Inelastic wave mixing and multi-photon
destructive interference based induced transparency
in coherently prepared media

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Abstract

We consider a multi-level medium initially prepared with a coherent superposition of two initial levels that are not
directly dipole-coupled. We study the production of multi-wave mixing in the coherent medium, where the process
begins on one and terminates on the other of the two prepared levels. We find that a new type of wave mixing field
can be efficiently generated in such a system with characteristics of both conventional four-wave mixing and stimulated
hyper-Raman emission. We also show that two multi-photon destructive interferences build up simultaneously, leading
to simultaneous reduction of the attenuation of the pump and wave-mixing fields. As a consequence a pair of ultra slow,
temporally and group-velocity matched pump and wave-mixing fields can be generated. This type of multi-photon
induced transparency and its consequences are qualitatively different from the conventional EIT where the destructive
interference occurs between two single photon channels.

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Electromagnetically induced transparency (EIT) [1] describes the modification, by an external electromagnetic field, of the absorption profile of

an optical field tuned to a strong optical transition. In the well-known three-state EIT process, a control laser results in a destructive interference between two single photon excitation channels, and leads to significant reduction of the absorption of a probe field tuned on a strong one-photon resonance [2]. Recently, this index manipulation
technique has been applied to several nonlinear processes including nonlinear phase shift [3], four-wave mixing (FWM) [4,5], FWM with channel opening technique [6,7], and hyper-Raman generation [8] in optically dense media.

In this Communication, we extend research in this field through investigations of multi-wave mixing in a coherently prepared medium under conditions that may lead to ultra slow wave propagation. Specifically, we show, in a full time-dependent treatment, that in a medium that has been coherently prepared, a new type of wave mixing field can be efficiently generated. This new type of mixing field has both FWM and hyper-Raman character and has never has been reported before. We further show that when an auxiliary pump field is sufficiently intense in a ladder system studied here, two multi-photon destructive interferences [9] evolve simultaneously, resulting in simultaneous suppression of the residual excitations of a dark state and the upper state associated with the wave-mixing field. As a consequence of these multi-photon destructive interferences a pair of well matched [10] ultra slow pulses can be produced even if their frequencies are quite different. This is qualitatively different from the well-known three-state EIT process where the destructive interference occurs between two single photon channels.

We consider a lifetime broadened five-level optically dense atomic medium that interacts with three laser fields (Fig. 1). Here, states $|0\rangle$, $|2\rangle$, and $|3\rangle$ together with a pump field ($\Omega_p$) and a continuous wave (cw) control field ($\Omega_c$) form a usual three-state ladder system where induced transparency at the pump frequency ($\omega_p$) can be achieved with sufficiently intense $\Omega_c$. Indeed, with appropriate control laser intensity this process can significantly reduce the absorption of a pump wave tuned to $|0\rangle \rightarrow |2\rangle$ transition, opening a possible efficient up conversion channel [11]. When a cw auxiliary pump field ($\Omega_a$, coupling states $|3\rangle$ and $|4\rangle$) is introduced for wave mixing, the induced transparency by $\Omega_c$ is significantly altered and a four-wave-mixing (FWM) field is efficiently generated.

In the present study, we choose laser polarizations and atomic state configuration so that a wave-mixing field is generated via the transition $|4\rangle \rightarrow |1\rangle$ [12]. We note that one of the key feature of our work is that the medium is “prepared” by first establishing a coherence between levels $|1\rangle$ and $|0\rangle$. This leads to very different wave mixing dynamics as compared to four-wave mixing process using double-$A$ schemes [13]. As we will show, if the coherence between states $|0\rangle$ and $|1\rangle$ is maintained during the arrival of the pulsed pump field, the generated wave has every characteristic of a uni-directional, highly coherent FWM field. It is, however, a new type of FWM because the net momentum, energy and population transfers at the end of the excitation cycle is similar to that in a conventional stimulated hyper-Raman (SHR) process. But unlike SHR generation, the process here is only produced by co-propagating source beams in co-propagating uni-directional, group-velocity matched waves, 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

![Fig. 1. Energy level diagram showing relevant laser couplings.](image-url)
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 start with the atomic equations of motion in Schrödinger picture and the wave equations for the pump and IFWM fields:

\[
\frac{\partial A_2}{\partial t} = -\gamma_2 A_2 + i\Omega_p A_0 + i\Omega_c A_3, \tag{1a}
\]

\[
\frac{\partial A_3}{\partial t} = -\gamma_3 A_3 + i\Omega_c^* A_2 + i\Omega_a A_4, \tag{1b}
\]

\[
\frac{\partial A_4}{\partial t} = i(\delta_m + i\gamma_4) A_4 + i\Omega_m^* A_1 + i\Omega_a A_3, \tag{1c}
\]

\[
\left( \frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) \Omega_{p(m)} = i\kappa_0(14)A_{2(4)}A_{0(1)}. \tag{1d}
\]

Here, \(2\Omega_\beta (\beta = p,c,a,m)\) is the Rabi frequency of the respective excitation field and \(\gamma_j (j = 2,3,4)\) is the decay rate of the state \(|j\rangle\) (see Fig. 1 for definition of detunings). \(\kappa_{ji} = 2\pi N|\omega_\beta D_{ji}|^2/(hc)\) with \(N\) and \(D_{ij}\) being the concentration and dipole moment for the transition \(|i\rangle \rightarrow |j\rangle\).

Our calculation starts with the assumption that the system is coherently prepared so that a prescribed amount of population \((0.1 \leq P_1 \leq 0.5)\) has been coherently transferred from state \(|0\rangle\) to the state \(|1\rangle\) [14,15]. This can be accomplished with coherent population transfer techniques [16] frequently used in atomic initial state preparations. Our objective is to seek the system response to cw laser fields \(\Omega_c\) and \(\Omega_a\), and the pulsed pump field \(\Omega_p\) applied immediately after the initial states are prepared. With the restriction of \(|\Omega_p| < |\Omega_c|\), depletions of states \(|0\rangle\) and \(|1\rangle\) from the initial values are negligible and \(A_0\) and \(A_1\) can be treated as constants in the following calculations [17] and the equations of motion for \(A_0\) and \(A_1\) have been neglected in Eq. (1). Since the mixing does not form a closed loop, circularly polarized light can be utilized. An alkali atom with \(F = 2, M_F = -2\) as state \(|0\rangle\) and \(F = 2, M_F = +2\) as \(|1\rangle\) in an \(s \rightarrow p \rightarrow d \rightarrow p \rightarrow s\) type of sequence is a practical system with existing lasers. With zero magnetic field levels \(|0\rangle\) and \(|1\rangle\) are degenerate in energy.

With these assumptions we now take the Fourier transform of Eq. (1) and obtain

\[
\begin{align*}
x_2 &= -A_0 \frac{D_p A_p^* + A_1 \Omega_p \Omega_a}{D_m A_m^*}, \tag{2a}
ex_3 &= -A_0 \frac{\Omega_c^*(\omega + \delta_m + i\gamma_4) A_p^* - A_1}{D} \times \frac{\Omega_a (\omega + i\gamma_2)}{D} A_m^*, \tag{2b}
ex_4 &= A_0 \frac{\Omega_c \Omega_a^*}{D} A_p - A_1 \frac{D_c}{D} A_m^*, \tag{2c}
\end{align*}
\]

\[
\left( \frac{\partial}{\partial z} - \frac{i\omega}{c} \right) A_{p(m)}^* = i\kappa_0(14)A_{2(4)}A_{0(1)}^*. \tag{2d}
\]

where \(D_a = |\Omega_a|^2 - (\omega + i\gamma_3)(\omega + \delta_m + i\gamma_4),\)
\(D_c = |\Omega_c|^2 - (\omega + i\gamma_2)(\omega + i\gamma_3),\)
\(D = |\Omega_a|^2 + D_a^2 + D_c^2 (\omega + \delta_m + i\gamma_4),\)
and \(\gamma_j (j = 2,3,4)\) and \(A_{p(m)}^*\) are Fourier transforms of \(A_p^* (j = 2,3,4)\) and \(\Omega_{p(m)}^*\), respectively. Solving Eqs. (2d), we obtain

\[
A_p^*(z,\omega) = A_p^*(0,\omega) \times \left[ \frac{\left[ (r_+ - i\omega/c - Q_2) e^{i\omega z} - (r_- - i\omega/c - Q_2) e^{-i\omega z} \right]}{r_+ - r_-} \right], \tag{3a}
\]

\[
A_m^*(z,\omega) = A_p^*(0,\omega) S_2 \left( e^{i\omega z} - e^{-i\omega z} \right) \frac{r_+ + r_-}{r_+ - r_-}, \tag{3b}
\]

\[
r_\pm = \mp i \frac{\omega}{c} + S_1 + Q_2 \pm \sqrt{(S_1 - Q_2)^2 + 4Q_1 S_2}, \tag{3c}
\]

where \(S_1 = -i\kappa_0 |\Omega_a|^2 D_a / D,\)
\(S_2 = i\kappa_{14} A_p^* A_0 \Omega_c^* \Omega_a^*/ D,\)
\(Q_1 = i\kappa_0 A_0^* A_1 \Omega_c \Omega_a / D,\)
\(Q_2 = -i\kappa_{14} A_1^* D_c / D.\)

We note that the only assumptions and approximations made in obtaining Eqs. (3a) and (3b) are slowly-varying-amplitude and plane-wave approximations for the wave equations, and the assumption of persisting coherence between states \(|0\rangle\) and \(|1\rangle\) during the propagation of the pulsed pump and IFWM fields.

In general, the inverse transform of Eqs. (3a) and (3b) cannot be evaluated analytically. However, much of physical insight can be gained by choosing circumstances where \(r_\pm\) can be expanded in a power series of \((S_2/Q_2)/(S_1/Q_1)\). The justification for this type of perturbation treatment is the fact that usually \(\kappa_{12} \gg \kappa_{14}\) for a typical FWM type
of processes. In this same spirit, we also assume that both the cw control and the auxiliary pump fields are sufficiently intense to insure that \(|\Omega_0^2 \gg |(\omega + i\gamma_0)(\omega + \delta_m + i\gamma_4)|, \) and \(|\Omega_2^2 \gg |(\omega + i\gamma_2)(\omega + i\gamma_3)|. \) (This still permits ultra slow propagation, see the numerical example below.) Under these conditions, we have \(S_1, Q_1 \gg S_2, Q_2\) and Eqs. (3a)–(3c) become

\[
A_p^a(z, \omega) = \frac{A_p^a(0, \omega)}{A_0} \times \left[ (S_1^2 - Q_2 S_1 + Q_1 S_2) e^{i\omega z} + Q_1 S_2 e^{i\omega z} \right],
\]

\[
A_m^a(z, \omega) = \frac{A_m^a(0, \omega) S_1 S_2}{A_0} \left( e^{i\omega z} - e^{i\omega z} \right),
\]

\[
r_{+}^{(0)} = \frac{i\omega}{c} + S_1 + \frac{Q_1}{S_1} S_2, \quad r_{-}^{(0)} = \frac{i\omega}{c} + Q_2 - \frac{Q_1}{S_1} S_2,
\]

where \(A_0 = S_1^2 - Q_2 S_1 + 2Q_1 S_2.\) Detailed analysis of Eq. (4c) reveals that \(r_{+}^{(0)}\) has large negative real and imaginary parts in the regime specified. Therefore, after a very short propagation distance, \(z,\) we have

\[
A_p^a(z, \omega) = \frac{S_2 Q_1}{A_0} A_p^a(0, \omega) e^{i\omega z},
\]

\[
A_m^a(z, \omega) = -\frac{S_1 S_2}{A_0} A_p^a(0, \omega) e^{i\omega z}.
\]

These results have profound consequences to the propagation of the pump and IFWM fields. First, Eqs. (5a) and (5b) indicate that at this depth into the medium, where \(r_{+}^{(0)}\) component has decayed away, both the pump and IFWM fields are group-velocity matched and are proportional to each other. Taking the ratio of Eq. (5b) to Eq. (5a), we obtain

\[
\frac{A_m^a}{A_p^a} = -\frac{S_1}{Q_1 - \frac{A_0 D_4}{A_1 \Omega_c \Omega_a} \Omega_0^2 A_1 \Omega_c. \ (6)
\]

Inserting Eq. (6) into Eqs. (2a)–(2c), and carrying out inverse Fourier transform we obtain \(\Omega_c A_3 = -A_0 \Omega_p^a\) and \(\Omega_c A_4 \simeq -A_1 \Omega_m^a.\) When these expressions are inserted into Eqs. (1a) and (1c) we immediately see that at the propagation distance where Eqs. (5a) and (5b) are valid, we have simultaneously achieved \(A_2 = 0\) and \(A_4 = 0.\) Therefore, excitations to states \(|2\) and \(|4\) are simultaneously suppressed. Physically, when deep inside the medium so that Eqs. (5a) and (5b) are valid, two multi-photon destructive interferences are simultaneously established: (1) State \(|2\) is coupled through two pathways: \(|1\rangle \rightarrow |2\rangle\) via \(\Omega_m + \Omega_c^* + \Omega_c\) and \(|0\rangle \rightarrow |2\rangle\) via \(\Omega_p,\) and also (2) level \(|4\) is doubly coupled through \(|0\rangle \rightarrow |4\rangle\) via \(\Omega_p + \Omega_c + \Omega_a\) and \(|1\rangle \rightarrow |4\rangle\) via \(\Omega_m).\) These processes lead to simultaneous suppression of the amplitudes of states \(|2\) and \(|4\) from multi-photon destructive interference through the two pathways that connect each of the two states. This is qualitatively different from the conventional EIT process where the destructive interference and the induced transparency resulting from it occur between two single photon channels. Further analysis and numerical evaluations of the amplitude of state \(|2\) have shown more than four orders of magnitude further reduction when compared to the case where the multi-photon destructive interference is ineffective. Notice that \(\pi \Delta A_0^a\) describes the dispersion properties of the medium at the pump wave frequency. Before the destructive interference is effective, the medium is highly dispersive and also absorptive to both pump and IFWM fields, as can be seen from the expressions of \(\pi \Delta A_0^a\) and \(\pi \Delta A_4^a\) at \(\Omega_m \rightarrow 0\) and non-depleted pump limit (see Eqs. (2a) and (2c)). As the destructive interference rapidly builds in with increasing \(z,\) both \(\pi \Delta A_0^a\) and \(\pi \Delta A_4^a\) are strongly suppressed, and the medium becomes highly transparent to both pump and IFWM field. In the present case, the two multi-photon destructive interferences mediated by the internally generated IFWM field have supplanted the two single photon channels occurring in the conventional EIT, resulting in very strong reduction of the absorption of the pump and generated waves. Indeed, at this depth of the medium, both the pump and IFWM fields have evolved into a pair of temporally and group-velocity matched pulses that propagate free of dispersion or distortion [18]. It is remarkable that the internally generated IFWM field can lead to such an efficient induced transparency effect [19].

We now establish, through extensive numerical calculations, the validity of the analytical treat-
ment presented above using experimentally achievable parameters. In Fig. 2 we have plotted $|z_2(z,w)\rangle/\langle 0,0|z_2(0,w)\rangle$ and $|z_4(z,w)\rangle/\langle 0,0|z_4(0,w)\rangle$ as functions of propagation distance $z$ for $w = \omega \tau = -6.5$. Dash-dotted (dashed) line: $z_2$ ($z_4$) obtained from direct numerical solution of Eqs. (1a)–(1d). Solid circle (diamond): $z_2$ ($z_4$) obtained using approximate solutions of Eqs. (4a)–(4c). Parameters used: $\tau = 10^{-5}$ s, $\Omega_2 \tau = 1000$, $\Delta_{p} \tau = 200$, $\Delta_{m} \tau = 10$, $\gamma_1 \tau = 10$, $\gamma_2 \tau = 624$, $\gamma_3 \tau = 4$, $\gamma_4 \tau = 4$, $\kappa_2 \tau = 10^4$ cm$^{-1}$, $\kappa_4 \tau = 10^4$ cm$^{-1}$, and $|A_0(0)| = |A_1(0)| = 1/\sqrt{2}$. When the two multi-photon destructive interferences become effective both fields propagate dispersion-free with a matched ultra slow propagation velocity of $=10^3$ cm/s.

Fig. 2. Plot of $|z_2(z,w)\rangle/\langle 0,0|z_2(0,w)\rangle$ and $|z_4(z,w)\rangle/\langle 0,0|z_4(0,w)\rangle$ as functions of propagation distance $z$ for $w = \omega \tau = -6.5$. Dash-dotted (dashed) line: $z_2$ ($z_4$) obtained from direct numerical solution of Eqs. (1a)–(1d). Solid circle (diamond): $z_2$ ($z_4$) obtained using approximate solutions of Eqs. (4a)–(4c). Parameters used: $\tau = 10^{-5}$ s, $\Omega_2 \tau = 1000$, $\Delta_{p} \tau = 200$, $\Delta_{m} \tau = 10$, $\gamma_1 \tau = 0$, $\gamma_2 \tau = 624$, $\gamma_3 \tau = 4$, $\gamma_4 \tau = 4$, $\kappa_2 \tau = 10^4$ cm$^{-1}$, $\kappa_4 \tau = 10^4$ cm$^{-1}$, and $|A_0(0)| = |A_1(0)| = 1/\sqrt{2}$. When the two multi-photon destructive interferences become effective both fields propagate dispersion-free with a matched ultra slow propagation velocity of $=10^3$ cm/s.

In Fig. 3 we have plotted the coefficients of Eqs. (2a) and (2c) in Fourier space for small and large $z$. In the small $z$ regime where the IFWM is very weak, we neglect $A_0/A_1$ in Eqs. (2a) and (2c). It is seen that the auxiliary field $\Omega_2$ has introduced loss inside the transparency window (Fig. 3(a)) and the IFWM field grows linearly (Fig. 3(b)) [20]. In the large $z$ regime, we use Eq. (6) to replace $A_0/A_1$ in Eqs. (2a)–(2c). We notice that excitations of both states $|2\rangle$ and $|4\rangle$ are strongly suppressed (within our approximation) by multi-photon destructive interference, as predicted. Fig. 4 is a surface plot of $\sqrt{\kappa_2 \kappa_4} |A_m/\Omega_p(0,0)\rangle$ as a function of propagation distance $z$ and

Fig. 3. Surface plot of $|z_2(z,w)\rangle/\langle 0,0|z_2(0,w)\rangle$ and $|z_4(z,w)\rangle/\langle 0,0|z_4(0,w)\rangle$ for small $z$ (panel (a), (b)), where no destructive interference is present, and large $z$ (panel (c), (d)) where destructive interference is effective. Parameters used: $\tau = 10^{-5}$ s, $\gamma_1 \tau \approx 0$, $\gamma_2 \tau = 60$, $\gamma_3 \tau = \gamma_4 \tau = 0.4$, $\Delta_{m} \tau = 1$, $|\Omega_2 \tau| = 20$. These parameters are similar to that of Na and experimentally achievable with typical MOT trapped cold atoms.
dimensionless frequency \( \omega \tau \) for the same set of parameters. As can be seen that the generated field rises sharply and then falls to a saturate value when the multi-photon destructive interference becomes effective. This saturate value can be maintained for a large propagation distance, indicating the medium is highly transparent.

Before concluding the present study, we comment on the issue of decoherence of the prepared states that is the key to the IFWM generation reported here. As has been described before, the present study rests on the assumption of negligible decay of coherence between states \( |0\rangle \) and \( |1\rangle \) during the wave mixing process. For a typical alkali vapor trapped in a magneto-optical trap (to eliminate Doppler broadening), the decoherence time between the two hyperfine levels of the ground state manifold can be as long as few milliseconds. Thus, a pump pulse of a few 10 \( \mu \)s will not sense appreciable decoherence even with ultra slow group velocity. Within the same ground state manifold (such as the \( F = 2 \) manifold) with zero magnetic field (as the hot vapor magneto-optical induction type of setup), much longer decoherence time (long-lived Zeemann coherence) has been demonstrated [21]. For these systems the decoherence of the prepared states does not present a problem for an ultra slow pump pulse of a few 10 \( \mu \)s. When the probe pulse becomes long, i.e., \( \tau \leq 1 \) ms, the issue of decoherence becomes important [22]. As the prepared coherence decays away, the generated field gradually loses its characteristic of highly direction FWM, and exhibits the features of ordinary hyper-Raman radiation which, as described before, will lead to bi-directional or even counter-propagating field.

The multi-photon destructive-interferences-based induced transparencies discussed here may provide a different avenue for achieving lossless propagation. These transparency processes rely on multi-photon destructive interference rather than an interference between two single photon channels as is the case in the conventional EIT. As a consequence, both the pump and the IFWM waves, after a characteristic propagation, can be made to propagate with well matched temporal profiles and ultra slow group velocities, free of dispersion, in a highly resonant medium. It is remarkable that the internally generated IFWM field can provide such efficient suppressions to the loss of the pump field. This method may have potential applications in opto-electronic device design or in certain types of signal processing. The availability of a pair of well matched ultra slow pulses may lead to new research opportunities. As already noted, the wave-mixing field is of a new type, having both hyper-Raman and conventional FWM characteristics. We finally note that effects described in the present study are also anticipated in many other nonlinear optical processes, regardless in fast or slow regimes.

References

It should be noted that this work is a steady-state treatment without prepared states, and only treats conventional SHR.


It can be shown that when the driving field is very weak, the pump field is strongly absorbed, leading to a partially incoherent excitation of state $j_3$, a situation that must be avoided in coherent multi-wave mixing. See also Ref. [6].

Typically the state $|1\rangle$ should be a hyper-fine level of the ground state manifold (such as a Zeeman level, see for example, D. Budker, et al., Phys. Rev. Lett. 81 (1998) 5788,) for long coherence time and small net energy transfer.

We take $\rho_{00} = |A_0|^2 = 1 - P_1$, $\rho_{11} = |A_1|^2 = P_1$, and $\rho_{01} = A_1^* A_0 = e^{i\phi} \sqrt{(1 - P_1)} P_1$ where $\phi$ is a constant phase factor. Choices of $P_1 = 1/2$ and $\gamma_1 > 1$ indicate a system with a maximum and persisted Raman coherence. Such a persistent Raman coherence gives the generated field described here both the characteristics of a conventional FWM and hyper-Raman radiation.

We note that approximations made in obtaining Eqs. (4)–(6) are not necessary conditions for the destructive interference to occur. Extensive numerical calculations have shown that effective three-photon destructive interferences can be established without making any approximation other than neglecting the depletions of states $|0\rangle$ and $|1\rangle$. Experimental and theoretical studies on this and other nonlinear optical processes including hyper-Raman, parametric FWM, etc. using ladder and double-$A$ schemes with prepared states are in progress.

We note that once the conditions leading to Eqs. 5(a) and (b) are satisfied, the resulting multi-photon destructive interferences described here are very robust and immune to power fluctuation of the control and auxiliary fields. In fact, these destructive interferences can be maintained in a broad range of density, propagation distance and driving field strengths.

We note that in the $|\Omega_0| \to 0$ limit, the peak in the center of Fig. 3(a) disappears and we recover the transparency window of a conventional 3-level EIT scheme.
