Matched ultraslow propagation of highly efficient four-wave mixing in a closely cycled double-ladder system

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We present a fully time dependent, adiabatic solution, and steady-state analysis for the ultraslow propagation of the nondegenerate four-wave mixing (NDFWM) signal and the weak probe beam in a closely-cycled double-ladder system. Under appropriate (especially power balance) conditions, the two-mode probe and phase-matched NDFWM pulses, after a characteristic propagation length, evolve into a pair of amplitude and group velocity matched pulses. Double transparency for the probe and NDFWM beams can be achieved owing to an efficient one- and three-photon destructive interference involving the NDFWM beam and its back reaction to the probe beam. The forward and backward configurations are investigated in this double-ladder system, and their efficiencies are calculated and compared.

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I. INTRODUCTION

Efficiencies of nonlinear optical processes in multilevel atomic systems can be greatly enhanced through light-induced atomic coherence [1–5]. It is clear that the Kerr nonlinear coefficient can be greatly modified and enhanced near electromagnetically induced transparency (EIT) [6,7] resonance in three-level atomic systems [8]. In four-level atomic systems, more energy level configurations can be envisioned, and nonlinear optical processes can be optimized by suppressing linear absorption through EIT (destructive interference) and increasing the nonlinear optical coefficient through constructive interferences in three-photon processes [9–11].

In recent years, there have been many experimental demonstrations and theoretical calculations of enhancing different nonlinear optical processes in various four-level atomic systems. One of these interesting nonlinear optical processes is the nondegenerate four-wave mixing (NDFWM), which normally has high efficiency in closely-cycled multilevel systems such as double-Λ [3,4,12], four-level cascade [13], and double-ladder systems. The distinct features of the double-Λ systems are its symmetry in laser frequencies and near degeneracy between the probe beam and the generated signal beam. For a specially arranged laser beam configuration [two strong pump beams share one lower state, but connect to different excited states, as shown in Fig. 1(a)], both the probe beam and the generated signal beam can satisfy EIT condition simultaneously to minimize linear absorptions, and, at the same time, the NDFWM can have high efficiency. Recent studies have predicted 100% NDFWM efficiency in backward double-Λ configuration [14], but the forward NDFWM efficiency can only reach 25%. Since a near Doppler-free condition can be easily satisfied in such double-Λ system, a hot atomic vapor cell can be used for experimental demonstrations of the predicted effects. However, since the generated frequency is similar to the probe beam frequency (in the near degenerate case), no up-converted beam can be generated. On the other hand, recent theoretical studies on cascade four-level ladder system, as shown in Fig. 1(b), predicted high efficiency (75%) in forward NDFWM and generation of up-converted light beam. Due to the large difference in the generated signal frequency from the probe beam, near Doppler-free configuration is impossible to satisfy in this case, so experiments in this configuration cannot be done in atomic vapors.

Another important issue in such nonlinear optical processes is the pulse matching between the weak probe beam $E_1$ and the generated signal beam $E_s$. Since both the probe and signal beams are under EIT conditions, their group

![Figure 1](https://example.com/fig1.png)

**FIG. 1.** The closely-cycled four-level systems: double-Λ (a) and four-level cascade (b) systems, double-ladder systems (c) and (d), where $\delta$ is the controllable detuning factor; The forward (e) and backward (f) NDFWM schemes. The solid arrows, dashed arrows, and dotted-dashed arrows represent the applied laser field, the emitted NDFWM signal, and the back-reaction of NDFWM, respectively.

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velocities are reduced owing to the sharp slope changes accompanying the EIT dips. The high efficiency in NDFWM is due to the slowing down of the group velocities of these two pulses and matching between them in propagation. In previous theoretical studies using an atomic amplitude technique [12–14], a four-state system interacting with long and short laser pulses in a weak probe beam approximation has been investigated. It was shown that with three-photon destructive interference, the conversion efficiency can reach as high as 100% [12]. A pulsed probe field and a pulsed NDFWM field of considerably different frequency can evolve into a pair of matched solitons with the same temporal shape and ultraslow group velocity [13]. The 100% NDFWM efficiency of the backward NDFWM configurations in the four-level system has been analyzed [14].

In this work, we investigate yet another related four-level system, i.e., four-level double-ladder configuration, as shown in Figs. 1(c) and 1(d), which can be easily achieved in rubidium or sodium atoms. This system can have features that combine the advantages of individual system of double-Λ [Fig. 1(a)] and four-level cascade [Fig. 1(b)] configurations, as discussed earlier. For example, if level $3$ is chosen to be near level $1$, the NDFWM efficiency and pulse matching behaviors will behave similarly to the double-Λ configuration (with near degenerate frequencies). However if level $3$ is chosen to be near level $2$, then the NDFWM, as well as pulse matching behaviors, will approach that of the four-level cascade system [Fig. 1(b)].

In comparison to other related four-state models, there are the advantages of double-ladder system shown in NDFWM efficiency, applicability to Doppler-free techniques in atomic vapors, and the ability to generate up-converted light. However the last two criteria typically are mutually exclusive. There are four usual single-photon interference EIT configurations (two $\omega_1+\omega_2$ and $\omega_3+\omega_4$ counterpropagating ladder-types, two copropagating $\omega_1+\omega_2$ $\Lambda$ type and $\omega_1+\omega_4$ $V$ type), and two three-photon interference EIT configurations (three photon $\omega_1+\omega_2-\omega_3$ and $\omega_4$, three photon $\omega_1+\omega_3-\omega_2$ and $\omega_4$). The forward NDFWM scheme [Fig. 1(e)] with a maximum 50% efficiency is good for the Doppler-free configuration requirements of all four typical EIT subsystems [7] and the two three-photon interference EIT subsystems. Specifically, two Doppler-free schemes of three-photon interference EIT are $k_1v-k_2v+k_3v=k_1p$ and $k_4p-k_5v+k_6v=k_1p$ ($v$ is atomic velocity). While the backward NDFWM scheme [Fig. 1(f)] with a maximum 100% efficiency is only good for the Doppler-free configuration requirements of the two ladder-type EITs.

The system parameters for desired objectives in this double-ladder system are calculated and optimized. Also, by considering the effect of “back action” [the NDFWM process due to the generated signal, as indicated in Fig. 1(d)], we will analyze the interplay between the NDFWM and cross-phase modulation (XPM) conditions to better understand the underlying mechanism to achieve pulse matching between the probe beam and the signal beam. Finally, we give a fully time-dependent adiabatic solution and steady-state density-matrix analysis of the forward and backward NDFWM schemes in an ultraslow propagation regime. We present the analytical expressions of a pulsed probe laser, NDFWM-generated pulse, competitions from different linear and nonlinear contributions, ultraslow group velocities, and high NDFWM efficiencies as well. The matched ultraslow propagations of the probe and NDFWM pulses are linked to back-and-forth population transfer and coherent coupling in this double-ladder EIT system. The adequate balance condition is remarkably important for the probe and NDFWM matched propagation. A larger coupling field is good for NDFWM generation, while a smaller coupling field is good for NDFWM ultraslow propagation. On the other hand, a larger pump field is good for probe conversion, while a smaller pump field is good for probe ultraslow propagation. Two destructive interferences via one- and three-photon excitation pathways connecting the ground and terminal states strongly compete with each other, leading to simultaneous attenuation reductions of the probe and NDFWM pulses in this double-ladder system. After a characteristic propagation distance, the nonlinear absorption and dispersion contributions of the cross-Kerr terms and the NDFWM terms all cancel out each other. As a consequence a pair of ultraslow, temporally, and group-velocity matched probe and NDFWM pulses is generated. This unique type of one- and three-photon induced transparency and its consequences are qualitatively different from the standard EIT where the destructive interference occurs between two single photon transition channels.

This paper is organized as follows. Section II presents the basic theory for a closely-cycled double-ladder system. We present a fully time-dependent, adiabatic solution (Sec. III) and steady-state analysis (Sec. IV) for the ultraslow propagation of the NDFWM signal and the probe beam in forward and backward schemes. Finally, Sec. V gives the discussion and conclusion.

II. BASIC THEORY

The closed-loop NDFWM can be greatly enhanced by induced atomic coherences involving two destructive interferences via one- and three-photon pathways (Fig. 1). The nonlinear effects are particularly strong when four-photon closed-loop paths with resonant energy levels are possible [3,4,11–14]. Here, we consider the closed-loop NDFWM in a double-ladder type system, as shown in Figs. 1(c) and 1(d), where lifetime broadened four-state atoms interact with two continuous wave (CW) coupling ($\omega_1$) and pump ($\omega_4$) fields. When a weak probe ($\omega_3$) laser pulse is injected into the system, a pulsed NDFWM field ($\omega_2$) can then be generated efficiently. We shall show that this NDFWM field can acquire the same ultraslow group velocity and pulse shape of the probe pulse and that the maximum NDFWM efficiency of forward scheme is greater than 50%. When the generated NDFWM field is sufficiently intense, efficient feedback to the NDFWM generating state becomes important. It is remarkable that the internally generated feedback NDFWM field can provide such efficient suppression to the loss of the probe field (probe EIT) [Fig. 1(d)]. This feedback pathway also leads to competitive multiphoton excitation of the
NDFWM generating state by three supplied and one internally generated field (NDFWM EIT) [Fig. 1(c)]. Physically, the strong competition is destructive in nature, resulting in induced transparencies due to multiphoton destructive interferences that efficiently suppress the amplitudes of the states involved.

In the four-level double-ladder system depicted in Figs. 1(c) and 1(d) weak probe laser (driving the transition |0⟩→|1⟩) with Rabi frequency $G_1$ and a coupling laser (driving the transition |1⟩→|2⟩) with Rabi frequency $G_2$ form a standard ladder-type EIT configuration. A pump laser drives the transition |2⟩→|3⟩ with Rabi frequency $G_3$, and facilitates a closed coherent NDFWM path, $|0⟩\rightarrow|1⟩\rightarrow|2⟩\rightarrow|3⟩\rightarrow|0⟩$, which results in the generation of photons with wave vector $k_f$ at frequency $\omega_f$. The required phase-matching condition is given by $k_f = k_1 + k_3 - k_2$. When the generated NDFWM field is sufficiently intense, efficient feedback [$(0)\rightarrow(3)\rightarrow(2)\rightarrow(1)$] as shown in Fig. 1(d)] becomes important, which forms the second closed coherent NDFWM path and generates photons with wave vector $k_1$ at frequency $\omega_1$. The required feedback phase-matching condition is given by $k_1 = k_2 + k_3 - k_1$. This NDFWM feedback excitation pathway $|0⟩\rightarrow|3⟩\rightarrow|2⟩\rightarrow|1⟩\rightarrow|0⟩$ leads to multiphoton induced transparency of the probe field through one- $(|0⟩\rightarrow|1⟩)$ and three-photon $(|0⟩→|3⟩→|2⟩→|1⟩)$ destructive interference at $|1⟩$. Furthermore, the back-and-forth population transfer and coherent coupling in this system cause the phase-matched coherent NDFWM field to have the same group velocity and pulse shape as that of the ultraslow probe field.

We start with the atomic density-matrix equations of motion. By using transformations of $\rho_{10}(t) = \rho_{10} e^{-i\omega_1 t}$, $\rho_{20}(t) = \rho_{20} e^{-i\omega_2 t}$, $\rho_{30}(t) = \rho_{30} e^{-i\omega_3 t}$, $\rho_{13}(t) = \rho_{13} e^{i\Delta_f t}$, $\rho_{21}(t) = \rho_{21} e^{-i\omega_1 t}$, and $\rho_{23}(t) = \rho_{23} e^{-i\omega_2 t}$, we obtain

$$\frac{\partial \rho_{10}}{\partial t} = -(i\Delta_1 + \Gamma_{10}) \rho_{10} + i G_1 e^{i k_1 r} \rho_{10} + i G_2 e^{-i k_2 r} \rho_{20} - i G_1 e^{i k_1 r} \rho_{11} - i G_2 e^{-i k_2 r} \rho_{13},$$  

$$\frac{\partial \rho_{20}}{\partial t} = -(i\Delta_2 + \Gamma_{20}) \rho_{20} + i G_2 e^{i k_2 r} \rho_{20} + i G_3 e^{i k_3 r} \rho_{30} - i G_2 e^{i k_2 r} \rho_{21} - i G_3 e^{i k_3 r} \rho_{23},$$  

$$\frac{\partial \rho_{30}}{\partial t} = -(i\Delta_3 + \Gamma_{30}) \rho_{30} + i G_3 e^{-i k_3 r} \rho_{30} + i G_4 e^{i k_4 r} \rho_{00} - i G_3 e^{-i k_3 r} \rho_{31} - i G_4 e^{i k_4 r} \rho_{33}.$$  

The Rabi frequencies of the NDFWM field, probe field, coupling field and pump field are $G_1 = \epsilon_1 \mu_1 / \hbar$, $G_2$, $G_3$, $G_4$, and $G_f = \epsilon_f \mu_f / \hbar$, respectively; while the actual laser and NDFWM fields are $E_1 = e^{i k_1 r - i \omega_1 t}$ and $E_f = e^{i k_f r - i \omega_f t}$. The decoherence rate of polarization $\rho_{00}$ is denoted by $\Gamma_{00}$. The dipole moment between states $|i⟩$ and $|j⟩$ is $\mu_{ij}$, where $\mu_{10} = \mu_{01} = \mu_1$, $\mu_{21} = \mu_{12} = \mu_2$, $\mu_{32} = \mu_{23} = \mu_3$, $\mu_{30} = \mu_{03} = \mu_f$. The frequency detuning is $\Delta_f = \Omega_f - \omega_0$, where $\Omega_f$ is the corresponding atomic energy splitting. The closely-cycled condition requires that $\Delta_f = \Delta_1^2 - \Delta_3$, where $\Delta_1^2 = \Delta_1 + \Delta_2$. We also define a controllable detuning factor $\delta = \Omega_f - \Omega_3$.

The task of solving Eqs. (1a)–(1c) starts with the nondepleted ground-state approximation, i.e., $\rho_{00} = 1$ and weak probe and NDFWM field approximations in the four-level double-ladder system. We note that the last two terms in Eqs. (1a)–(1c) are higher-order terms with small quantities $G_1$ and $G_f$ and small atomic state amplitudes. For a small signal treatment, we neglect these higher-order terms, only keep energy conserving terms in the equations of motion. The Eqs. (1a)–(1c) can then be recast into

$$\frac{\partial \rho_{10}}{\partial t} = -(i\Delta_1 + \Gamma_{10}) \rho_{10} + i G_1 e^{i k_1 r} \rho_{00} + i G_2 e^{-i k_2 r} \rho_{20},$$  

$$\frac{\partial \rho_{20}}{\partial t} = -(i\Delta_2 + \Gamma_{20}) \rho_{20} + i G_2 e^{i k_2 r} \rho_{10} + i G_3 e^{i k_3 r} \rho_{30},$$  

$$\frac{\partial \rho_{30}}{\partial t} = -(i\Delta_3 + \Gamma_{30}) \rho_{30} + i G_3 e^{-i k_3 r} \rho_{20} + i G_4 e^{i k_4 r} \rho_{00}.$$  

III. A FULLY TIME DEPENDENT, ADIABATIC TREATMENT FOR THE MATCHED PROBE AND NDFWM PULSES

We assume the standard ladder-type EIT conditions $G_2 \gg G_1$ and $G_3 \gg G_f$ [7], such that depletions of the CW coupling and the pump fields can be neglected. We derive the field propagation equations under the slowly-varying amplitude approximation. For the forward NDFWM scheme, the probe and NDFWM conventional propagation equations are

$$\left( \frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) G_j = \frac{i4 \pi \omega_j M \mu_j^2}{c h} \rho_{00} = i \xi_{0j} \rho_{00},$$  

$$\left( \frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) G_j = \frac{i4 \pi \omega_j M \mu_j^2}{c h} \rho_{30} = i \xi_{30} \rho_{30}.$$  

Here, $\xi_{0j}(30) = 4 \pi \omega_j (1/j) / c h$.

Taking the Fourier transforms of Eqs. (2) and (3), and using the nondepleted ground state approximation (i.e., $\rho_{00} = 1$), we first obtain

$$D_{10} = M_1 Q_1 + \frac{G_1 G_2^*}{M} Q_f,$$  

$$D_{20} = -\frac{N_1 G_2}{M} Q_1 - \frac{N_1 G_3}{M} Q_f,$$  

$$D_{30} = \frac{M_1}{M} Q_f + \frac{G_2 G_3^*}{M} Q_1.$$  

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Here $D_{10}, D_{20}, D_{30}, Q_1$, and $Q_f$ are the Fourier transforms of $ho_{10}, \rho_{20}, \rho_{30}, G_1$, and $G_f$, respectively. $M_1 = M_f = N_2 = N_3 = \frac{G_3}{2}$, $M = N = \frac{G_2}{2}$, $N_1 = N_2 = N_3 - N_1$, $N_2 = N_3 - N_1$, $N_3 = N_1 + N_2$, $d_1 = -\Delta_1 + \Gamma_1$, $d_2 = -\Delta_2 + \Gamma_2$, and $d_3 = -\Delta_3 + \Gamma_3$. We then obtain the following results for the forward probe and NDFWM schemes [Fig. 1(e)] [initial conditions $Q_1(z = 0, \omega)$ and $Q_f(z = 0, \omega) = 0$ at the entrance]

\begin{equation}
\left( \frac{\partial}{\partial z} - \frac{i \omega}{c} \right) Q_1 = i \xi_0 M_1 Q_1 + \frac{G_3 G^*_f}{M} Q_f, \tag{5a}
\end{equation}

\begin{equation}
\left( \frac{\partial}{\partial z} - \frac{i \omega}{c} \right) Q_f = i \xi_0 M_1 Q_f + \frac{G_3 G^*_f}{M} Q_1. \tag{5b}
\end{equation}

This calculation is similar to the one used in Ref. [12] for the double-$\Lambda$ system. For field propagation equations for the forward probe and backward NDFWM configuration [Fig. 1(f)] [initial conditions given $Q_1(z = 0, \omega)$ and $Q_f(z = 1, \omega) = 0$ at the entrance] are as follows:

\begin{equation}
\left( \frac{\partial}{\partial z} + \frac{i \omega}{c} \right) Q_1 = i \xi_0 M_1 Q_1 + \frac{G_3 G^*_f}{M} Q_f, \tag{6a}
\end{equation}

\begin{equation}
\left( \frac{\partial}{\partial z} + i \omega/c \right) Q_f = -i \xi_0 M_1 Q_f + \frac{G_3 G^*_f}{M} Q_1. \tag{6b}
\end{equation}

Where the propagation is through a medium of length $l$. The second terms on the right-hand side of Eqs. (5) and (6) represent the back-and-forth coupling between the probe field and the NDFWM field while the first terms denote both linear and nonlinear absorptions and dispersions of the atomic medium to their respective fields.

**A. Forward NDFWM**

For given $Q_1(z = 0, \omega)$ with $Q_f(z = 0, \omega) = 0$ in the forward NDFWM configuration [Fig. 1(e)], Eqs. (5a) and (5b) can be solved analytically, yielding

\begin{equation}
Q_1(z, \omega) = Q_1(0, \omega) \psi_e^{i k_2 - i k_2} / (\psi_e - \psi_e) \tag{7a}
\end{equation}

\begin{equation}
Q_f(z, \omega) = Q_f(0, \omega) \psi_e^{i k_2 - i k_2} / (\psi_e - \psi_e) \tag{7b}
\end{equation}

where

\begin{equation}
\psi_e = \psi_e(\omega) = \psi_e(0) + o(\omega), \quad k_2 = k_2(0) + k_2' \omega + o(\omega^2)
\end{equation}

This adiabatic regime is physically well believed by neglecting both $o(\omega^2)$ terms in $k_2(0)$ and $o(\omega)$ terms in $\psi_e(\omega)$. This adiabatic approximation is well justified under the condition

\begin{equation}
G_2^2, G_3^2 > max(|d_1|, |d_2|, |d_3|)
\end{equation}

which can be easily satisfied for typical parameters [12,13]. The group velocities $v_g^\pm$ are determined by $1/v_g^\pm = Re(k_2^\mp) = |d_2^\pm|/|\partial k_2^\mp/\partial \omega|$. We denote that $k_2(0) = \beta_e + i \alpha_e$ where $\beta_e$ denote the phase shifts per unit length, and $\alpha_e$ are absorption coefficients for the two propagation modes. It follows that $\beta_e = -\Delta_2 \alpha_e / \Gamma_{20} = -\Delta_2 \xi_0 G_3 / \Gamma_1$, $\beta_2 = -\Delta_2 \xi_0 G_3 / \Gamma_1$, and $\alpha_2 = \Delta_2 \xi_0 G_3 / \Gamma_1$, with $A_1 = \xi_0 G_3 / \Gamma_1$ and $A_2 = \xi_0 G_3 / \Gamma_1$. Under this condition, one of the components in Eqs. (8a) and (8b) decays much faster than the other. Consequently, after a short characteristic propagation distance we have

\begin{equation}
G_f(z, \omega) = G_f(0, \omega) \psi_e^{i k_2 - i k_2} / (\psi_e - \psi_e), \tag{9}
\end{equation}

**A. Forward NDFWM**

For given $Q_1(z = 0, \omega)$ with $Q_f(z = 0, \omega) = 0$ in the backward NDFWM configuration [Fig. 1(f)], Eqs. (6a) and (6b) can be solved analytically, yielding

\begin{equation}
G_1(z, \omega) = G_1(0, \omega) \psi_e^{i k_2 - i k_2} / (\psi_e - \psi_e) \tag{8a}
\end{equation}

\begin{equation}
G_f(z, \omega) = G_f(0, \omega) \psi_e^{i k_2 - i k_2} / (\psi_e - \psi_e) \tag{8b}
\end{equation}

where

\begin{equation}
\eta = e^{-2 \xi_0 G_3 / \Gamma_1} / (1 + \delta) \Omega \xi_0 G_3 / G_2, \quad \text{where} \quad I_{10} = |\xi_0 G_3|^2 / |\xi_0 G_2|^2 \mu_1^2.
\end{equation}

This maximum NDFWM efficiency is achieved $\xi_0 G_3^2 = |\xi_0 G_2|^2$. Then it is straightforward to obtain

\begin{equation}
\eta = e^{-2 \xi_0 G_3 / \Gamma_1} / (1 + \delta) \Omega \xi_0 G_3 / G_2
\end{equation}

Under the balance condition $Q_f/Q_1 = \psi_e = G_3^2/G_2^2$, $\Delta_0 = 0$, and $D_{30} = 0$ are satisfied simultaneously for all $\omega$ [Eqs. (4a) and (4c)]. Two multiphoton destructive interferences occur between the excitation pathways for the states $|1 \rangle$ and $|3 \rangle$ as depicted in Figs. 1(c) and 1(d). The expressions (7) can be recast into

\begin{equation}
Q_1(z, \omega) = Q_1(0, \omega) \psi_e^{i k_2 - i k_2} / (\psi_e - \psi_e).
\end{equation}

Inserting this result into Eq. (4b) and carrying out its inverse Fourier transform, we obtain $G_1 = -G_2 \rho_{20}$ and $G_f = -G_2 \rho_{20}$. When these relations are inserted into Eqs. (2a) and (2c), it is straightforward to see that at the propagation distance where Eq. (9) is satisfied, the efficient simultaneous $\rho_{10} = 0$ and $\rho_{30} = 0$, and then the probe and the NDFWM fields can propagate in the medium without any attenuation or amplification. Physically, this dual EIT mechanism can be understood as follows. When deep inside the
medium (so that $G_1 \approx -G_{20}^2$ and $G_f \approx -G_{30}^2$ are valid), two multiphoton (three photon and one photon) destructive interferences on the [1] and [3] states are simultaneously established: (a) State [3] is excited through two pathways; $$|0\rangle \xrightarrow{w_1} |1\rangle \xrightarrow{w_2} |2\rangle \xrightarrow{w_3} |3\rangle$$ via phase matched $G_1G_2G_3^*$ [Fig. 1(e)] and $|0\rangle \xrightarrow{w_4} |3\rangle \xrightarrow{w_5} |2\rangle \xrightarrow{w_6} |1\rangle$ via $G_1G_2G_3^*$ feedback excitation [Fig. 1(d)] and $|0\rangle \xrightarrow{w_7} |1\rangle$ via probe $G_1$. These processes lead to simultaneous suppressions of the amplitudes of states [1] and [3] from multiphoton destructive interference through the two pathways. This is qualitatively different from the standard EIT process where the destructive interference (the induced transparency) results from two one-photon channels.

Before the destructive interference is effective, the medium is highly dispersive and also absorptive to both probe and NDFWM fields, as can be seen from Eqs. (5) and (6) for the weak NDFWM ($G_1 \approx 0$), nondepleted coupling and pump limit. As the destructive interference rapidly builds up (increasing $G_f$) with increasing distance, both $\rho_{10}$ and $\rho_{30}$ are strongly suppressed, and the medium becomes highly transparent to both probe and NDFWM fields. Indeed, at a certain depth in the medium, the probe and NDFWM pulses will evolve into a pair of temporally and group-velocity matched pulses that propagate free of distortion. It is remarkable that the generated NDFWM field can lead to such an efficiently induced transparency effect.

Based on Eqs. (8a) and (8b), there are two mixed dressed states as follows:

$$G_+(z,t) = G_1(z,t) + G_f(z,t) = [G_1^*(z,t) + G_f^*(z,t)] + [G_1^*(z,t) + G_f^*(z,t)],$$

(10a)

$$G_-(z,t) = G_1(z,t) - G_f(z,t) = [G_1^*(z,t) - G_f^*(z,t)] + [G_1^*(z,t) - G_f^*(z,t)].$$

(10b)

First, due to the absorption coefficient $\alpha_s \gg \alpha_+$, we can have

$$G_+(z,t) = G_1^*(z,t) + G_f^*(z,t) = G_1(0,t - z/v_g) \varphi_+ e^{i\beta_+ z - \alpha_+ c(1 + \varphi_+)}(\varphi_+ + \varphi_-),$$

(11a)

$$G_-(z,t) = G_1^*(z,t) - G_f^*(z,t) = G_1(0,t - z/v_g) \varphi_- e^{i\beta_+ z + \alpha_+ c(1 - \varphi_-)}(\varphi_+ + \varphi_-).$$

(11b)

The maximum NDFWM efficiency is achieved at $\xi_0 = |G_f|^2 \approx \xi_0 |G_1|^2 (\varphi_+ = 1)$. Equation (11) evolve into $G_+(z,t) = 2G_1(0,t - z/v_g)e^{i\beta_+ z - \alpha_+ c}$ and $G_-(z,t) = 0$. Since probe $G_1(z,t)$ and NDFWM $G_f(z,t)$ photons both have EIT, there exists only as one mixed dressed state $G_+(z,t)$ or polariton of the matched probe and NDFWM pulses.

Second, at different conditions of $v_g^+ = v_g^- = v_g$, $|\alpha_+| \ll 1$, $\beta_+ z = \beta_+ z = \pi$, $\Delta_1 = 2\Delta_2 = 3\Delta_3 = 1$, $\varphi_+ \approx \varphi_-$, we can obtain

$$G_1(z,t) \approx 0 \quad \text{and} \quad G_f(z,t) = -2\varphi_+ \varphi_- G_1(0,t-z/v_g)/(\varphi_+ + \varphi_-).$$

(12)

Finally, at yet another situation with $v_g^+ = v_g^- = v_g$, $|\alpha_+| \ll 1$, $\beta_+ z = \beta_+ z = 0$, $\Delta_2/\Gamma_{30} \gg 1$, and $\Delta_3 = 0$, we can obtain $G_1(z,t) = G_1(0,t-z/v_g)$ and $G_f(z,t) = 0$. Then

$$G_+(z,t) = G_-(z,t) = G_1(0,t-z/v_g).$$

(13)

Thus, the perfect EITs (resonant EIT and detuning EIT) of the matched probe and NDFWM pulses are both established with $\Delta_1 = \Delta_2 = \Delta_3 = 0$, i.e., all lasers are detuned exactly to the unperturbed resonances. On the other hand, one can detune the two CW strong lasers from their perspective resonances in order to avoid the multi-photon destructive interferences. In this case the constructive interference between two modes does occur in Eqs. (7) and (8), the NDFWM conversion efficiency can reach close to 100% whenever the difference between $v_g^+$ and $v_g^-$ is small, the absorption factors $|\alpha_+| \ll 1$, the difference of the phase shifts is $\pi$ [i.e., $\beta_+ z = 0$, $\beta_+ z = \pi$ (or $\beta_+ z = \pi$, $\beta_+ z = 0)$], $\Delta_1 = 0$, and $\Delta_3/\Gamma_{30} \gg 1$. Under these conditions the probe efficiency $\eta_p = G_1(z = l)^2/G_1(z = 0)^2$ is close to zero and all probe creation photons are converted to the signal photons through the NDFWM process. This case is actually very similar to the backward scheme that is discussed in the next section. The multi-photon destructive interference of state [3] no longer occurs, the propagating NDFWM field is strongly enhanced by two modes constructive interference, while the multiphoton destructive interference of state [1] may occur in this case. On the contrary, the NDFWM conversion efficiency is close to zero whenever the phase shifts are the same, i.e., $\beta_+ z = \beta_+ z = n\pi$ (n is integer), $\Delta_2/\Gamma_{30} \gg 1$ and $\Delta_3 = 0$.

B. Backward NDFWM

Based on Eqs. (6a) and (6b), the field propagation equations for the forward probe and backward NDFWM scheme can be recast as

$$\frac{\partial Q_1}{\partial z} = i\theta_1 Q_1 + i\theta_2 Q_f,$$

(14a)

$$\frac{\partial Q_f}{\partial z} = i\theta_3 Q_1 + i\theta_4 Q_f,$$

(14b)

where $\theta_1 = \frac{\xi_0 G_1}{M + \frac{w}{c}}$, $\theta_2 = \frac{\xi_0 G_2 G_1^*}{M + \frac{w}{c}}$, $\theta_3 = -\frac{\xi_0 G_1}{M + \frac{w}{c}}$, $\theta_4 = \frac{\xi_0 G_2 G_1^*}{M + \frac{w}{c}}$. For given initial conditions $Q_1(z = 0, \omega) = Q_f(z = l, \omega) = 0$ at the entrance of this backward NDFWM scheme, Eqs. (14a) and (14b) can be solved analytically as

$$Q_1(z, \omega) = Q_1(0, \omega) \left[\psi_+ e^{i\xi_1 z} - \psi_- e^{i\xi_2 z}\right]/(\psi_+ - \psi_-),$$

(15a)

$$Q_f(z, \omega) = Q_f(0, \omega) \left[\psi_+ e^{i\xi_1 z} + \psi_- e^{i\xi_2 z}\right]/(\psi_+ - \psi_-),$$

(15b)
where \( \psi'_s = (k'_s - \theta) / \theta = \psi'_s (0) + o(\omega) \) and \( k'_s = ((\theta_1 + \theta_3) \pm (\theta_1 - \theta_3) i) / 2 = k'_s (0) + k'_s (0) \omega + o(\omega^2) \).

After the inverse Fourier transforms, Eqs. (15a) and (15b) can be recast into
\[
G_1(z,t) = G_1(z,t) + G_1^*(z,t),
\]
\[
G_1(z,t) = G_1^*(z,t) + G_1^*(z,t),
\]
where \( G_1^*(z,t) = \psi'_s (0) e^{i \beta'_s z - a'_s} + G_1 (0,t) \) and \( G_1^*(z,t) = \psi'_s (0) e^{i \beta'_s z - a'_s} + G_1 (0,t) \).

As mentioned in the previous discussions, the NDFWM efficiency in the backward scheme is greater than that in the forward scheme and may reach 100\% under certain conditions. The balance condition may be satisfied throughout the whole medium [14]. The higher efficiency in the backward scheme can be understood from the effect of the two-mode constructive interference.

The backward NDFWM efficiency can be defined as the ratio of the intensities of the generated NDFWM field at its exit \( z=0 \) and the probe field at its entrance \( z=0 \), i.e.,

\[
\eta = I(z=0) / I(z=0) = \left( i(\omega_1 \epsilon_0 / \omega_2 \epsilon_0) G_1 (z=0) / G_1 (z=0) \right)^2 = 1 + \delta / \Omega_1 \left[ \xi_0^2 / (2 \xi_3 \Gamma_{30} + \xi_0 \omega_0) \right] [14].
\]

For a sufficiently high atom density and enough propagation distance, the efficiency is close to 100\%.

**IV. STEADY-STATE ANALYSIS**

After reaching the balance condition of \( G_1(z,t) G_2^* \) through propagation in the medium, we can consider the high efficiency and ultrasonic propagation behaviors of the probe and NDFWM fields in steady state. By setting \( \rho_{10} = \rho_{20} = \rho_{30} = 0 \) in Eqs. (2), we can obtain
\[
\rho_{10} = \left( i(\Delta_2 + \Gamma_{30}) (\Delta_2 + \Gamma_{30}) + |G_3|^2 G_1 - G^* G_2 G_1 \right) e^{i \beta_1 / \gamma_1 / D},
\]
\[
\rho_{30} = \left( i(\Delta_1 + \Gamma_{10}) (\Delta_1 + \Gamma_{10}) + |G_2|^2 G_1 - G^* G_2 G_1 \right) e^{i \beta_1 / \gamma_1 / D},
\]
where \( D = (\Delta_1 + \Gamma_{10}) (\Delta_2 + \Gamma_{20}) (\Delta_1 + \Gamma_{10}) + |G_3|^2 (\Delta_2 + \Gamma_{30}) + |G_2|^2 (\Delta_1 + \Gamma_{10}) \).

The required phase-matching conditions are given by \( k_f = k_1 + k_2 - k_3 \) for NDFWM \( G_1 G_2 G_3 \) and \( k_1 = k_f + k_3 - k_2 \) for feedback NDFWM \( G_1 G_2 G_3 \). Close inspection of Eqs. (17a) and (17b) shows that three key contributions are involved in the propagation characteristics of the probe and NDFWM fields, which are the linear response term, the cross-Kerr nonlinear term, and the phase-matched coherent NDFWM term. These three contributions provide the total linear and nonlinear absorption and dispersion of the atomic medium. When the generated NDFWM is weak enough (i.e., \( G_f \ll |G_f| / |G_2| \)), which is far away from the balance condition, Eqs. (17) can be written as \( \rho_{10} = i(\Delta_2 + \Gamma_{30}) (\Delta_2 + \Gamma_{30}) G_1 + |G_3|^2 G_1 e^{i \beta_1 / \gamma_1 / D} \) and \( \rho_{30} = -i G_1 G_2 G_1 e^{i \beta_1 / \gamma_1 / D} \).

As the fields propagate in the medium, the NDFWM will build up sufficiently, and the ratio \( G_f(z,t) / G_1(z,t) \) becomes a constant. Then it is straightforward to obtain \( \rho_{10} = i G_1 e^{i \beta_1 / \gamma_1 / D} \) (the two nonlinear terms cancel each other). The opposite signs of the cross-Kerr term and the NDFWM term in Eqs. (17a) and (17b) show normal and abnormal nonlinear dispersion, respectively. After a characteristic propagation distance, the nonlinear absorption and dispersion contributions of these two terms exactly cancel each other. As a consequence, a pair of ultraslow, temporally and group-velocity matched probe and NDFWM pulses is generated. Thus, the \( G_f \) and \( G_1 \) fields show self-regulation during their propagation in such four-level double-ladder atomic system.

Since \( e \chi_{10} E_1 = N \mu_{10} \rho_{10} \) and \( e \chi_{10} E_1 = N \mu_{30} \rho_{30} \), we can write down the susceptibilities of the probe and NDFWM fields, respectively. The group velocities of the probe \( (\chi_1) \) and NDFWM \( (\chi_f) \) pulses are given by \( v_{g1} = c / [1 + \omega_1 (\delta \chi_1 / \delta \omega_0) \Delta_{10} / \omega_0] \) and \( v_{gf} = c / [1 + \omega_1 (\delta \chi_f / \delta \omega_0) \Delta_{10} / \omega_0] \). Comparing with the previous work [4], we have considered the additional NDFWM contribution in the probe group velocity, and obtained the NDFWM group velocity directly (Figs. 2–4). Next, we show that, under appropriate conditions, cross-Kerr modulation and NDFWM coupling can precisely balance nonlinear group velocity dispersion in the ultraslow propagation regime, leading to the formation of ultraslow matched probe and NDFWM pulses (i.e., \( v_{g1} \approx v_{gf} \)).

The total real part \( \chi_r \) of the susceptibility includes linear and nonlinear parts (cross-Kerr and NDFWM), i.e., \( \chi_r = \chi_{rr} + \chi_{rn} \) (\( \chi_{rn} = \chi_{Ker} + \chi_{FWM} \)). Figures 2 and 3 show the competitions of these three dispersion slope contributions for the propagation group velocities of the probe and NDFWM pulses. It needs to be mentioned that the NDFWM part always shows normal dispersion at resonance \((1 / [\delta \chi_f / \delta \omega_0]) \Delta_{10} / \omega_0 > 0\) while the cross-Kerr part corresponds to anomalous dispersion in general \((1 / [\delta \chi_K / \delta \omega_0]) \Delta_{10} / \omega_0 < 0\). The total dispersions in Fig. 2 are dominated by the linear contribution, while the total dispersions in Fig. 3 are dominated by the NDFWM part. A negative slope of the total dispersion profile tends to produce subluminal light, while a positive slope produces superluminal light in Figs. 2 and 3. The subluminal and superluminal characteristics of the probe and NDFWM pulses have been shown in Fig. 4. The matched group velocities \( v_{g1} \) and \( v_{gf} \) of the probe and NDFWM pulses exist at \( G_f / \Gamma_{30} = 7 \) and \( G_f / \Gamma_{30} = 1.35 \) (Fig. 4). The probe light prefers \( G_1 \) to have ultraslow propagation (Fig. 2), while the NDFWM ultralow light prefers \( G_f \) (Fig. 3).

The unique dual induced transparency effects are caused by the one- and three-photon destructive interferences between two different excitation pathways connecting \(|0 \rangle \rightarrow |3 \rangle \) and \(|0 \rangle \rightarrow |1 \rangle \) in this four-level double-ladder system (Figs. 1(c) and 1(d)). We can obtain the balance condition \( G_f(z,t) G_1 \approx G_f(z,t) G_2 \) via close inspection of the ratios in Eq. (9)
after a sufficient propagation distance in the medium. With this result it is straightforward to show that $G_1 e^{i k_1 x} + G_2 e^{-i k_3 x} \rho_{20} = 0$, $G_2 e^{i k_2 x} \rho_{10} + G_3 e^{i k_3 x} \rho_{30} = 0$, and $G_5 e^{-i k_3 x} \rho_{20} + G_7 e^{i k_2 x} \rho_{30} = 0$. When these results are used in Eqs. (2) (note $\rho_{30} = 1$), it can be seen that the amplitudes of all three upper atomic states are strongly suppressed $| \rho_{10} |^2 \approx 0$. Physically, when the NDFWM field is sufficiently intense an additional feedback excitation channel to the state $| 3 \rangle$ occurs, i.e., $| 0 \rangle \quad \rightarrow \quad | 3 \rangle \quad \rightarrow \quad | 2 \rangle \quad \rightarrow \quad | 1 \rangle$ via $G_6 G_3 G_2^* NDFWM$ feedback excitation [Fig. 1(d)]. This excitation is $\pi$ out of phase with respect to the excitation $| 0 \rangle \quad \rightarrow \quad | 1 \rangle$ provided by $G_1$, resulting in suppression of state $| 1 \rangle$, as indicated by $G_1 e^{i k_1 x} + G_2 e^{-i k_2 x} \rho_{20} = 0$.

By inserting $G_3^* e^{-i k_3 x} \rho_{20} = -G_7 e^{i k_2 x} \rho_{30}$ into Eq. (2c), we find that at a sufficient depth into the medium where $G_3^* e^{-i k_3 x} \rho_{20} = -G_7 e^{i k_2 x} \rho_{30}$ is valid, $\rho_{30} / \rho_{20} = -[i(\Delta_1 + \Delta_2 - \Delta_3) + \Gamma_{30} / \Gamma_{30}]$. This indicates that further propagation from this point in the medium the two coupling terms in Eq. (2c) interfere destructively, and no further excitation can be made to the NDFWM generating state $| 3 \rangle$. Physically, when the generated NDFWM field is sufficiently intense, the absorption of the generated NDFWM field opens the second excitation pathway to the state $| 3 \rangle$. This excitation is $\pi$ out of phase with respect to the three laser NDFWM excitation $G_1 G_2 G_3$ to the same state, resulting in a unique destructive interference that suppresses further excitation of the state $| 3 \rangle$. Therefore, the production of the phase-matched NDFWM field saturates. When the NDFWM field becomes sufficiently intense, a small detuning $\Delta_3$ will also lead to a strong absorption of this generated wave via the one-photon process. Such

FIG. 2. The probe dispersion versus $\Delta_1$ with different $G_2$, (a) total probe dispersion, (b) linear part, (c) cross-Kerr part, and (d) NDFWM part of the probe dispersion, respectively. The parameters are $\delta \approx 0$, $\Delta_2 = \Delta_3 \approx 0$, $\Gamma_{10} / \Gamma_{20} = 5.4$ MHz, $\Gamma_{20} / \Gamma_{30} = 1.8$ MHz, $\Gamma_{30} / \Gamma_{30} = 5.9$ MHz, $G_j / 2 \pi = 0.15$ MHz, $G_j / 2 \pi = 5$ MHz, $G_j / 2 \pi = 3$ MHz (solid curve), 10 MHz (dashed curve), 25 MHz (dotted curve), and 30 MHz (dash dotted curve).

FIG. 3. The NDFWM field dispersion versus $\Delta_1$ with different $G_j$, (a) total NDFWM dispersion, (b) linear part, (c) cross-Kerr part, and (d) NDFWM part of the NDFWM dispersion, respectively. The parameters are $\delta \approx 0$, $\Delta_1 = \Delta_2 = \Delta_3 \approx 0$, $\Gamma_{10} / \Gamma_{20} = 5.4$ MHz, $\Gamma_{20} / \Gamma_{30} = 1.8$ MHz, $\Gamma_{30} / \Gamma_{30} = 5.9$ MHz, $G_j / 2 \pi = 0.15$ MHz $G_j / 2 \pi = 5$ MHz, $G_j / 2 \pi = 3$ MHz (solid curve), 10 MHz (dashed curve), 25 MHz (dotted curve), and 100 MHz (dash dotted curve).

FIG. 4. The matched group velocities $v_{\chi f}$ (dashed curve) and $v_{\chi f}$ (solid curve) of the probe and NDFWM pulses versus $G_j / \Gamma_{30}$. The parameters are $\delta \approx 0$, $G_j / \Gamma_{30} = 7$, $\Omega_j / \Gamma_{30} = 2.7 \times 10^4$, $\Delta_1 = \Delta_2 = \Delta_3 \approx 0$, $\Gamma_{10} / \Gamma_{30} = 0.002$, $\Gamma_{20} / \Gamma_{30} = 0.7$, and $G_j / \Gamma_{30} = 0.15$. 
which have the initial conditions \(G = 0\) at the entrance curve

\[NDFWM \quad \text{configuration} \quad H_{20851} \quad \text{pump} \quad H_{9003} \quad \text{curve} \quad H_{20849}\]

For given \(G = 0\), the NDFWM conversion efficiency can reach as high as 50%. However, even with the three-photon destructive interference, the field propagation equations of the forward probe and backward NDFWM are

\[\partial G_1/\partial z = i\xi_{10}p_{10} = \xi_{10}(D_2 G_f/D - D_1 G_1/D), \quad (18a)\]

\[\partial G_f/\partial z = i\xi_{30}p_{30} = \xi_{30}(D_2^* G_f/D - S_1 G_1/D), \quad (18b)\]

where \(D_1 = (i\Delta_1^2 + \Gamma_{20})(i\Delta_2 + \Gamma_{30}) + |G_1|^2\), \(D_2 = G_2^* G_3\), and \(S_1 = (i\Delta_1 + \Gamma_{10})(i\Delta_2^* + \Gamma_{20}) + |G_2|^2\).

Thus we can easily obtain

\[G_1^* + [(S_1 + D_1)/D]G_1^* - [(D_2^2 - S_1 D_1)/D^2]G_1 = 0, \quad (19a)\]

\[G_f^* + [(S_1 + D_1)/D]G_f^* - [(D_2^2 - S_1 D_1)/D^2]G_f = 0. \quad (19b)\]

For given \(G_1(z=0) = 0\) and with \(G_f(z=0) = 0\) in the forward NDFWM configuration [Fig. 1(e)], Figs. 5–8 show the steady-state numerical solutions. For the purpose of comparison, the field propagation equations of the forward probe and backward NDFWM are

\[\partial G_1/\partial z = i\xi_{10}p_{10} = \xi_{10}(D_2 G_f/D - D_1 G_1/D) \quad \text{and} \quad \partial G_f/\partial z = -i\xi_{30}p_{30} = -\xi_{30}(D_2^* G_f/D - S_1 G_1/D),\]

which have the initial conditions \(G_1(z=0) = 0\) and \(G_f(z=l) = 0\) at the entrance [Fig. 1(f)].

An efficiently generated NDFWM field can acquire the same ultrasonic group velocity and pulse shape of the probe pulse and the maximum forward NDFWM efficiency can be greater than 50% in the four-level double-ladder system (Figs. 5–8). One can see from the simulated results that the high NDFWM efficiency prefers the backward scheme [Figs. 1(f) and 9], smaller coupling and pump frequency detunings, and larger coupling and pump Rabi frequencies. The pulse matching of the probe and NDFWM pulses needs sufficient propagation distance in the atomic medium (Figs. 6–9). Specifically, large \(G_2\) is good for NDFWM generation, while large \(G_3\) is good for probe field conversion (Figs. 5 and 6). The maximum NDFWM efficiency (Figs. 5 and 6) qualitatively correlates with the slower group velocity which has also been demonstrated in the double-\(\Lambda\) system [3,4]. Due to \(\sin(\Delta_3^{-1} z)\) and \(\cos(\Delta_3^{-1} z)\) factors obtained from Eqs. (9) [12], the spatial period in the probe and NDFWM propagation curves increases, and the NDFWM generation efficiency be-

FIG. 5. The forward NDFWM conversion efficiency \(\eta_f = \eta_f(\xi_{10}/\Gamma_{20} = 10)/\eta_f(z=0)\) versus (a) the pump Rabi frequency \(G_1\) and (b) the coupling Rabi frequency \(G_2\). The parameters are \(\delta = \Omega_1, \Delta_1 = \Delta_2 = \Delta_3 = 0, \Gamma_{10}/\pi = 5.4 \text{ MHz}, \Gamma_{20}/\pi = 1.8 \text{ MHz}, \) and \(\Gamma_{30}/\pi = 5.9 \text{ MHz}\).

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comes extremely low when we increase $\Delta_i$ (Fig. 8). If $\Delta_2/\Gamma_{30}=\Delta_3/\Gamma_{30}=15$ the matched pulses need a relatively short propagation distance to establish, and the spatial oscillation also disappears [Fig. 7(b)]. This is similar to the $\Delta_2/\Gamma_{30}=\Delta_3/\Gamma_{30}=0$ case [14]. Since the probe and NDFWM EITs have been degraded, Figs. 7(a) and 8(b) with small $G_2$ ($G_3$) or large frequency detuning show the decay effect. Larger $G_2$ is good for NDFWM generation, while smaller $G_2$ is good for NDFWM ultraslow propagation; On the other hand, larger $G_3$ is good for probe conversion, while smaller $G_3$ is good for probe ultraslow propagation. The adequate choices of the balance parameters of a specific double-ladder EIT scheme lead to the high efficiency and ultraslow propagation matched pulse pair of the probe and NDFWM fields. Actually, when two pulses are matched, the group velocities are slowed, but not minimized, and can give best conversion efficiency. However, if we want one of the fields (either probe or NDFWM) to propagate very slow, then the other field will propagate faster. This will break the matched pair and therefore reduce the conversion efficiency.

V. DISCUSSION AND CONCLUSION

The closed-loop coherent NDFWM with double-ladder EIT system has been studied in detail. The matched probe and NDFWM ultraslow propagation is linked to back-and-forth population transfer and coherent coupling in this double-ladder system. One possible experimental candidate for the proposed system ($S \rightarrow P \rightarrow D \rightarrow P \rightarrow S$ type) is Na atoms with energy levels $|0\rangle=|S_{1/2}\rangle$, $|1\rangle=|P_{3/2}\rangle$, $|2\rangle=|D_{3/2}\rangle$, and $|3\rangle=|P_{3/2}\rangle$. The respective transitions are $|0\rangle \rightarrow |1\rangle$ at 590 nm (weak short pulse probe laser) ($\Gamma_1 = 16.9$ ns, $\Gamma_1^{\text{eff}} = 5.7$ ps), $|1\rangle \rightarrow |2\rangle$ at 449 nm (strong cw or long pulse coupling laser), $|3\rangle \rightarrow |2\rangle$ at 1.12 $\mu$m (strong cw or long pulse pump laser), and $|0\rangle \rightarrow |3\rangle$ at 330 nm (ultraviolet NDFWM short pulse radiation). This experimental system will not be very good for Doppler-free conditions in an atomic vapor since $\alpha_0$ and $\alpha_f$ have big difference, and it will be good for UV generation [similar to the four-level cascade system as in Fig. 1(b)]. Another possible experimental candidate for the proposed system is $^{87}$Rb atoms with energy levels $|0\rangle=|5S_{1/2}\rangle$, $|1\rangle=|5P_{3/2}\rangle$, $|2\rangle=|5P_{1/2}\rangle$, and $|3\rangle=|5P_{1/2}\rangle$. The respective transitions are $|0\rangle \rightarrow |1\rangle$ at 795 nm (weak short pulse probe laser) ($\gamma_{10} = 5.4$ MHz, where $\gamma_i$ is term due to spontaneous emission (longitudinal relaxation rate) between states $|i\rangle$ and $|j\rangle$), $|1\rangle \rightarrow |2\rangle$ at 762 nm (strong cw or long pulse coupling laser) ($\gamma_{12} = 0.8$ MHz), $|3\rangle \rightarrow |2\rangle$ at 776 nm (strong cw or long pulse pump laser) ($\gamma_{23} = 0.97$ MHz), and $|0\rangle \rightarrow |3\rangle$ at 780 nm (NDFWM short pulse radiation) ($\gamma_{30} = 5.9$ MHz). This system is similar to the four-level double-$\Lambda$ system and can easily satisfy two-photon Doppler-free configurations in hot Rb vapor [7,15]. For a comparison with the previous works [12–14], we adopt near-unity efficiency in Figs. 7–9. On the other hand, we had the real experiment in mind and used the realistic parameters for real atoms ($^{87}$Rb) in Figs. 5 and 6. The transverse relaxation rate $\Gamma_{ij}$ between states $|i\rangle$ and $|j\rangle$ can be obtained by $\Gamma_{ij}=(\Gamma_i+\Gamma_j)/2$ ($\Gamma_i=0$, $\Gamma_1=\gamma_{10}$, $\Gamma_2=\gamma_{21}+\gamma_{23}$, $\Gamma_3=\gamma_{30}$).

It is interesting to consider the competition between the NDFWM process $G_1G_2G_3$ and feedback NDFWM process...
$G_1^0 G_2^0$ in this four-level double-ladder atomic system with two counter-propagating beam pairs [i.e., probe beam $\omega_1$ and coupling beam $\omega_2$ as one ladder system, and pump beam $\omega_3$, and the generated NDFWM beam $\omega_f$ as another ladder system in Figs. 1(e) and 1(f)]. The forward NDFWM scheme (NDFWM efficiency $\geq 50\%$) as shown in Fig. 1(e) is good for matched pulse pair propagation; while backward NDFWM scheme (NDFWM efficiency $\approx 100\%$) as shown in Fig. 1(f) is good for NDFWM generation.

This four-level double-ladder system has features that combine the advantages of the individual systems of the double-$\Lambda$ [Fig. 1(a)] and the four-level cascade [Fig. 1(b)] configurations. Therefore, we can calculate and optimize the system parameters for desired objectives. The maximum entanglement between the well-matched probe and NDFWM pulses (that propagate with the same ultraslow group velocity) can be obtained only after a characteristic propagation distance in the atomic medium. The matched probe and NDFWM photon pair is an ideal candidate for a quantum correlated photon source for quantum information processing and quantum networking [16].

In conclusion, the suppression of linear absorption of the probe and NDFWM fields (double EIT) facilitates the enhanced NDFWM efficiency in the double-ladder EIT system. After a characteristic propagation distance, the nonlinear absorption and dispersion contributions of the cross-Kerr term and the NDFWM term cancel out each other. As a consequence, a pair of ultraslow, temporally and group-velocity matched probe and NDFWM pulses emerge. The generation of the phase-matched coherent NDFWM field and its feedback NDFWM process are limited by the two destructive interferences between the three-photon and one-photon excitation paths. The maximum generated NDFWM field is achieved at the balance condition $G_f(z,t)/G_1(z,t) = G_2^0/G_2^0$ for this double-ladder system. For the forward NDFWM, the onset of the balance condition occurs deep inside the atomic medium, which limits the NDFWM generation (the NDFWM efficiency is still up to $50\%$ in the ideal case). Two multiphoton destructive interferences can be avoided by detuning the two strong CW (coupling and pump) lasers from their perspective resonances. Under certain conditions, the backward NDFWM efficiency is close to $100\%$ which results from two modes constructive interference. The balance condition may be satisfied throughout the whole medium in the backward NDFWM scheme. To get matched pulses (or correlated photon pairs), we will want $50\%$ probe and $50\%$ NDFWM pulses, so the forward scheme with two multiphoton destructive interferences is better for this case. Three configurations (first, two multiphoton destructive interferences of states $|1\rangle$ and $|3\rangle$; second, the multiphoton destructive interference of state $|3\rangle$ no longer occurs and the NDFWM field is strongly enhanced by constructive interference of the two propagating modes; third, the multiphoton destructive interference of state $|1\rangle$ no longer occurs and the propagating probe field is strongly enhanced by constructive interference of the two modes) of the entangled NDFWM and probe photon pairs can be switched by the frequency detuning which may give one potential application for optical memory.

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