Nuclear-state population transfer by a train of coincident pulses

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Population transfer of three-level Λ-like nuclei interacting with x-ray free electron lasers (XFEL) via a train of coincident pulses has been investigated theoretically. This study uses copropagating beams in which the frequency of the pump laser is different from that of the Stokes laser. We calculate the required laser intensities for each step which satisfy the condition of coincident-pulse technique in different nuclei. By employing the master equation and considering the effect of spontaneous emission, we show that complete nuclear population transfer occurs by choosing the appropriate number of pulse pairs. It is shown that the effect of laser intensity fluctuation is suppressed by increasing the number of pulse pairs. In this scheme, after each step, the populations of system are in stable states and the time delays between the neighboring pulses do not affect the transmission efficiency.

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I. INTRODUCTION

Coherent population transfer in Λ-like quantum systems by external resonant laser fields represents an important notion in quantum mechanics. Stimulate Raman adiabatic passage (STIRAP) [1–5] and π pulse [6,7] are two important techniques that are used to complete population transfer between the two end states in a three-state Λ-like system. STIRAP can be implemented using exact resonance (or two-photon resonance) counterintuitive pump and Stokes laser fields (Stokes before pump) with large pulse areas in time. STIRAP is an approximate technique and for sufficiently large laser pulses on the time scale of the system's evolution is robust against variation in the pulse intensity, the pulse delay, and the intermediate state detuning. In return, π pulse needs a small pulse area compared to STIRAP; however, it is sensitive to exact pulse area and exact resonance field conditions, and also the excited state takes a significant population during the evolution.

In recent years with the development of the x-ray free electron laser (XFEL) [8–13] coherent control of nuclear states has been taken into consideration and has opened a prominent field in nuclear quantum optics [14–17] and nuclear quantum information processing [18]. Recently, nuclear coherent population transfer (NCPT) with x-ray laser pulses has been implemented in two-level [14] and three-level Λ-like systems [19,20] with transition energies of a few hundred keV. In Refs. [19,20], two π pulses have been used for nuclei with long-lifetime excited states and STIRAP has been implemented for nuclei with short-lifetime excited states. In the proposed schemes in Refs. [14,19,20], due to the need for short wavelengths, the nuclei should be accelerated (Doppler shift) in order to match the resonance for transition between energy levels of the nuclei.

Considering that π pulse requires precise designs of the pulse area and STIRAP needs large pulse areas, which may be hard to reach experimentally, piecewise adiabatic passage (PAP) has been proposed in Refs. [21,22] to complete population transfer in three-state and two-state systems. PAP extends the concepts of coherent accumulation [23,24] and Ramsey's experiment [25], where a slow process is implemented in a piecewise manner by using a train of short, mutually coherent pulse pairs, each of which produces a small change in the populations. Recently Rangelov and Vitanov [26] proposed a technique to complete population transfer in three-state quantum systems by a train of N pairs of coincident pulses, in which the population in the excited state is suppressed to negligible small value by increasing the pulse pairs. In this technique [26] the number of pulse pairs is arbitrary and the robustness of system against deviation from exact pulse areas and spontaneous emission from excited state rise with increasing numbers of pulse pairs. In addition to the complete population in three-state systems, the coincident pulse technique [26] has been used very recently to coherent superposition in multistate linkage patterns [27–29] and synthesis of fast qudit gates [30]. Given that in some nuclei the lifetimes of excited states are very short and in some others they are long, the coincident pulse technique can be a good choice for NCPT.

In this paper we investigate the coincident-pulse technique [26] in order to NCPT in three-state Λ-like systems. Throughout the present study we assume a fully coherent XFEL source such as the future XFEL Oscillator (XFELO) [31] or the seeded XFEL (SXFEL) [8–13] for both pump and Stokes lasers and at each stage the required laser intensities will be calculated exactly. To take into account the spontaneous emission of excited state into two ground states, we study the population transfer of the system by master equation. Some of the advantages of this method are as follows: (i) This technique can be used for both short- and long-lifetime excited-state nuclei. (ii) The robustness of the system against the small variation of laser intensities and spontaneous emission rise by increasing the number of pulse pairs. (iii) This technique is exact and the laser intensities which have been calculated exactly for each step can be valuable practice for implementation. (iv) In this method, after each sequence of pulse pairs, the populations of system are in the stable states and as a result the time delays between the neighboring pulses will not affect population transfer efficiency.

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This paper is organized as follows. We define the technique of coincident pulses in Sec. II and nuclear population transfer by a train of coincident pulses is introduced in Sec. III. In Sec. IV we demonstrate compensation of laser intensity fluctuation by a train of coincident pulses. The conclusions are summarized in Sec. V.

II. COINCIDENT PULSES TECHNIQUE

Figure 1 shows the linkage pattern of radiative interactions in a three-level Λ-like system, which is related to coincident-pulse technique. The internal dynamics of the system is described by the time-dependent Schrödinger equation (TDSE),

$$i\hbar\frac{\partial}{\partial t}|\Psi(t)\rangle = \hat{H}(t)|\Psi(t)\rangle, \tag{1}$$

where $\hat{H}(t)$ is the Hamiltonian matrix in the subspace $S = \{|1\rangle,|2\rangle,|3\rangle\}$ for the system and its interaction with the pulses. The state vector $|\Psi(t)\rangle$ is a three-component column vector. The Hamiltonian $\hat{H}(t)$ in the rotating-wave approximation [6,7] in exact resonance and in the absence of decoherence reads

$$\hat{H}(t) = \frac{\hbar}{2}\begin{pmatrix} 0 & 0 & \Omega_P(t) \\ 0 & 0 & \Omega_S(t) \\ \Omega_P(t) & \Omega_S(t) & 0 \end{pmatrix}, \tag{2}$$

where $\Omega_P(t)$ and $\Omega_S(t)$ are the Rabi frequencies of the pump and Stokes pulses. We impose the condition that the Rabi frequencies are pulse-shaped functions with the same time dependences, but possibly with different magnitudes. The interest in Hamiltonian (2) is mainly determined by the existence of the dark state. The dark state is a coherent superposition of initial state, $|1\rangle$, and the final state, $|2\rangle$. A new basis which is called the bright-dark state or Morris-Shore basis [32–36] will be defined as follows:

$$|v_1\rangle = \sin\varphi|1\rangle + \cos\varphi|2\rangle, \tag{3a}$$

$$|v_2\rangle = \cos\varphi|1\rangle - \sin\varphi|2\rangle, \tag{3b}$$

$$|v_3\rangle = |3\rangle, \tag{3c}$$

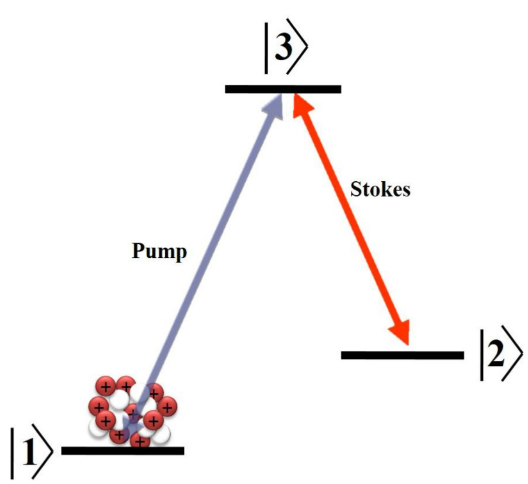

FIG. 1. Linkage pattern of a three-level Λ-like system. All initial population is in state $|1\rangle$.

where

$$\tan\varphi = \frac{\Omega_P(t)}{\Omega_S(t)}. \tag{4}$$

The Hamiltonian in the basis of dark and bright states $S^T = \{|v_1\rangle,|v_2\rangle,|v_3\rangle\}$ can be written as

$$\hat{H}_T(t) = \hat{T}^\dagger\hat{H}(t)\hat{T} = \frac{\hbar}{2}\begin{pmatrix} 0 & 0 & \Omega(t) \\ 0 & 0 & 0 \\ \Omega(t) & 0 & 0 \end{pmatrix}, \tag{5}$$

where $\Omega(t) = \sqrt{\Omega_P(t)^2 + \Omega_S(t)^2}$ and $\hat{T}$ is

$$\hat{T} = \begin{pmatrix} \sin\varphi & \cos\varphi & 0 \\ \cos\varphi & -\sin\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{6}$$

The corresponding propagator in the dark-bright basis can be calculated as follows:

$$\hat{U}_T(t) = e^{-i\int_{t_i}^{t}\hat{H}_T(t)dt} = \begin{pmatrix} \cos\frac{1}{2}A(t) & 0 & -i\sin\frac{1}{2}A(t) \\ 0 & 1 & 0 \\ -i\sin\frac{1}{2}A(t) & 0 & \cos\frac{1}{2}A(t) \end{pmatrix}, \tag{7}$$

where the rms pulse area is $A = \int_{t_i}^{t}\Omega(t)dt$. Taking $A(t) = 2\pi$, the propagator in the original basis S at the end of the evolution is

$$\hat{U}(\varphi) = \hat{T}\hat{U}_T(t)\hat{T}^\dagger = \begin{pmatrix} \cos 2\varphi & -\sin 2\varphi & 0 \\ -\sin 2\varphi & -\cos 2\varphi & 0 \\ 0 & 0 & -1 \end{pmatrix}. \tag{8}$$

According to propagator (8) one can find the exact analytic solution for complete population transfer from state $|1\rangle$ to the final state $|2\rangle$. For $\varphi = \pi/4$, the population of state $|1\rangle$ can be transposed to the final state $|2\rangle$. Then, the excited state $|3\rangle$ takes a significant population during evolution. In order to suppress the population of excited state, we use a sequence of N pairs of coincident pulse each with $A(t) = 2\pi$ and mixing angels φ_k. Finally the following total evolution matrix [26] is produced as follows:

$$\hat{U}^N = \hat{U}(\varphi_N)\hat{U}(\varphi_{N-1})\ldots\hat{U}(\varphi_k)\ldots\hat{U}(\varphi_1), \tag{9}$$

where φ_k is given by

$$\varphi_k = \frac{(2k-1)\pi}{4N} \quad (k = 1,2,3,\ldots,N). \tag{10}$$

The maximum population of state $|3\rangle$ in the middle of each pulse pair is damped to small values by increasing the number of pulse sets as follows:

$$P_3^{\max} = \sin^2\left(\frac{\pi}{4N}\right). \tag{11}$$

III. NUCLEAR POPULATION TRANSFER BY A TRAIN OF COINCIDENT PULSES

We study the collider system depicted in Fig. 2, which is composed of an accelerated nuclear beam that interacts with two incoming coincident XFEL pulses. The explanation of the

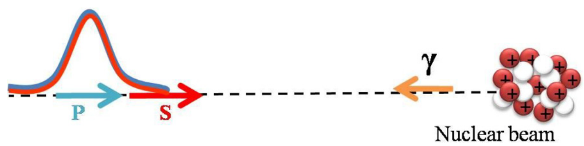

FIG. 2. Two-color scheme (copropagating beams) in the laboratory frame. The frequency of pump laser is different from that of the Stokes laser. The nuclear beam is accelerated such that $\gamma(1+\beta)\omega_{P(S)} = ck_{31(2)}$ is fulfilled.

notations used throughout the following text and the equation can be found in Table I. The nuclear excitation energies are typically higher than the designed photon energy of the XFELO and SXFEL. The accelerated nuclei can interact with two Doppler-shifted x-ray coincident laser pulses as shown in Fig. 2. The two laser frequencies (two color) and the relativistic factor γ of the accelerated nuclei have to be chosen such that in the nuclear rest frame both of one-photon resonances are fulfilled.

The XFEL-nuclei interaction in the nuclear rest frame has been illustrated by the level scheme in Fig. 1. The nuclear dynamics is governed by the master equation for the nuclear density matrix $\hat{\rho}(t)$ [2,19,20,37]

$$\frac{\partial}{\partial t}\hat{\rho} = \frac{1}{i\hbar}[\hat{H},\hat{\rho}] + \hat{\rho}_s + \hat{\rho}_d, \tag{12}$$

where the Hamiltonian $\hat{H}(t)$ is given by Eq. (2); $\hat{\rho}_s$ is the decoherence matrix caused by spontaneous emission from the upper level; and $\hat{\rho}_d$ is an additional dephasing matrix to model laser field pulses with limited coherence times. The decoherence matrix, $\hat{\rho}_s$, is

$$\hat{\rho}_s = \frac{\Gamma}{2}\begin{pmatrix} 2B_{31}\rho_{33} & 0 & -\rho_{13} \\ 0 & 2B_{32}\rho_{33} & -\rho_{23} \\ -\rho_{31} & -\rho_{32} & -2\rho_{33} \end{pmatrix}, \tag{13}$$

TABLE I. The notations used throughout the text. The indices $i,j = 1,2,3$ denote the three nuclear states showed in Fig. 1. The label (Rest) indicates that the corresponding values are in the laboratory (nuclear rest) frame.

Notation	Frame	Explanation
c	Any	Speed of light in vacuum
β	Lab	Velocity of the nuclear particle, in units of c
γ	Lab	Relativistic factor
ϵ_0	Any	Vacuum permittivity
k_{3i}	Rest	Wave number of $\|3\rangle \longrightarrow \|i\rangle$ transition
$\Gamma_{LP(S)}$	Lab	Laser bandwidth of pump (Stokes)
$\tau_{P(S)}$	Rest	Temporal peak position of pump (Stokes) laser
$T_{P(S)}$	Lab	Pulse duration of pump (Stokes) laser
$I_{1(2)}$	Any	Angular momentum of ground state $\|1\rangle$ ($\|2\rangle$)
$L_{1(2)}$	Any	Multipolarity of the corresponding nuclear $\|i\rangle \longrightarrow \|3\rangle$ transition
$B(\varepsilon/\mu L_{i3})$	Rest	Reduced transition probability for the nuclear electric (ε) or magnetic (μ)$\|i\rangle \longrightarrow \|3\rangle$ transition

where Γ is linewidth of state $|3\rangle$ and B_{3i} is branching ratio of $|3\rangle \to |i\rangle (i = 1,2)$. The dephasing effect can be avoided by using a fully coherent XFEL to derive nuclear transitions. Considering a fully coherent XFEL source for both pump and Stokes lasers, in our method the term $\hat{\rho}_d$ in Eq. (12) is zero. The initial conditions are

$$\rho_{ij}(0) = \delta_{i1}\delta_{1j}. \tag{14}$$

Considering the nuclear physics and relativistic treatment for the nuclei in the accelerated beam, and in order to implement the technique of coincident pulses, Rabi frequencies can be obtained as follows [38,39]:

$$\Omega_{p(S)}(t) = \frac{4\sqrt{\pi}}{\hbar}\left[\frac{\gamma^2(1+\beta)^2 I^{\mathrm{eff}}_{P(S)}(L_{1(2)3}+1)(2I_{1(2)}+1)B(\varepsilon/\mu L_{1(2)3})}{c\epsilon_0 L_{1(2)3}}\right]^{1/2}\frac{k_{31(2)}^{L_{1(2)3}-1}}{(2L_{1(2)3}+1)!!}\exp\left\{-\left[\frac{\gamma(1+\beta)(t-\tau_{p(S)})}{\sqrt{2}T_{p(S)}}\right]^2\right\}. \tag{15}$$

The values of parameters for all nucleus are given in Tables II–IV. Laser quantities in Eq. (15) is written in the nuclear rest frame, which leads to

(i) the angular frequency $\gamma(1+\beta)\omega_{p(S)}$,
(ii) bandwith $\gamma(1+\beta)\Gamma_{p(S)}$,
(iii) pulse duration $T_{p(S)}/[\gamma(1+\beta)]$,
(iv) laser peak intensity $\gamma^2(1+\beta)^2 I_{p(S)}$.

Here it is necessary to explain the concept of effective intensity, $I^{\mathrm{eff}}_{p(S)}$. For the long laser pulse case, the bandwidth of the incident laser of intensity, $I_{p(S)}$, is narrower than the linewidth, Γ, of the considered nuclear transition (i.e., $\Gamma \geqslant \gamma(1+\beta)\Gamma_{p(S)}$) and as a result, $I^{\mathrm{eff}}_{p(S)}$, will be equal to $I_{p(S)}$. For the short pulse case the effective intensity is significantly reduced since the bandwidth of the incident laser is wider than Γ (i.e., $\Gamma < \gamma(1+\beta)\Gamma_{p(S)}$) and the effective intensity, $I^{\mathrm{eff}}_{p(S)}$, will be different from $I_{p(S)}$. The relationship between $I^{\mathrm{eff}}_{p(S)}$ and $I_{p(S)}$ can be written as follows:

$$I^{\mathrm{eff}}_{p(S)} = \begin{cases} I_{p(S)} & \text{if } \Gamma \geqslant \gamma(1+\beta)\Gamma_{p(S)} \\ I_{p(S)}\frac{\Gamma}{\gamma(1+\beta)\Gamma_{p(S)}} & \text{if } \Gamma < \gamma(1+\beta)\Gamma_{p(S)} \end{cases}. \tag{16}$$

TABLE II. Fixed laser parameters for all nuclei. The pump photon energies are 12.4 keV for SXFEL and 25 keV for XFELO, respectively. The $\Gamma_{Lp(S)}$ is laser bandwidth of pump (Stokes) and $T_{p(S)}$ is pulse duration of pump (Stokes) laser in the laboratory frame.

XFEL	$T_{p(S)}$ (ps)	$\Gamma_{p(S)}$ (meV)	$\hbar\omega_p$ (keV)
SXFEL	0.1	10	12.4
XFELO	1	1	25

TABLE III. Nuclear, XFEL, and nuclear beam parameters, linewidth of state $|3\rangle$, and branching ratio of $|3\rangle \rightarrow |i\rangle (i = 1,2)$, which are related to spontaneous decay. E_i is the energy of state $|i\rangle$ with $i \epsilon 1,2,3$ (in keV). The multipolarities and reduced matrix elements (in Weisskopf units) for the transitions $|i\rangle \longrightarrow |3\rangle$ with $i \epsilon 1,2$ have been given. The accelerated nuclei have the relativistic factor, γ, determined by the one-photon resonance condition, $\gamma(1+\beta)\hbar\omega_P = ck_{31}$. $\hbar\omega_S$ denotes the Stokes photon energy.

Nucleus	E_3	E_2	E_1	Line with	$\varepsilon/\mu L$		$B(\varepsilon/\mu L)$ (wsu)		Branching ratio		SXFEL		XFELO	
				of $\|3\rangle$(meV)	L_{13}	L_{23}	$\|1\rangle \longrightarrow \|3\rangle$	$\|2\rangle \longrightarrow \|3\rangle$	B_{31}	B_{32}	γ	$\hbar\omega_s$(keV)	γ	$\hbar\omega_s$(keV)
^{223}Ra	50.13	29.86	0.00	0.00144	$E1$	$E1$	1.19×10^{-3}	5×10^{-4}	0.9728	0.0272	2.1	5.01	1.3	10.11
^{113}Cd	522.26	316.21	263.54	0.00145	$E2$	$E1$	4.42×10	1.19×10^{-6}	0.9879	0.1205	10.5	9.88	5.2	19.91
^{97}Tc	567.00	324.00	96.57	0.61	$E2$	$E1$	5×10^{2}	6.7×10^{-5}	0.9653	0.0058	22.6	7.36	11.2	14.83
^{154}Gd	1241.00	123.00	0.00	300	$E1$	$E1$	4.4×10^{-2}	4.9×10^{-2}	0.5167	0.4752	50.1	11.17	24.8	22.52
^{172}Yb	1599.87	78.74	0.00	42.50	$E1$	$E1$	1.8×10^{-3}	1.23×10^{-3}	0.3911	0.6017	64.5	11.79	32.0	23.77
^{168}Er	1786.00	79.00	0.00	133.16	$E1$	$E1$	3.2×10^{-3}	9.1×10^{-3}	0.2908	0.7092	72	11.85	35.7	23.88

The multipolarities $B(\varepsilon/\mu L_{1(2)3})$ are in Weisskopf units so that for the $E_1(|i\rangle \longrightarrow |j\rangle)$ transition which corresponds to $L_{ij} = 1$, $B(\varepsilon/\mu L_{1(2)3})$, in Table III should be multiplied by $0.06446 \times A^{(2/3)}\, e^2\, \mathrm{fm}^2$ and for the $E_2(|i\rangle \longrightarrow |j\rangle)$ transition which corresponds to $L_{ij} = 2, B(\varepsilon/\mu L_{1(2)3})$, in Table III should be multiplied by $0.05940 \times A^{(4/3)}\, e^2\, \mathrm{fm}^4$.

In order to implement the technique of a train of pairs of the coincident pulses, the Rabi frequencies described in Eq. (15) for each nuclear (X) in the kth set can be rewritten in the following form:

$$\Omega_{pX}(t) = \Omega_{0pX}\sqrt{\tilde{I}_p}\sin\varphi_k \exp\left\{-\left[\frac{(t-\tau_p)}{\tilde{T}_p}\right]^2\right\}, \quad (17a)$$

$$\Omega_{SX}(t) = \Omega_{0SX}\sqrt{\tilde{I}_S}\cos\varphi_k \exp\left\{-\left[\frac{(t-\tau_S)}{\tilde{T}_S}\right]^2\right\}, \quad (17b)$$

where

$$\Omega_{0p(S)X} = \frac{4\sqrt{\pi}k_{31(2)}^{L_{1(2)3}-1}}{\hbar(2L_{1(2)3}+1)!!}\left[\frac{\gamma^2(1+\beta)^2(L_{1(2)3}+1)(2I_{1(2)}+1)B(\varepsilon/\mu L_{1(2)3})}{c\epsilon_0 L_{1(2)3}}\right]^{1/2}, \quad (18a)$$

$$\tilde{T}_{p(S)} = \frac{\sqrt{2}T_{p(S)}}{\gamma(1+\beta)}, \quad (18b)$$

$$\tilde{I}_p = \frac{I_p^{\mathrm{eff}}}{\sin^2\varphi_k}, \quad (18c)$$

$$\tilde{I}_S = \frac{I_S^{\mathrm{eff}}}{\cos^2\varphi_k}. \quad (18d)$$

The first condition for implementation of the coincident-pulse technique is the pump and Stokes pulses, in which in each step has the same time dependance, which means that $\tau_p = \tau_S = \tau$ and $\tilde{T}_p = \tilde{T}_S = \tilde{T}$ or $T_p = T_S = T$. The second condition is that the rms pulse area at the end of each step should satisfy the following condition:

$$A(t_f) = \int_{t_i}^{t_f}\sqrt{\Omega_{pX}(t)^2 + \Omega_{SX}(t)^2}dt = 2\pi. \quad (19)$$

To take into view the equation

$$\tilde{I}_{p(S)} = \left(\frac{2\sqrt{\pi}}{\tilde{T}\Omega_{0p(S)X}}\right)^2, \quad (20)$$

the second condition of coincident-pulse technique will be satisfied. In order to implement the coincident-pulse technique, we should adjust the lasers intensities at each step so that they satisfy condition (20). Obviously, for long-lived excited state, $|3\rangle$, of nucleus, complete population transfer will occur by one step of coincident pulses. In Fig. 3, we clearly see the working conditions for coincident pulses technique using SXFEL pulses in ^{97}Tc, in which the lifetime of state $|3\rangle$ (0.76 ps) is longer than the laser pulse duration in the nuclear rest frame. As shown in Fig. 3, complete population transfer occurs by one step of coincident pulses. Figure 4 shows another example of complete population transfer using SXFEL pulses in ^{154}Gd, in which the lifetime of state $|3\rangle$ (1.54 fs) is similar to the laser pulse duration in the nuclear rest frame. As can be expected, the longer pulse trains reduce the effect of spontaneous emission of the excited state $|3\rangle$ in ^{154}Gd. The target state population is 0.84 for a single pulse pair, 0.93 for $N = 2$ pulse pairs, and just 0.99 for a train of $N = 5$ pulse pairs. We emphasize that in this method after each sequence of pulse pairs, the populations of system are in the stable states $|1\rangle$ and $|2\rangle$ and as a result the time delays between the neighboring pulses will not affect population transfer efficiency. In Table IV we calculate needed intensities at each step of coincident pulses, which is also investigated for the cases of ^{113}Cd, ^{223}Ra, ^{168}Er, and ^{172}Yb and finally leads to complete nuclear coherent population transfer. It should be noted that we present $\tilde{I}_{p(S)}$ ($\tilde{I}_p = I_p^{\mathrm{eff}}/\sin^2\varphi_k$, $\tilde{I}_S = I_S^{\mathrm{eff}}/\cos^2\varphi_k$) for different nuclei in

TABLE IV. The quantum numbers of ground states and peak intensities of pump and Stokes laser pulse calculated in the kth step of coincident pulses.

Nucleus	Quantum Numbers of ground state		Peak intensities of pump and Stokes laser pulse (SXFEL)		Peak intensities of pump and Stokes laser pulse (XFELO)	
	I_1	I_2	$I_p^{\rm eff}/\sin^2\varphi_k({\rm W/cm^2})$	$I_S^{\rm eff}/\cos^2\varphi_k({\rm W/cm^2})$	$I_p^{\rm eff}/\sin^2\varphi_k({\rm W/cm^2})$	$I_S^{\rm eff}/\cos^2\varphi_k({\rm W/cm^2})$
^{223}Ra	3/2	5/2	5.7002×10^{20}	9.0443×10^{20}	5.7001×10^{18}	9.0443×10^{18}
^{113}Cd	11/2	5/2	7.1528×10^{21}	1.1958×10^{24}	7.2771×10^{19}	1.1957×10^{22}
^{97}Tc	1/2	5/2	1.0000×10^{21}	2.3514×10^{22}	1.0049×10^{19}	2.3514×10^{20}
^{154}Gd	0	2	7.8927×10^{19}	1.4175×10^{19}	7.8929×10^{17}	1.4175×10^{17}
^{172}Yb	0	2	1.7923×10^{21}	5.2484×10^{20}	1.7923×10^{19}	5.2484×10^{18}
^{168}Er	0	2	1.0241×10^{21}	7.2024×10^{19}	1.0241×10^{19}	7.2024×10^{17}

Table IV, which are essential parameters [see Eqs. (17)] for the numerical study. So in order to calculate $I_{p(S)}^{\rm eff}$ in the kth step, the values in Table IV must be multiplied by $\sin^2\varphi_k(\cos^2\varphi_k)$ where φ_k for a certain value, N, is given by Eq. (10).

XFELO lasers have long pulse duration compared to SXFEL lasers for small target nucleus accelerations and it is expected that the effect of spontaneous emission which uses XFELO lasers may be more significant than SXFEL lasers. However, for large γ factors both XFELO and the SXFEL pulse durations in the nuclear rest frame will be much shorter than spontaneous decay time.

IV. COMPENSATION OF LASER INTENSITY FLUCTUATIONS BY A TRAIN OF COINCIDENT PULSES

Complete nuclear population transfer by one pair of pulses needs precise adjustment of pump and Stokes laser intensities. Deviations from exact laser intensities which have been calculated in Table IV lead to deviation of transition probability from the desired value. In this section, we show numerically that the coincident pulse technique does not require highly precise control of the pump and Stokes laser intensities by increasing the number of coincident pulse pairs. Based on the description in Sec. II, complete population transfer which uses N pairs of coincident pulses requires that the rms pulse area should be 2π at the end of each sequence. The effective intensities in Table IV satisfy the condition of $A(t)=2\pi$ at the end of each step and thus we numerically study the robustness of this scheme with respect to deviation from effective intensities presented in Table IV. We consider the complete population transfer in ^{97}Tc by using XFELO laser pulses. Figure 5 shows the contour plot of the final population of state $|2\rangle$ as a function of $I_p^{\rm eff}/\sin^2\varphi_k$ and $I_S^{\rm eff}/\cos^2\varphi_k$ for $N=2,5,10$ sequence of pulse pairs. One can notice that

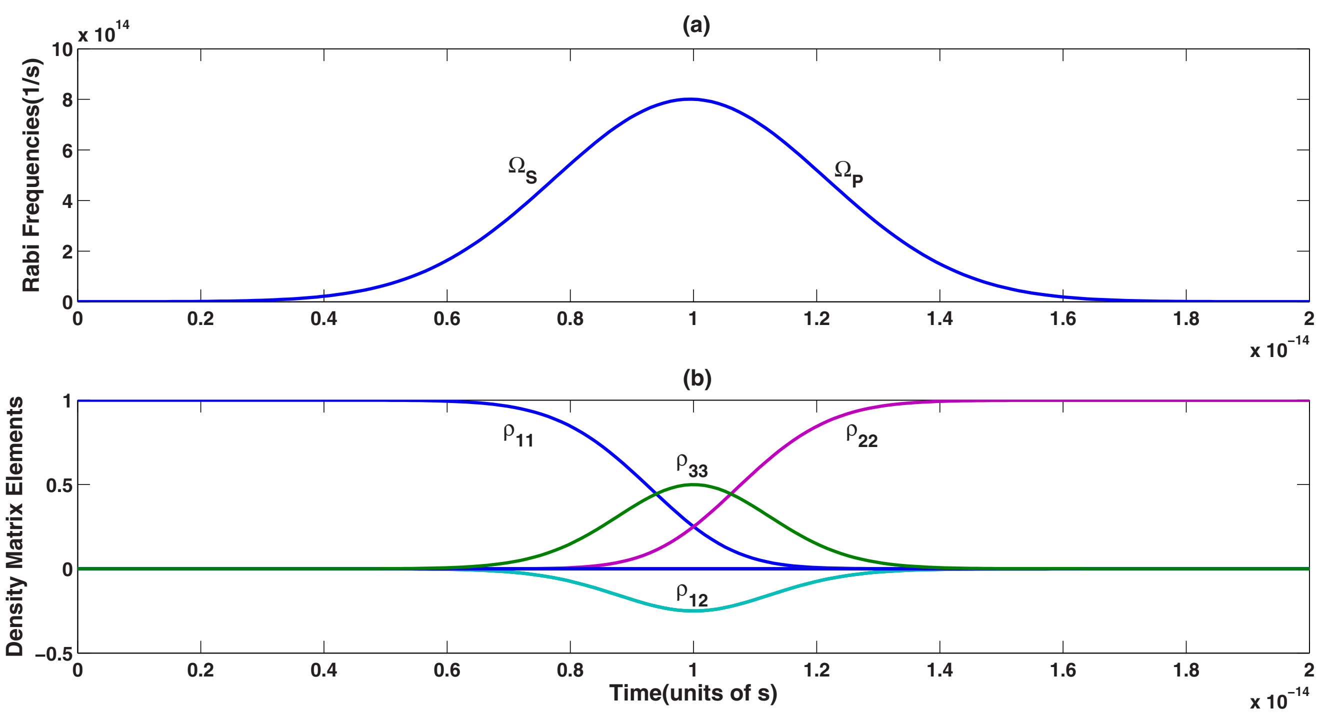

FIG. 3. Rabi frequencies in nuclear rest frame (a) and density matrix elements (b) vs time for ^{97}Tc using SXFEL parameters in master equation (12). As can be seen, complete population transfer occurs using one step of coincident pulses. See Tables II–IV for numerical parameters.

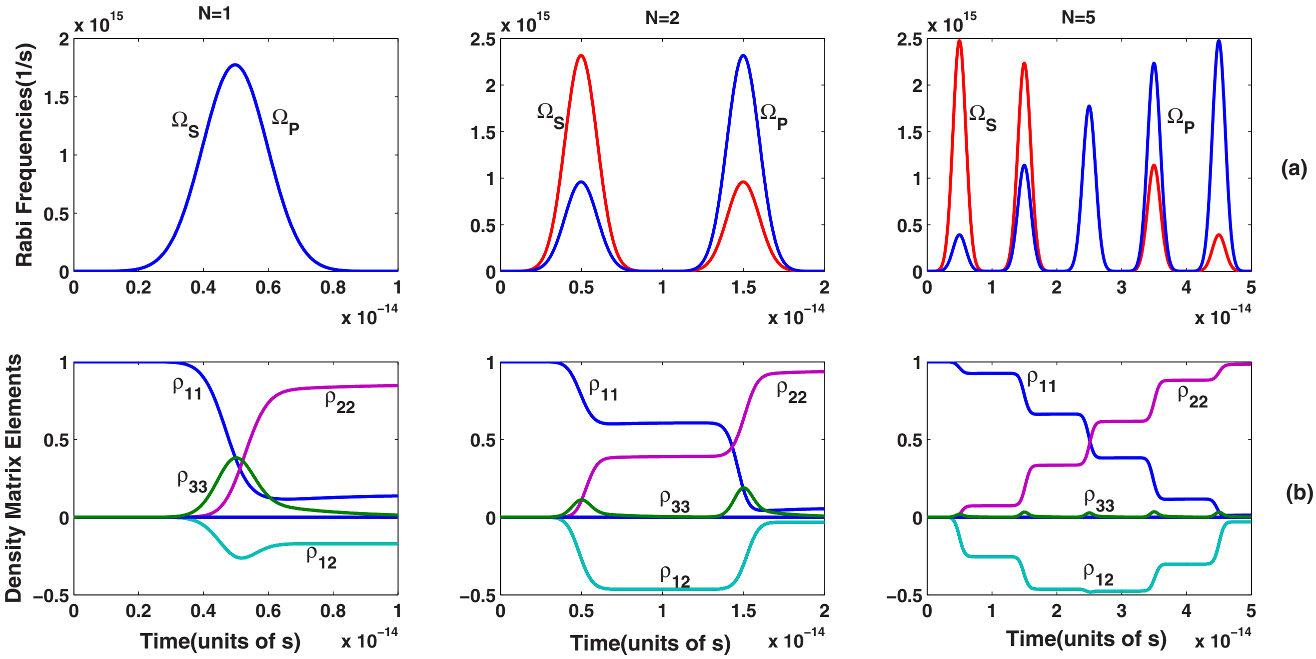

FIG. 4. Rabi frequencies in nuclear rest frame (a) and density matrix elements (b) vs time for ^{154}Gd by using SXFEL parameters in master equation (12). As can be seen, complete population transfer occurs by using five steps of coincident pulses. See Tables II–IV for numerical parameters.

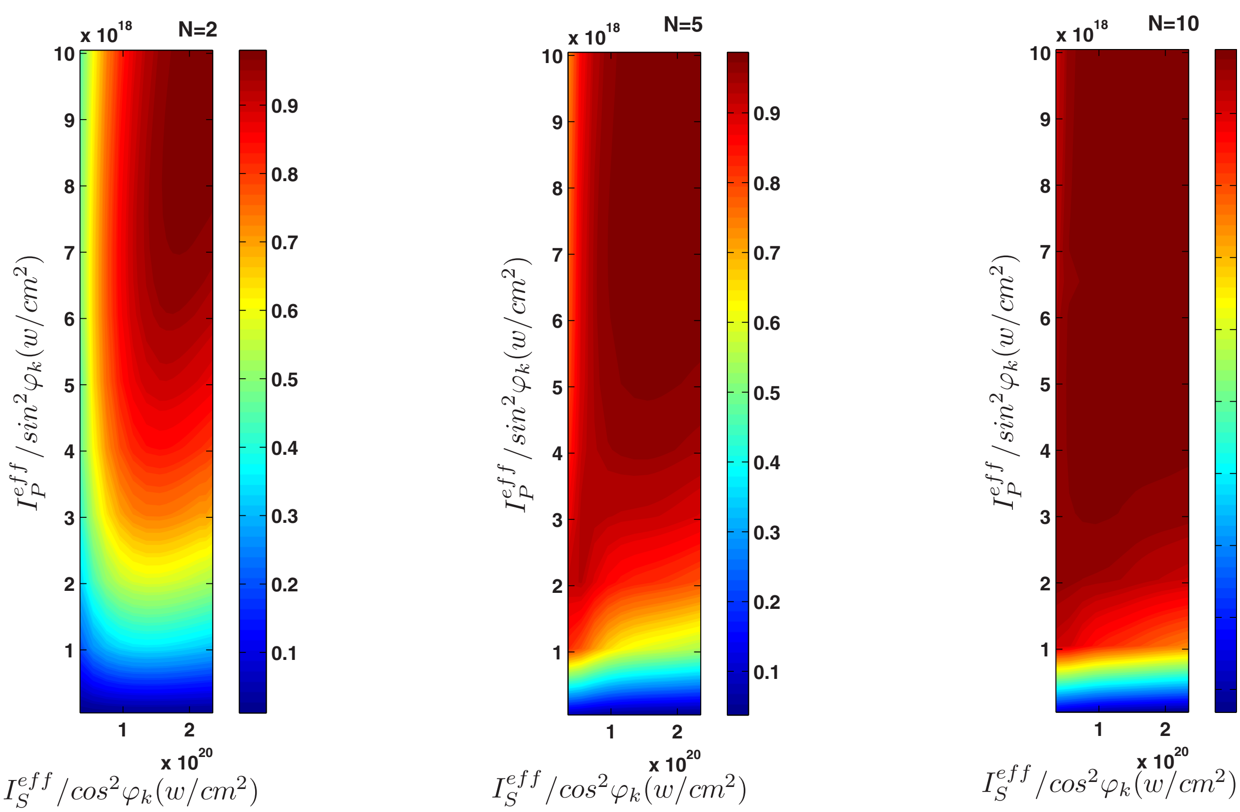

FIG. 5. Contour plot of the final population of state $|2\rangle$ as a function of $I_p^{\mathrm{eff}}/\sin^2\varphi_k$ and $I_S^{\mathrm{eff}}/\cos^2\varphi_k$ for $N = 2,5,10$, a sequence of pulse pairs which is implemented in ^{97}Tc by using XFELO laser parameters.

the robustness of system in deviation from the quantities in Table IV increases by increasing the number of coincident pulses.

V. DISCUSSION AND CONCLUSION

In this research, we have investigated NCPT by a train of pairs of coincident pulses using a collider system which is composed of two fully coherent XFEL (SXFEL and XFELO) beams. The required pump and Stokes laser intensities for different nuclei including ^{223}Ra, ^{113}Cd, ^{97}Tc, ^{154}Gd, ^{172}Yb, and ^{168}Er have been calculated in order to satisfy the coincident-pulse technique conditions. We have shown that this method is not very sensitive to small fluctuations in the intensity of the laser pulses and for the nuclei with short excited-state lifetimes, the effect of spontaneous emission decreases by increasing the number of pulse pairs. The present method has significant potential to create a nuclear battery [40–42], offering clean storage of nuclear energy. Although in this paper complete population transfer between nuclear states has been investigated, the present method can also be used for fractional population transfer between nuclear states which can be important in nuclear quantum information processing. In Eq. (8) if we take $0 < \varphi < \pi/4$, fraction of the population will be transferred from states $|1\rangle$ to $|2\rangle$. In order to implement a train of pairs of coincident pulse in fractional population transfer, for a certain value of φ,φ_k is defined as follows [28]:

$$\varphi_k = \frac{(2k-1)\varphi}{N} \quad (k = 1,2,3,\ldots,N). \tag{21}$$

Using the total evaluation matrix (9), fractional population will be transferred from states $|1\rangle$ to $|2\rangle$ by a train of coincident pulses.

For completion, we give some details about the experimental feasibility of the XFEL train in comparison with STIRAP. STIRAP needs the long pulse in the time scale of the system's evolution; moreover, the STIRAP condition requires a slowly changing field envelope. Because of the ultrashort duration and narrow bandwidth of XFEL lasers in the rest frame, the above conditions are difficult to satisfy with XFEL pulses. The present method does not require the above conditions and can have valuable practice for implementation. In this scheme, we have used a two-color copropagating laser beam setup which has experimental advantages. In the copropagating beam setup, it is estimated that the number of the coherently excited nuclei is significant [20] and can realized experimentally in the near future.

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