Achieving nonlinear optical modulation via four-wave mixing in a four-level atomic system

Hai-Chao Li,[1] Guo-Qin Ge,[2] and M. Suhail Zubairy[2,3]
[1]*College of Science, China Three Gorges University, Yichang 443002, People's Republic of China*
[2]*School of Physics, Huazhong University of Science and Technology, Wuhan 430074, People's Republic of China*
[3]*Institute for Quantum Science and Engineering (IQSE) and Department of Physics and Astronomy, Texas A&M University, College Station, Texas 77843-4242, USA*

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We propose an accessible scheme for implementing tunable nonlinear optical amplification and attenuation via a synergetic mechanism of four-wave mixing (FWM) and optical interference in a four-level ladder-type atomic system. By constructing a cyclic atom-field interaction, we show that two reverse FWM processes can coexist via optical transitions in different branches. In the suitable input-field conditions, strong interference effects between the input fields and the generated FWM fields can be induced and result in large amplification and deep attenuation of the output fields. Moreover, such an optical modulation from enhancement to suppression can be controlled by tuning the relative phase. The quantum system can be served as a switchable optical modulator with potential applications in quantum nonlinear optics.

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I. INTRODUCTION

Generation and manipulation of optical signals induced by coherent evolution of atom-field interaction at the quantum level have always been subjects of intense research in cavity quantum electrodynamics [1] and play a fundamentally important role in the practical applications of quantum information and computation [2]. A variety of useful techniques for signal generation, amplification, and control have been proposed and carried out in experiments. For example, a known approach to realizing light amplification is lasing [3–5] developed by quantum theory. As an intriguing and counterintuitive quantum optical phenomenon, lasing has already obtained fruitful progress in the past few decades. At the same time the research systems range from previous natural atoms to various new types of quantum media, such as quantum dots [6], nitrogen-vacancy color centers [7], optomechanical systems [8], and superconducting quantum circuits [9].

Another feasible tool to emit and modulate light waves is nonlinear multiwave mixing [10–14]. As a typical high-order nonlinear effect, four-wave mixing (FWM) in multilevel atomic systems has attracted a great deal of attention recently. For instance, an experimental observation of efficient FWM in the pulsed regime at low-light levels was reported in an N-type atomic system [15]. Frequency characteristics of FWM for far-detuned two-photon excitation was examined in a diamond-type system [16]. A resonant FWM conversion scheme with two spatially-varied control fields was proposed in a Λ-shaped configuration structure [17]. Besides, enhancement and suppression of FWM in a four-level system [18] and even six-wave mixing (SWM) in a five-level system [19] was investigated by using the dressing effect. Furthermore, interplays including competition, energy exchange, and interference between these two wave-mixing processes were demonstrated experimentally in an inverted Y-type atomic system [20]. Except for FWM and SWM processes, eight-wave mixing was observed in two different five-level atomic systems [21,22].

However, all these works focus on $2n$-wave mixing itself and intermixing only between nonlinear wave-mixing processes. To our knowledge, the relevant topic of exploring controllable modulation of input signal resulting from interaction of the generated FWM field with the incident field and its possible applications in nonlinear optics has received little attention in atomic systems.

In this paper we propose an accessible scheme for realizing controllable nonlinear optical modulation via a FWM approach in a four-level atomic system. In our model with engineered atom-field interaction, two coexisting FWM processes with three-order nonlinearity can occur. Under suitable input-field conditions, strong constructive and destructive interferences between the input signals and the generated FWM signals are induced and play a crucial role in amplifying and attenuating corresponding output fields. Furthermore, by tuning the relative phase of the incident beams, the output signal transition from enhancement to suppression can be achieved in a large range. Understanding the mechanism for efficient interplays of the input signals with the generated signals will help us to deepen and extend research issues in the field of multiwave mixing, and such a controllable light manipulation can have important applications in designing novel nonlinear optical devices.

II. THEORETICAL MODEL

Let us consider a four-level ladder-type atomic system driven by four laser beams, as shown in Fig. 1. Two weak incident lights, the probe field E_p ($\omega_p, \mathbf{k}_p$, and Rabi frequency Ω_p) and the signal field E_s ($\omega_s, \mathbf{k}_s$, and Rabi frequency Ω_s), connect a common ground state, $|0\rangle$, to two upper states, $|1\rangle$ and $|3\rangle$. Meanwhile, another weak driving field, E_d (ω_d, $\mathbf{k}_d$, and Rabi frequency Ω_d), and a strong control field, E_c (ω_c, $\mathbf{k}_c$, and Rabi frequency Ω_c), couple the transitions $|1\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |3\rangle$, respectively. It is obvious from Fig. 1 that the composite atom-field system forms a cyclic four-level configuration which

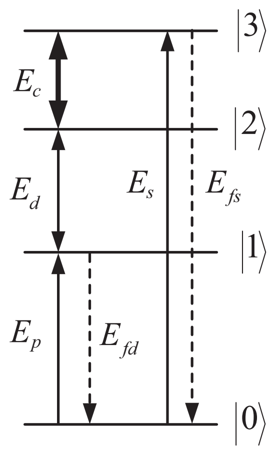

FIG. 1. Schematic of a four-level atomic system interacting with four incident fields (solid lines). In this model with engineered atom-field interaction, two coexisting FWM processes involving the sum-frequency field $E_{\rm fs}$ and the difference-frequency field $E_{\rm fd}$ (dashed lines) are generated synchronously.

indicates the possible transition $|0\rangle \to |1\rangle \to |2\rangle \to |3\rangle \to |0\rangle$ and its reverse pathway. As a result, there are two coexistent FWM processes with three-order nonlinearity via different optical transitions. Specifically, a FWM sum-frequency field $E_{\rm fs}$ with the phase-matching condition $\mathbf{k}_{\rm fs} = \mathbf{k}_p + \mathbf{k}_d + \mathbf{k}_c$ is generated via the perturbation chain $\rho_{00}^{(0)} \to \rho_{10}^{(1)} \to \rho_{20}^{(2)} \to \rho_{30}^{(3)}$ and another FWM difference-frequency field $E_{\rm fd}$ with the phase-matching condition $\mathbf{k}_{\rm fd} = \mathbf{k}_s - \mathbf{k}_c - \mathbf{k}_d$ is generated via the perturbation chain $\rho_{00}^{(0)} \to \rho_{30}^{(1)} \to \rho_{20}^{(2)} \to \rho_{10}^{(3)}$.

The Hamiltonian describing the atom-field interaction for the system is given by ($\hbar = 1$)

$$H = \sum_{j=0}^{3} \omega_j |j\rangle\langle j| - \frac{1}{2}(\Omega_p e^{-i\omega_p t}|1\rangle\langle 0| + \Omega_{\rm fd} e^{-i\omega_{\rm fd} t}|1\rangle\langle 0| + \Omega_d e^{-i\omega_d t}|2\rangle\langle 1| + \Omega_c e^{-i\omega_c t}|3\rangle\langle 2| + \Omega_s e^{-i\omega_s t}|3\rangle\langle 0| + \Omega_{\rm fs} e^{-i\omega_{\rm fs} t}|3\rangle\langle 0| + \text{H.c.}), \tag{1}$$

where $\Omega_{\rm fs}$ ($\Omega_{\rm fd}$) is the Rabi frequency of the generated FWM sum-frequency (difference-frequency) field, and $\omega_{\rm fs} = \omega_p + \omega_d + \omega_c$ ($\omega_{\rm fd} = \omega_s - \omega_c - \omega_d$) is the corresponding traveling frequency. Switching to the interaction picture, the transformed Hamiltonian can be written as

$$H_I = \Delta_p|1\rangle\langle 1| + (\Delta_p + \Delta_d)|2\rangle\langle 2| + (\Delta_p + \Delta_d + \Delta_c)|3\rangle\langle 3| - \frac{1}{2}(\Omega_p|1\rangle\langle 0| + \Omega_{\rm fd}|1\rangle\langle 0| + \Omega_d|2\rangle\langle 1| + \Omega_c|3\rangle\langle 2| + \Omega_s|3\rangle\langle 0| + \Omega_{\rm fs}|3\rangle\langle 0| + \text{H.c.}), \tag{2}$$

where $\Delta_p = \omega_{10} - \omega_p$, $\Delta_d = \omega_{21} - \omega_d$, and $\Delta_c = \omega_{32} - \omega_c$ are the detunings of the probe, driving, and control fields. Here we emphasize that the following frequency relation among four incoming beams should be satisfied in the above calculation, i.e., $\omega_s = \omega_p + \omega_d + \omega_c$. Combining it with the frequency expressions of the two generated FWM fields, we show that the difference-frequency $E_{\rm fd}$ (sum-frequency $E_{\rm fs}$) generation has the same frequency with the probe E_p (signal E_s) field, which is a crucial condition for the realization of light amplification and attenuation as seen later in this paper.

The evolution of dynamics for the atomic system can be governed by the master equation

$$\frac{d\rho}{dt} = -i[H_I,\rho] + (\text{relaxation terms}), \tag{3}$$

where the first term indicates the coherent interaction and the second term denotes the environment-induced dissipation processes. It is well known that linear and nonlinear polarizations for a multilevel atomic system can be described by first-order and high-order off-diagonal density matrix elements. According to Eqs. (2) and (3), the equations of motion for the off-diagonal matrix elements are expressed as

$$\dot\rho_{10} = -(\gamma_{10} + i\Delta_p)\rho_{10} + \frac{i}{2}(\Omega_p + \Omega_{\rm fd})(\rho_{00} - \rho_{11}) + \frac{i}{2}\Omega_d^*\rho_{20} - \frac{i}{2}(\Omega_s + \Omega_{\rm fs})\rho_{13}, \tag{4a}$$

$$\dot\rho_{21} = -(\gamma_{21} + i\Delta_d)\rho_{21} + \frac{i}{2}\Omega_d(\rho_{11} - \rho_{22}) + \frac{i}{2}\Omega_c^*\rho_{31} - \frac{i}{2}(\Omega_p^* + \Omega_{\rm fd}^*)\rho_{20}, \tag{4b}$$

$$\dot\rho_{20} = -[\gamma_{20} + i(\Delta_p + \Delta_d)]\rho_{20} + \frac{i}{2}\Omega_d\rho_{10} + \frac{i}{2}\Omega_c^*\rho_{30} - \frac{i}{2}(\Omega_p + \Omega_{\rm fd})\rho_{21} - \frac{i}{2}(\Omega_s + \Omega_{\rm fs})\rho_{23}, \tag{4c}$$

$$\dot\rho_{32} = -(\gamma_{32} + i\Delta_c)\rho_{32} + \frac{i}{2}\Omega_c(\rho_{22} - \rho_{33}) - \frac{i}{2}\Omega_d^*\rho_{31} + \frac{i}{2}(\Omega_s + \Omega_{\rm fs})\rho_{02}, \tag{4d}$$

$$\dot\rho_{31} = -[\gamma_{31} + i(\Delta_d + \Delta_c)]\rho_{31} + \frac{i}{2}\Omega_c\rho_{21} - \frac{i}{2}\Omega_d\rho_{32} + \frac{i}{2}(\Omega_s + \Omega_{\rm fs})\rho_{01} - \frac{i}{2}(\Omega_p^* + \Omega_{\rm fd}^*)\rho_{30}, \tag{4e}$$

$$\dot\rho_{30} = -[\gamma_{30} + i(\Delta_p + \Delta_d + \Delta_c)]\rho_{30} + \frac{i}{2}\Omega_c\rho_{20} + \frac{i}{2}(\Omega_s + \Omega_{\rm fs})(\rho_{00} - \rho_{33}) - \frac{i}{2}(\Omega_p + \Omega_{\rm fd})\rho_{31}, \tag{4f}$$

where the damping rate $\gamma_{ij} (i > j)$ is inserted phenomenologically. Assuming that the atomic system is initially populated in the ground state $|0\rangle$, the steady-state solutions of the off-diagonal matrix elements responsible for the first-order and third-order optical processes are obtained by a formal perturbation expansion:

$$\rho_{10}^{(1)} = \frac{i(\Omega_p + \Omega_{\rm fd})}{2\Gamma_{10}}, \tag{5a}$$

$$\rho_{30}^{(1)} = \frac{i\Gamma_{20}(\Omega_s + \Omega_{\rm fs})}{2\xi}, \tag{5b}$$

$$\rho_{10}^{(3)} = \frac{i^3\Omega_s\Omega_c^*\Omega_d^*}{8\Gamma_{10}\xi} + \frac{i^3\Omega_{\rm fs}\Omega_c^*\Omega_d^*}{8\Gamma_{10}\xi}, \tag{5c}$$

$$\rho_{30}^{(3)} = \frac{i^3\Omega_p\Omega_d\Omega_c}{8\Gamma_{10}\xi} + \frac{i^3\Omega_{\rm fd}\Omega_d\Omega_c}{8\Gamma_{10}\xi}, \tag{5d}$$

where $\Gamma_{10} = \gamma_{10} + i\Delta_p$, $\Gamma_{20} = \gamma_{20} + i(\Delta_p + \Delta_d)$, $\Gamma_{30} = \gamma_{30} + i(\Delta_p + \Delta_d + \Delta_c)$, and $\xi = \Gamma_{20}\Gamma_{30} + |\Omega_c|^2/4$. Equations (5a) and (5b) are usually used to quantify the linear susceptibilities, which govern the dispersion and absorption properties of the probe, signal, and two FWM fields. The first terms in Eqs. (5c) and (5d) demonstrate the FWM difference- and sum-frequency conversions with third-order nonlinearity, and the second terms display the backward nonlinear processes of the generated FWM fields. In light of the slowly varying amplitude approximation [23], two sets of coupled equations mastering the energy transfers for the fields E_p and $E_{\rm fs}$ and the fields E_s and $E_{\rm fd}$ during their propagation are expressed as

$$\frac{\partial\Omega_p}{\partial z} = i\kappa_{01}\left[\frac{i\Omega_p}{2\Gamma_{10}} + \frac{i^3\Omega_{\rm fs}\Omega_c^*\Omega_d^*}{8\Gamma_{10}\xi}\right], \tag{6a}$$

$$\frac{\partial\Omega_{\rm fs}}{\partial z} = i\kappa_{03}\left[\frac{i\Gamma_{20}\Omega_{\rm fs}}{2\xi} + \frac{i^3\Omega_p\Omega_d\Omega_c}{8\Gamma_{10}\xi}\right], \tag{6b}$$

$$\frac{\partial\Omega_s}{\partial z} = i\kappa_{03}\left[\frac{i\Gamma_{20}\Omega_s}{2\xi} + \frac{i^3\Omega_{\rm fd}\Omega_d\Omega_c}{8\Gamma_{10}\xi}\right], \tag{7a}$$

$$\frac{\partial\Omega_{\rm fd}}{\partial z} = i\kappa_{01}\left[\frac{i\Omega_{\rm fd}}{2\Gamma_{10}} + \frac{i^3\Omega_s\Omega_c^*\Omega_d^*}{8\Gamma_{10}\xi}\right]. \tag{7b}$$

III. RESULTS AND DISCUSSION

We now intend to present the controllable nonlinear optical amplification and attenuation based on the synergetic mechanism of FWM conversion and optical interference. It should be pointed out again that all the equations from Eq. (2) are calculated in the frame of $\omega_s = \omega_p + \omega_d + \omega_c$. When another condition, $\mathbf{k}_s = \mathbf{k}_p + \mathbf{k}_d + \mathbf{k}_c$, among four incident beams is satisfied simultaneously, using previous two phase-matching conditions we show that the signal and FWM sum-frequency (probe and FWM difference-frequency) fields propagate along the same direction $\mathbf{k}_s$ ($\mathbf{k}_p$). As a result, each pair of optical fields is indistinguishable [12,18] and the total output amplitude $E_s^{\rm tot}$ ($E_p^{\rm tot}$) can be deemed as a coherent superposition of these two signals, i.e., $E_s^{\rm tot} = E_s + E_{\rm fs}$ ($E_p^{\rm tot} = E_p + E_{\rm fd}$). In this case, two sets of optical interferences are induced and play a fundamentally essential role in the output light amplification and attenuation. Solving Eqs. (6) and (7), we have

$$\begin{aligned} E_s^{\rm tot}/E_{s0} = {} & \frac{\kappa b - a + \lambda}{2\lambda}\exp\left[(a+\kappa b-\lambda)Z/2\right] \\ & - \frac{\kappa b - a - \lambda}{2\lambda}\exp\left[(a+\kappa b+\lambda)Z/2\right] \\ & + \frac{c|\Omega_{p0}|}{\lambda|\Omega_{s0}|}e^{-i\phi}\{\exp\left[(a+\kappa b+\lambda)Z/2\right] \\ & - \exp\left[(a+\kappa b-\lambda)Z/2\right]\}, \end{aligned} \tag{8}$$

$$\begin{aligned} E_p^{\rm tot}/E_{p0} = {} & \frac{a - \kappa b + \lambda}{2\lambda}\exp\left[(a+\kappa b-\lambda)Z/2\right] \\ & - \frac{a - \kappa b - \lambda}{2\lambda}\exp\left[(a+\kappa b+\lambda)Z/2\right] \\ & + \frac{\kappa c|\Omega_{s0}|}{\lambda|\Omega_{p0}|}e^{i\phi}\{\exp\left[(a+\kappa b+\lambda)Z/2\right] \\ & - \exp\left[(a+\kappa b-\lambda)Z/2\right]\}, \end{aligned} \tag{9}$$

where $\lambda = \sqrt{(\kappa b - a)^2 + 4\kappa c^2}$, $\kappa = \kappa_{01}/\kappa_{03}$, $a = -\Gamma_{20}/(2\xi)$, $b = -1/(2\Gamma_{10})$, $c = |\Omega_d||\Omega_c|/(8\Gamma_{10}\xi)$, and $Z = \kappa_{03}z$ is the effective propagation distance. Note that the Rabi frequencies Ω_p, Ω_d, Ω_c, and Ω_s are treated as complex parameters: $\Omega_p = |\Omega_p|e^{-i\phi_p}$, $\Omega_d = |\Omega_d|e^{-i\phi_d}$, $\Omega_c = |\Omega_c|e^{-i\phi_c}$, and $\Omega_s = |\Omega_s|e^{-i\phi_s}$, where ϕ_p, ϕ_d, ϕ_c and ϕ_s are the phases of the probe, driving, control and signal fields, respectively. The first two terms on the right side of Eqs. (8) and (9) dominate the evolutional processes of the probe and signal beams while the third terms are responsible for the propagation dynamics of the generated FWM beams. It is seen from these equations that two output signals, $E_s^{\rm tot}$ and $E_p^{\rm tot}$, are sensitive to the relative phase $\phi = \phi_p + \phi_d + \phi_c - \phi_s$ and then can be controlled effectively by tuning ϕ.

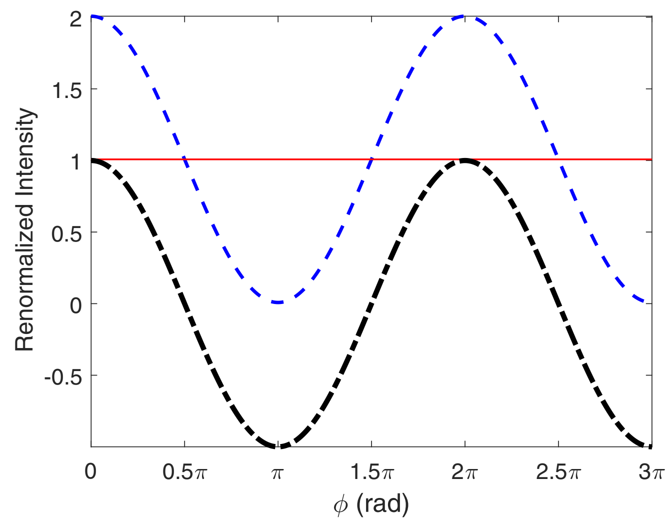

FIG. 2. Output signal intensity $|E_s^{\rm tot}/E_{s0}|^2$ (blue dashed line), the sum of $|E_{\rm fs}/E_{s0}|^2 + |E_s/E_{s0}|^2$ (red solid line) and the interference term $|E_s^{\rm tot}/E_{s0}|^2 - |E_{\rm fs}/E_{s0}|^2 - |E_s/E_{s0}|^2$ (black dash-dotted line) as a function of the relative phase ϕ. The parameters are $\gamma_{20} = 0.05\gamma_{10}$, $\gamma_{30} = \gamma_{10}$, $Z = 10\gamma_{10}$, $\kappa = 0.1$, $\Omega_d = 0.15\gamma_{10}$, $\Omega_c = 1.8\gamma_{10}$, $|\Omega_{p0}|/|\Omega_{s0}| = 2.5$, and $\Delta_p = \Delta_d = \Delta_c = 0$.

According to Eq. (8), it is obvious that when $|E_s^{\rm tot}/E_{s0}|^2$ is larger than 1, the signal field E_s will be amplified after passing through the four-level atomic system. On the contrary, if it is smaller than 1, the signal field will be attenuated. Figure 2 plots the evolutional curves of the renormalized output intensity $|E_s^{\rm tot}/E_{s0}|^2$, the sum of $|E_{\rm fs}/E_{s0}|^2 + |E_s/E_{s0}|^2$ and the interference term $|E_s^{\rm tot}/E_{s0}|^2 - |E_{\rm fs}/E_{s0}|^2 - |E_s/E_{s0}|^2$ as a function of the relative phase ϕ. From this picture, we see that the output intensity and the interference term present an interesting periodic oscillating behavior as the relative phase ϕ increases. In addition, the peak value of the interference profile is roughly equal to the sum $|E_{\rm fs}/E_{s0}|^2 + |E_s/E_{s0}|^2$, which can bring about significant interference-induced light enhancement and suppression. To be specific,the maximum output intensity is obtained at $\phi = 0$, accompanied by the strongest constructive interference between the incoming light E_s and the generated light $E_{\rm fs}$. The minimum intensity reduces to zero at $\phi = \pi$ where the complete destructive interference occurs. And the signal-field output recovers its maximal value at $\phi = 2\pi$. Thus we demonstrate that the output signal $E_s^{\rm tot}$ can vary from large amplification to deep attenuation by tuning the relative phase, which indicates the atom-field system is able to serve as a phase-dependent amplitude modulator.

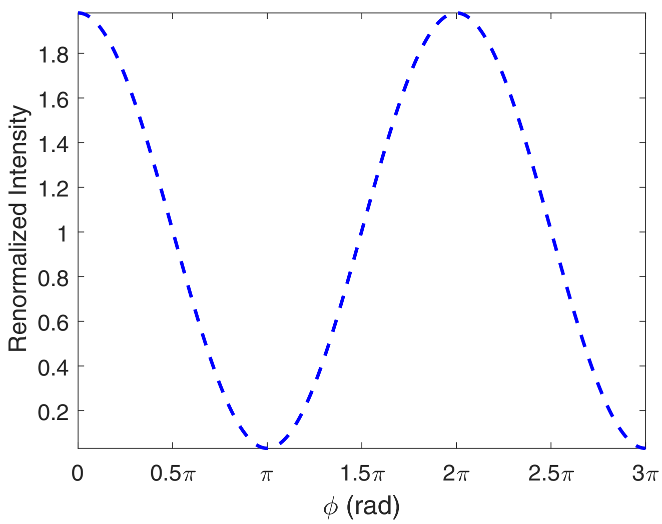

FIG. 3. Output signal intensity $|E_p^{\rm tot}/E_{p0}|^2$ versus the relative phase ϕ. The parameters are $\gamma_{20} = 0.05\gamma_{10}$, $\gamma_{30} = \gamma_{10}$, $Z = 10\gamma_{10}$, $\kappa = 0.1$, $\Omega_d = 0.15\gamma_{10}$, $\Omega_c = 1.2\gamma_{10}$, $|\Omega_{s0}|/|\Omega_{p0}| = 25$, and $\Delta_p = \Delta_d = \Delta_c = 0$.

By simply analyzing Eqs. (8) and (9), we evaluate that the intensity of the output field $E_p^{\rm tot}$ should possess characters analogous to those of $E_s^{\rm tot}$ in the nonlinear regime, as verified in Fig. 3 where we diagram the evolution of $|E_p^{\rm tot}/E_{p0}|^2$, versus the relative phase ϕ. Consequently, we also achieve the phase-dependent amplification and attenuation in a large range for the output field $E_p^{\rm tot}$ and show that the driven system is capable of acting as a controllable amplitude modulator for the probe field.

Note that in the above discussion we do not take into account Doppler broadening because it is negligible in cold atoms. In recent years, many FWM experiments with cold atoms have been carried out [10,15,24,25], which shows our scheme is feasible in the current technology. A potential candidate for the proposed system is cold ^{85}Rb atoms confined in a magneto-optical trap, and the quantum states are chosen as follows: $5S_{1/2}, F = 2$ ($|0\rangle$), $5P_{1/2}$ ($|1\rangle$), $5D_{3/2}$ ($|2\rangle$), $nP_{3/2}$ ($|3\rangle$), with $n > 10$. The respective transitions are $|0\rangle \to |1\rangle$ at 795 nm, $|1\rangle \to |2\rangle$ at 762 nm, and $|2\rangle \to |3\rangle$ at 1.3–1.5 μm.

IV. CONCLUSIONS

In conclusion, we have presented an efficient nonlinear scheme to achieve the controllable optical modulation in a cyclically driven four-level atomic system. Based on the synergetic mechanism of FWM conversion and optical interference, we have demonstrated large enhancement and deep suppression of two output beams. Furthermore, we have shown the output signal transition from amplification to attenuation by tuning the relative phase, indicating the driven system could act as a switchable optical modulator with widespread applications in optical devices, for example, an all-optical switch. Our study opens up an intriguing physical perspective in manipulating optical signals and may be used to extend potential subjects in the area of optical modulation and multiwave mixing.

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