depending on the concentration and operation conditions. As a comparison, the commonly known dark state picture in the conventional EIT does not rely on any propagation effect. In our case, before the characteristic propagation distance at which the fast damping terms becomes negligible, one speaks no dark state or the multi-photon induced transparency. Indeed, the view of changing from state \( |1\) to state \( |3\) by gradually extinguishing the driving field, as indicated in the dark state picture of the conventional EIT scheme, cannot be simply applied in the present problem because of the role of the internally generated field.

It should be pointed out that although examples demonstrated in Section 4.2 has shown that approximations made in the present study work better for double-\(A\) systems than ladder system because of much smaller \( |D_3| \) it is important to realize that ladder systems are more important for applications such as frequency up conversion. This is because that double-\(A\) systems do not have the advantages of being widely tunable and being able to yield wavelengths very different from any laser used in the process.

5. Summary

We have presented a theory for a new type of FWM, inelastic four-wave mixing (IFWM), in which the final state in the process is not the same as the initial state. This process has the characteristics of ordinary FWM and the inelastic feature that is peculiar to the hyper-Raman process. With experimentally achievable parameters we have shown that the conversion of photons between a weak probe field and the IFWM field can be close to 100%. In addition, we have shown that destructive interferences occur in all of the odd-photon resonances. These destructive interferences lead to interesting effects in which, at large \( z \) where the fast decaying terms in the probe and IFWM have been absorbed, both the probe and the IFWM fields can propagate through the highly dispersive medium with almost no absorption or distortion. The destructive interference between three- and one-photon channels are so effective that populations of these states can be reduced by as much as \( 10^{14} \) over the populations that would exist if IFWM field were not present. We have also shown that this new type of induced transparency is very different from the conventional EIT, and we have demonstrated how almost all of the features of these phenomena can be calculated accurately from relatively simple formulas. To validate the analytical treatment presented here we have presented extensive numerical simulations in which 7 differential equations are solved simultaneously with any approximation. These numerical tests have provided strong support to the analytical solutions to the problem.

We note that the ability to achieve close to 100% conversion from weak probe to IFWM field is not dependent on how weak the probe field is although it must be weak enough to not change the populations of states |5⟩ and

![Fig. 4. Plot of the amplitudes of state |2⟩ (|A_4|z, z/V_+/|) and state |4⟩ (|A_4|z, z/V_+/|) for a double-\(A\) system as functions of \( z \) at the time when the surviving components of the probe and IFWM fields are largest. Parameters are the same as in Fig. 3 except \( |D_{44}| = 200 \) and \( \delta_{z} = 10 \). Once the rapidly absorbed parts of the probe and IFWM fields are negligible we see that \( |A_2| \) (the dashed line) is suppressed by about a factor of \( 4 \times 10^5 \), while \( |A_4| \) (the dash-dot line) is suppressed by about a factor of \( 7 \times 10^7 \). At large \( z \) and at the time at which the probe and IFWM fields are a maximum, results from Eq. (9) agree very well with these numerical results.]{252 M.G. Payne et al. / Optics Communications 265 (2006) 246–254}

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\text{Fig. 4. Plot of the amplitudes of state } |2⟩ (|A_4|z, z/V_+/|) \text{ and state } |4⟩ (|A_4|z, z/V_+/|) \text{ for a double-}\(A\) \text{ system as functions of } z \text{ at the time when the surviving components of the probe and IFWM fields are largest. Parameters are the same as in Fig. 3 except } |D_{44}| = 200 \text{ and } \delta_z = 10. \text{ Once the rapidly absorbed parts of the probe and IFWM fields are negligible we see that } |A_2| \text{ (the dashed line) is suppressed by about a factor of } 4 \times 10^5 \text{, while } |A_4| \text{ (the dash-dot line) is suppressed by about a factor of } 7 \times 10^7 \text{. At large } z \text{ and at the time at which the probe and IFWM fields are a maximum, results from Eq. (9) agree very well with these numerical results.}
\]
Table B1  
Numerical coefficients

| $\delta \xi = 0$ | $a_{m0} = -0.000 + 195.1i$ | $a_{m1} = -38.44$ | $a_{m2} = -7.58i$ |
| $\delta \xi = 0$ | $a_{p0} = -0.000 + 0.050i$ | $a_{p1} = 0.025$ | $a_{p2} = 0.000$ |
| $\delta \xi = 10$ | $a_{m0} = -79.17 + 40.63i$ | $a_{m1} = -4.678 - 6.51i$ | $a_{m2} = -0.107 + 0.711i$ |
| $\delta \xi = 10$ | $a_{p0} = -0.00 + 0.050i$ | $a_{p1} = 0.025$ | $a_{p2} = 0.000$ |
| $\delta \xi = 50$ | $a_{m0} = -19.77 + 2.076i$ | $a_{m1} = 0.415 - 0.087i$ | $a_{m2} = -0.0077 + 0.0025i$ |
| $\delta \xi = 50$ | $a_{p0} = -0.00 + 0.050i$ | $a_{p1} = 0.025 + 0.000i$ | $a_{p2} = 0.000$ |
| $\delta \xi = 150$ | $a_{m0} = -6.652 + 0.227i$ | $a_{m1} = 0.00695 - 0.0055i$ | $a_{m2} = 0.000$ |
| $\delta \xi = 150$ | $a_{p0} = -0.00 + 0.050i$ | $a_{p1} = 0.025$ | $a_{p2} = 0.000$ |

Table B2  
Approximate coefficients

| $\delta \xi = 0$ | $a_{m0} = -0.000 + 195.1i$ | $a_{m1} = -38.44$ | $a_{m2} = -7.58i$ |
| $\delta \xi = 0$ | $a_{p0} = -0.000 + 0.050i$ | $a_{p1} = 0.025$ | $a_{p2} = 0.000$ |
| $\delta \xi = 10$ | $a_{m0} = -79.17 + 40.63i$ | $a_{m1} = -4.677 - 6.51i$ | $a_{m2} = -0.107 + 0.711i$ |
| $\delta \xi = 10$ | $a_{p0} = -0.000 + 0.050i$ | $a_{p1} = 0.0250$ | $a_{p2} = 0.000$ |
| $\delta \xi = 50$ | $a_{m0} = -19.77 + 2.078i$ | $a_{m1} = 0.415 - 0.087i$ | $a_{m2} = -0.0077 + 0.0024i$ |
| $\delta \xi = 50$ | $a_{p0} = -0.000 + 0.050i$ | $a_{p1} = 0.025$ | $a_{p2} = 0.000$ |
| $\delta \xi = 150$ | $a_{m0} = -6.657 + 0.227i$ | $a_{m1} = 0.00696 - 0.0051i$ | $a_{m2} = 0.000$ |
| $\delta \xi = 150$ | $a_{p0} = -0.000 + 0.050i$ | $a_{p1} = 0.025$ | $a_{p2} = 0.000$ |

[1] appreciably. Indeed, the present work is readily scalable to a quantum treatment of the probe and IFWM field, yielding Heisenberg equations of motion for the fields and density matrix for atomic response.

We finally note that all the features found for IFWM also apply to the FWM process described in Ref. [11]. We believe a FWM process which starts with a weak probe tuned between [1] and [2], as in Fig. 1, is better suited to cold and dense medium than a ladder scheme that starts with two strong lasers. In the latter scheme the process of stimulated hyper-Raman and parametric FWM out of state [3] could severely complicate the process. With our scheme there is never any appreciable excited state population.

Appendix A

Let $D_{0j} = \delta \xi + i \gamma / 2$ (j = 2, 3, 4) and $H_0 = D_{02}[\Omega_{43}^0 \tau]^2 + D_{04}[\Omega_{23}^0 \tau]^2$, we find

$$a_{\alpha0} = -\frac{M_m^2 + M_p^2}{H_0} + \left(\frac{T_2M_p + T_1M_m}{H_0}\right)D_{03},$$

$$a_{\alpha1} = \left(\frac{M_m^2 + M_p^2}{H_0}\right)[(\Omega_{23}^0 \tau)^2 + (\Omega_{43}^0 \tau)^2] - D_{02}D_{04} + \left(\frac{2T_2M_p + T_1M_m}{M_m^2 + M_p^2}\right)H_0,$$

$$a_{\alpha2} = -\left(\frac{M_m^2 + M_p^2}{H_0}\right)[(\Omega_{23}^0 \tau)^2 + (\Omega_{43}^0 \tau)^2] - D_{02}D_{04} - \left(\frac{2T_2M_p + T_1M_m}{M_m^2 + M_p^2}\right)H_0, \quad (A.1)$$

and

$$a_{\beta0} = \left(\frac{T_2M_m + T_1M_p}{M_m^2 + M_p^2}\right)D_{03},$$

$$a_{\beta1} = \left(\frac{T_1M_p + T_2M_m}{M_m^2 + M_p^2}\right)H_0, \quad a_{\beta2} = 0 \quad (A.2)$$

where we have defined $T_1 = M_mD_{04}/(\Omega_{43}^0 \tau)^2$ and $T_2 = M_pD_{02}/(\Omega_{23}^0 \tau)^2$.

Appendix B

See Tables B1 and B2.

References


With concentrations as high as $10^{14}$/cm$^3$, we must also keep the delay between state preparation and the incidence of the probe pulse small,
and the group velocities must be such that decay of coherence between $|1\rangle$ and $|5\rangle$ during the time of propagation is small. This will typically require that the delay plus the time of propagation be a few tens of microseconds.

[17] In the limit that $A_1$ and $A_5$ are constant during the presence of the probe pulse, Eqs. (2a)-(2c) are equivalent to two sets of equations of motion for the off-diagonal elements of the density matrix $\rho_{12}$ and $\rho_{54}$. Also, within our approximation the diagonal elements of the density matrix $\rho_{jj}$ are all very small. Thus, in this limit the Schrödinger picture approach is equivalent to a density matrix treatment of the problem except for the interpretation of the decay rates.