Nuclear coherent population transfer with x-ray laser pulses

Wen-Te Liao,* Adriana Pálffy,† and Christoph H. Keitel‡

Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

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Abstract

Coherent population transfer in a nuclear three-level system via stimulated Raman adiabatic passage is studied. To compensate for the lack of γ-ray laser sources, we envisage accelerated nuclei interacting with two copropagating or crossed x-ray laser pulses. The parameter regime for nuclear coherent population transfer using fully coherent light generated by future X-Ray Free-Electron Laser facilities and moderate or strong acceleration of nuclei is determined. We find that the most promising case requires laser intensities of 10^{17}-10^{19} W/cm^2 for complete nuclear population transfer. As relevant application, the controlled pumping or release of energy stored in long-lived nuclear states is discussed.

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*Electronic address: Wen-Te.Liao@mpi-hd.mpg.de

†Electronic address: Palffy@mpi-hd.mpg.de

‡Electronic address: Keitel@mpi-hd.mpg.de

Coherent population transfer in nuclei would be a powerful tool for preparation and detection in nuclear physics, especially for control of energy stored in nuclear states. In atomic physics, controlling matter via laser fields in techniques such as laser cooling [1], optical pumping [2] and stimulated Raman adiabatic passage (STIRAP) [3] has been achieved. The transfer of such schemes to nuclear systems, although encouraged by progress of laser technology, has not been accomplished due to the lack of γ-ray laser sources. The incentive is substantial due to the existence of nuclear isomers—long-lived excited states that can store large amounts of nuclear energy over long periods of time [4]. Control of isomer pumping or depletion is thus related to the concept of nuclear batteries.

To bridge the gap between x-ray laser frequency and nuclear transition energies, a key proposal is combining moderately accelerated target nuclei and novel x-ray lasers [5]. Using this scenario, the interaction of x-ray light from the European X-ray Free Electron Laser (XFEL) [6] with nuclear two-level systems was investigated theoretically [5, 7]. The manipulation of nuclear state population by STIRAP and the possibility of isomer triggering via coherent control have however never been addressed, partially because the poor coherence properties of the XFEL do not allow advanced nuclear quantum optics schemes.

In this Letter we investigate for the first time the nuclear coherent population transfer (NCPT) between two ground states in the Λ-level scheme showed in Figure 1(a) using two overlapping x-ray laser pulses in a STIRAP setup. This is a typical three-level scheme that can lead to the depletion of a metastable state, here the ground state $|1\rangle$, via a triggering level $|3\rangle$ to a level $|2\rangle$ whose decay to the nuclear ground state is no longer hindered by the long-lived isomer. We show that a fully coherent XFEL such as the future XFEL Oscillator (XFELO) [8] or the seeded (two-stage) XFEL (SXFEL) [6, 9–12] to provide both pump and Stokes laser, together with acceleration of the target nuclei to achieve the resonance condition, allow for NCPT. The coherence of the x-ray laser has as a result nuclear coherent control at much lower intensities than previous calculated values for laser driving of nuclear transitions [5], already at 10^{17}-10^{19} W/cm^2. In view of our results, the experimental prospects of isomer depletion are discussed and a setup to produce both pump and Stokes pulses with different frequencies from a single coherent x-ray beam is put forward. Until the first two-color XFEL becomes operational, this method may prove itself useful also for other x-ray multiple beam experiments in the near future.

The interaction of a nuclear Λ-level scheme with the pump laser P driving the $|1\rangle \rightarrow |3\rangle$ transition and the Stokes laser S driving the $|2\rangle \rightarrow |3\rangle$ transition is depicted in Figure 1(a). In STIRAP, the empty $|2\rangle$ and $|3\rangle$ states are first coupled by the Stokes laser, building a superposition of two unpopulated states. Subsequently, the pump laser couples the fully occupied $|1\rangle$ and the pre-built coherence of two empty states. The dark (trapped) state is formed and evolves with the time dependent Rabi frequencies of the pump and Stokes fields Ω_p and Ω_S, respectively [3].

Typically, the Λ-level scheme is not closed, i.e., the population in $|3\rangle$ will not only decay to $|1\rangle$ and $|2\rangle$ but also to other low energy levels through spontaneous radiative decay or by other decay mechanisms such as internal conversion or α decay. This open feature of $|3\rangle$ speaks against direct pumping, allowing us to identify two situations: (i) the lifetime of $|3\rangle$ is longer than the pulse duration. Since the population can stay in $|3\rangle$ long enough, apart from STIRAP, also NCPT via sequential isolated pulses such as π pulses is possible. A first π pulse can pump the nuclei in state $|3\rangle$, followed by a second π pulse of the Stokes laser for the stimulated $|3\rangle \rightarrow |2\rangle$ decay. The latter scenario lacks the robustness of STIRAP, having a sensitive dependence on the laser intensities. (ii) the lifetime of $|3\rangle$ is shorter than the pulse duration. Because of the high decay rate of $|3\rangle$, separated single pulses cannot produce NCPT and STIRAP provides the only possibility for population transfer.

The nuclear excitation energies in the two regimes described above are typically higher than the designed photon energy of the XFELO and SXFEL. Nuclei suitably accelerated can interact with two Doppler-shifted x-ray laser pulses. The two laser frequencies and the relativistic factor γ of the accelerated nuclei have to be chosen such that in the nuclear rest frame both one-photon resonances are fulfilled (multiphoton transitions are substantially less probable). Copropagating laser pulses should have different frequencies in the laboratory frame in order to match the nuclear

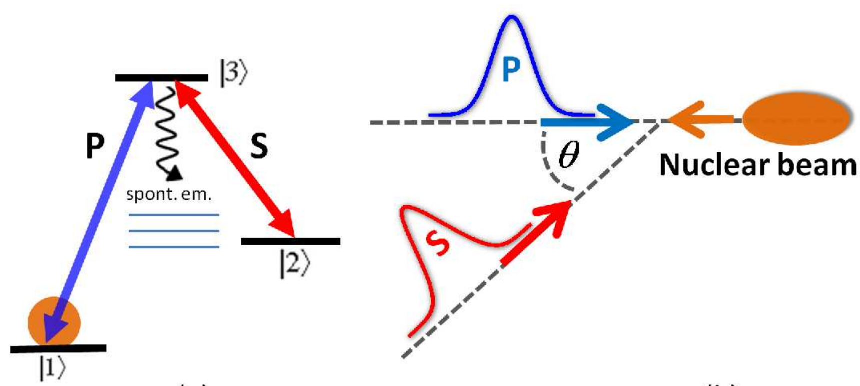

FIG. 1: (a) The nuclear Λ-scheme. The initial nuclear population is concentrated in state $|1\rangle$. The pump laser P drives the transition $|1\rangle \rightarrow |3\rangle$, while the Stokes laser S drives the transition $|2\rangle \rightarrow |3\rangle$). The upper state $|3\rangle$ decays also to other states through spontaneous emission. (b) Two partially overlapping x-ray laser pulses P (pump) and S (Stokes) interact with relativistically accelerated nuclei.

transition energies. To fulfill the resonance conditions with a single-color laser we envisage the pump and Stokes pulses meeting the nuclear beam at different angles, as shown in Figure 1(b).

In general, situation (i) is related to nuclear excitations of tens up to hundreds of keV, such that $\gamma \lesssim 10$. These low-lying levels have however energy widths of about 1 μeV or less, orders of magnitude smaller than the photon energy spread. In this case only a fraction of the incoming photons will drive the nuclear transition, leading to a small effective intensity [7]. For case (ii), the required γ for driving MeV transitions is on the order of 100. Typically, such transitions have widths ($\sim$ 1 eV) larger than the bandwidth of the XFELO or SXFEL. The effective and nominal laser intensity have in this case the same value, an advantage of the high-γ regime. A list of parameters for nuclei with suitable transitions for both (i) and (ii) regimes is presented in Table I.

TABLE I: Nuclear parameters. E_i is the energy of state $|i\rangle$ with $i \in \{1,2,3\}$ (in keV) [13], γ the relativistic factor required to bring the x-ray laser in resonance with the two nuclear transitions and θ the angle between the pump and Stokes pulses as shown in Figure 1(b). $|1\rangle$ is the ground state except for ^{97}Tc where $E_1 = 96.57$ keV.

			(a) SXFEL		(b) XFELO	
Nucleus	E_3	E_2	γ	θ (rad)	γ	θ (rad)
^{185}Re	284.200	125.359	11.5	1.4544	5.7	1.4596
^{97}Tc	656.900	324.476	22.6	1.3836	11.2	1.3848
^{154}Gd	1241.291	123.071	50.1	0.6407	24.8	0.6408
^{168}Er	1786.123	79.804	72.025	0.4260	35.7	0.4260

The theoretical study of the nuclear Λ three-level system interacting with two resonant x-ray lasers relies on the standard quantum optics approach performed in the nuclear rest frame. Considering the three level system denoted in Figure 1(a), the dynamics of the density matrix $\widehat{\rho}$ is governed by the master equation [3, 14] $\frac{\partial}{\partial t}\widehat{\rho} = \frac{1}{i\hbar}\left[\widehat{H},\widehat{\rho}\right] + \widehat{\rho}_{relax}$, with the interaction Hamiltonian

$$\widehat{H} = -\frac{\hbar}{2}\begin{pmatrix} 0 & 0 & \Omega_p \\ 0 & 2\left(\triangle_p - \triangle_S\right) & \Omega_S \\ \Omega_p^* & \Omega_S^* & 2\triangle_p \end{pmatrix}, \tag{1}$$

and the relaxation matrix $\widehat{\rho}_{relax}$ that includes the spontaneous decay. The initial conditions are $\rho_{ij}(0) = \delta_{i1}\delta_{1j}$. In the expression above, $\Delta_{p(S)} = \gamma(1+\beta)\omega_{p(S)} - ck_{31(2)}$ is the laser detuning, where γ and β denote the relativistic factors, $\gamma = 1/\sqrt{1-\beta^2}$, c is the speed of light, $\omega_{p(S)}$ is the pump (Stokes) laser angular frequency and k_{31} and k_{32} are the wave numbers of the corresponding transitions. The slowly varying effective Rabi frequencies $\Omega_{p(S)}(t)$ in the nuclear rest frame for transitions of electric (ε) or magnetic (μ) multipolarity L are given by [3, 7]

$$\Omega_{p(S)}(t) = \frac{4\sqrt{\pi}}{\hbar}\left[\frac{\gamma^2(1+\beta)^2 I_{p(S)}(L+1)B(\varepsilon/\mu\, L)}{c\epsilon_0 L}\right]^{1/2} \times \frac{k_{31(2)}^{L-1}}{(2L+1)!!}\mathrm{Exp}\left\{-\left[\frac{\gamma(1+\beta)(t-\tau_{p(S)})}{\sqrt{2}T_{p(S)}}\right]^2\right\}. \tag{2}$$

Here we have expressed the nuclear multipole moment with the help of the reduced transition probabilities $B(\varepsilon/\mu\, L)$ [7]. All the laser physical quantities have been transformed in Eq. (2) into the nuclear rest frame, leading to the angular frequency $\gamma(1+\beta)\omega_{p(S)}$, bandwidth $\gamma(1+\beta)\Gamma_{p(S)}$, pulse duration $T_{p(S)}/(\gamma(1+\beta))$, and laser peak intensity $\gamma^2(1+\beta)^2 I_{p(S)}$. A further important observation is that if the nuclear width is smaller than the laser bandwidth, only a fraction of the laser photons fulfills the resonance condition. We have therefore considered the effective laser intensity, $I^{\mathrm{eff}}_{p(S)} = I_{p(S)}\Gamma/(\gamma(1+\beta)\Gamma_{p(S)})$, with Γ the nuclear transition width and $\Gamma_{p(S)}$ the laser bandwidth. Further notations used in Eq. (2) are ϵ_0 the vacuum permittivity, $\hbar$ the reduced Planck constant, and $\tau_{p(S)}$ the peak position of the pump (Stokes) laser, respectively.

Unlike in other x-ray techniques such as nuclear forward scattering (NFS), where spatial coherence is required, the most important prerequisite for nuclear STIRAP is the *temporal* coherence of the x-ray lasers. The coherence parameters of the existent XFEL at the Linac Coherent Light Source (LCLS) in Stanford, USA are yet to be tested [11, 15] and the designed coherence time value for the European XFEL is 0.2 fs compared to the pulse duration of 100 fs [6]. The SXFEL, considered as an upgrade for the LCLS and the European XFEL, will deliver completely transversely and temporally coherent pulses, that can reach 0.1 ps pulse duration and about 10 meV bandwidth [10, 12]. Another option is the XFELO that will provide coherent photons up to 25 keV with coherence time on the order of the pulse duration ~ 1 ps, and meV narrow bandwidth [8]. We consider here the laser photon energy for the pump laser fixed at 25 keV for the XFELO and 12.4 keV for the SXFEL. The relativistic factor γ is given by the resonance condition $E_3 - E_1 = \gamma(1+\beta)\hbar\omega_p$. The frequency of the Stokes x-ray laser can be then determined depending on the geometry of the setup. For copropagating pump and Stokes beams (implying a two-color XFEL), the photon energy of the Stokes laser is smaller than that of the pump laser since $E_2 > E_1$. The alternative that we put forward is to consider two crossed laser beams generated by a single-color SXFEL meeting the accelerated nuclei as shown schematically in Figure 1(b). The angle θ between the two beams is determined such that in the nuclear rest frame the pump and

Stokes photons fulfill the resonances with two different nuclear transitions. The separation of the pump and Stokes beams out of the original XFEL beam requires dedicated x-ray optics such as the diamond mirrors [16] developed for the XFELO. X-ray reflections can also help tune the intensity of the two beams.

The relative coherence between the two ground states is crucial for successful NCPT via STIRAP. Since in our case the lifetime of $|2\rangle$ is much longer than the laser pulse durations, decoherence is related to the unstable central frequencies and short coherence times of the pump and Stokes lasers. In similar coherence-sensitive experiments such as STIRAP and electromagnetically-induced transparency in atomic quantum optics, acousto-optical modulators can be used to obtain two coherent beams of different frequencies out of a single one, thus canceling the effects of central frequency and phase jumps in the original laser pulse. For x-ray light, such devices are however not available. Our single-color XFEL crossed-beam setup accommodates the present lack of two-color x-ray coherent sources (only expected as a further upgrade of the LCLS [12]) and reduces the effect of laser central frequency jumps to equal detunings in the pump and Stokes pulses. Variations in detuning up to $\Delta_p = \Delta_S =$10 meV lead to less than 5$\%$ decrease in NCPT. One should mention however that due to time dilatation and pulse delay, a phase jump in the original x-ray beam does not act simultaneously on the pump and Stokes laser in the nuclear rest frame. Coherent population transfer in our setup therefore still requires temporal coherence for the whole pulse duration, as predicted for both SXFEL and XFELO.

In Figure 2 we compare our calculated population transfer for several cases in both regimes (i) and (ii) using XFELO and SXFEL parameters in a crossed-beam single-color XFEL setup with various laser intensities from $I_p = 10^{16}$ to 10^{26} W/cm^2. The optimal set of parameters is obtained by a careful analysis of the dependence between pump laser intensity and pulse delay $\tau_p - \tau_s$. For each value of the pump intensity the pulse delay is chosen such that the NCPT reaches its maximum. For regime (i) we considered the lowest three nuclear levels of ^{185}Re, with relativistic factors and Stokes beam crossing angles listed in Table I. The ^{185}Re nuclei start to be channeled at about $I_p = 10^{22}$ W/cm^2 (XFELO) and $I_p = 10^{25}$ W/cm^2 (SXFEL). NCPT is achieved here via sequential π pulses. At the exact π-pulse value of the pump intensity a peak in the population transfer for ^{185}Re can be observed, at $I_p = 6\times10^{25}$ W/cm^2 in Figure 2(a) and $I_p = 6\times10^{22}$ W/cm^2 in Figure 2(b). For higher intensities, oscillations become visible in Figure 2(b) until a plateau indicating NCPT via STIRAP is reached.

For case (ii), we present our results for ^{154}Gd and ^{168}Er, both requiring stronger nuclear acceleration with γ factors between 24 and 72 and fs pulse delays. The ^{154}Gd ground state population

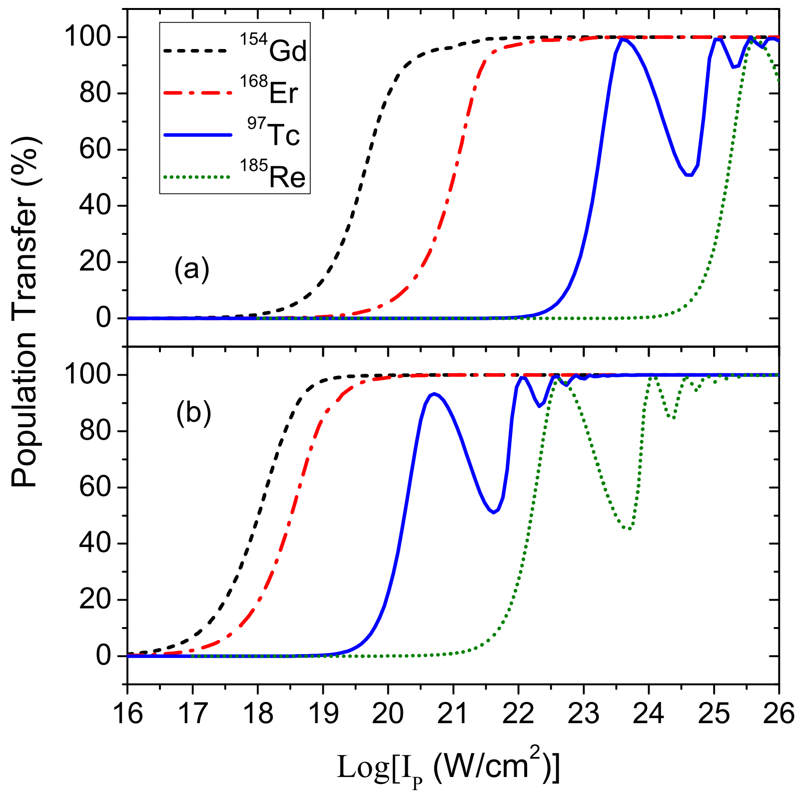

FIG. 2: NCPT for several nuclei as a function of the pump laser intensity using (a) SXFEL and (b) XFELO parameters in a crossed-beams setup as sketched in Fig. 1(b). The Stokes laser intensities were chosen $I_S = 0.02 I_p$ for ^{185}Re, $I_S = 0.33 I_p$ for ^{168}Er, $I_S = 0.89 I_p$ for ^{154}Gd and $I_S = 20.81 I_p$ for ^{97}Tc, respectively, according to the π pulse intensity ratios I_S^{π}/I_p^{π} . All detunings are $\triangle_p = \triangle_S = 0$. See discussion in the text and Table I for further parameters.

starts to be coherently channeled at about $I_p = 10^{17}$ W/cm^2 using XFELO and $I_p = 10^{19}$ W/cm^2 using SXFEL parameters, respectively. Up to $I_p = 10^{19}$ W/cm^2 (XFELO) and $I_p = 10^{21}$ W/cm^2 (SXFEL), more than 95$\%$ of the nuclei reach $|2\rangle$. In this case π pulses cannot provide the desired NCPT. The calculated intensities necessary for complete NCPT are within the designed intensities of the XFEL sources. Considering the operating and designed peak power of 20-100 GW [6, 10–12] for SXFEL (and about three orders of magnitude less for XFELO) and the admirable focus achieved for x-rays of 7 nm [17], intensities could reach as high as $10^{17} - 10^{18}$ W/cm^2 for XFELO [8] and $10^{21} - 10^{22}$ W/cm^2 for SXFEL [10].

One of the most relevant applications of NCPT is isomer pumping or depletion. In Figure 2 we present our result for NCPT for ^{97}Tc. The ^{97}Tc isomer lies at $E_1 = 96.57$ keV and has a half life of $\tau_1 = 91$ d. The intensities for which complete isomer depletion is achieved by STIRAP using SXFEL for ^{97}Tc are $I_p = 4 \times 10^{23}$ W/cm^2 and $I_S = 8 \times 10^{24}$ W/cm^2. These values are shifted by about three orders of magnitude towards lower intensities when considering the XFELO parameters. Compared to the case of high-energy nuclear transitions (ii), the intensities required for isomer depletion are in this case large, mainly due to the narrow transition width of state $|3\rangle$. Typically, triggering levels high above isomeric states are less well known. A detailed analysis of nuclear data in the search for the best candidate is required for successful isomer depletion.

NCPT is sensitive to the fulfillment of the resonance condition. This involves on the one hand precise knowledge of the nuclear transition energy and on the other hand good control of laser frequency and therefore nuclear acceleration. The former is usually attained in NFS by scanning first for the position of the nuclear resonance. In our setup, the relativistic factor γ influences the detunings and the effective pump and Stokes intensities and Rabi frequencies. For narrow-width excitations (i) it is necessary to first find the laser bandwidth window of the nuclear transition, since most of the transition energy values are not known with such precision. Once found, our procedure of considering an effective intensity which is scaled according to the number of resonant photons should provide the correct approach for a zero-detuning situation. For the case (ii) where the MeV nuclear transitions have eV widths, it is only necessary to tune the laser photons in the corresponding energy window. Especially for low-energy excitations, NCPT can thus be used for determination of nuclear transition energies. For instance, STIRAP from the ground state to the low-lying metastable state at 7.6 eV in ^{229}Th [18] via the 29.192 keV level could provide very precise transition energy values and help investigate the isomer properties.

Powerful ion accelerators are the key issue for achieving NCPT. In the low γ region, the forthcoming FAIR at GSI will provide high quality ion beams with energies up to 45 GeV/u [19]. The corresponding γ limit is about 48 and the precision $\Delta E/E \sim 2 \times 10^{-4}$. For the high γ region, the Large Hadron Collider (LHC) is currently the only suitable ion accelerator which can accelerate ^{208}Pb^{82+} up to $\gamma = 2963.5$ with low energy spread of about 10^{-4} [20]. LHC can also accelerate lighter ions to energies larger than 100 GeV [21]. Such acceleration can bridge the gap in energy between nuclear transition and x-ray photons as discussed in this work. For the strong acceleration regime, the resonance condition corresponds to an energy spread of the ion beam of 10^{-5}. This issue becomes more problematic for NCPT of nuclei in the moderate acceleration regime where the resonance condition requires a more precise γ value, $\Delta\gamma/\gamma = 10^{-6}$. A further study of the overlap efficiency for the laser beams and ion bunches shows that the copropagating laser beams setup is more advantageous. Using LHC beam size parameters [20] and a 10 μm focusing of the

XFEL beam, we estimate that for copropagating laser beams up to 10^5 nuclei meet the laser focus per bunch and laser pulse, while for crossed laser beams this number reduces to 30.

X-ray coherent light sources are not available today at the few large ion acceleration facilities. At present a case study for a low-energy ion beam at the European XFEL is in progress [22]. Furthermore, the new materials research center MaRIE in Los Alamos, USA, is also envisaged to have high-energy, high-repetition-rate, coherent x-ray capability along with accelerated charged-particle beams [23]. On the other hand, table-top solutions for both ion acceleration and x-ray coherent light would facilitate the experimental realization of isomer depletion in NCPT. Table-top x-ray undulator sources are already operational [24], with a number of ideas envisaging compact x-ray sources and table-top FELs [25, 26]. In conjunction with the crystal cavities that are designed to provide the XFELO with its remarkable coherence [8, 16], such table-top devices have the potential to become a key tool for the release on demand of energy stored in nuclei at large ion accelerator facilities. Alternatively, the use of table-top ion accelerators that rely on laser acceleration presents another option. For now however the shaped-foil-target ion accelerators [27] or radiation pressure dominant acceleration [28] do not provide the necessary stability and monochromaticity.

In conclusion, the parameter regime for which fully coherent x-ray laser pulses can induce population transfer between nuclear levels matches the predicted values for the envisaged XFELO and SXFEL facilities. Realization of NCPT and the future of nuclear batteries thus rely on the development of x-ray coherent sources and perhaps on high-precision table-top solutions for lasers and ion accelerators to be flexibly used in any location around the globe.

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