Nuclear coherent population transfer with x-ray laser pulses

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Abstract

Coherent population transfer between nuclear states using x-ray laser pulses is studied. The laser pulses drive two nuclear transitions between three nuclear states in a setup reminding of stimulated Raman adiabatic passage used for atomic coherent population transfer. 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.

Keywords: isomer decay, gamma transitions and levels, nuclear quantum optics, x-ray and gamma-ray lasers

Long-lived excited nuclear states, also known as isomers, can store large amounts of energy over longer periods of time. Isomer depletion, i.e., release on demand of the energy stored in the metastable state, has received great attention in the last one and a half decades, especially related to the fascinating prospects of nuclear batteries [1, 2, 3, 4]. Depletion occurs when the isomer is excited to a higher level, which is associated with freely radiating states and therefore releases the energy of the metastable state. Coherent population transfer between nuclear states would therefore not only be a powerful tool for preparation and detection in nuclear physics, but also especially useful for control of energy stored in isomers.

In atomic physics, a successful and robust way for atomic coherent population transfer is the stimulated Raman adiabatic passage (STIRAP) [5], a technique in which two coherent fields couple to a three-level system. 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 pursuit of coherent sources for wavelengths around or below 1 Å is supported however by the advent and commissioning of x-ray free electron lasers, the availability of which will stimulate the transfer of quantum optical schemes to nuclei.

To bridge the gap between x-ray laser frequency and nuclear transition energies, a key proposal is to combine moderately accelerated target nuclei and novel x-ray laser schemes [6]. Using this scenario, the interaction of x-ray from the European X-Ray Free Electron Laser (XFEL) [7] with nuclear two-level systems was studied theoretically [6, 8]. The manipulation of nuclear state population by STIRAP and the coherent control of isomers have however never been addressed, partially because of the poor coherence properties of the XFEL.

In this Letter we investigate for the first time the nuclear coherent population transfer (NCPT) between the two lower states in the nuclear A-level scheme showed in Fig. 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 using a fully coherent XFEL such as the future XFEL Oscillator (XFELO) [9] or the seeded XFEL (SXFELO) [10] for both pump and Stokes lasers, 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 [6], already

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at $10^{17}$–$10^{19}$ W/cm². In view of our results, the indeed challenging experimental prospects of isomer depletion are discussed and a setup to produce both pump and Stokes pulses with different frequencies in the nuclear rest frame from a single coherent x-ray beam is put forward.

The interaction of a nuclear A-level scheme with the pump laser $P$ driving the $|1⟩ \rightarrow |3⟩$ transition and the Stokes laser $S$ driving the $|2⟩ \rightarrow |3⟩$ transition is depicted in Fig. 1(a). In STIRAP, at first the Stokes laser creates a superposition of the two unpopulated states $|2⟩$ and $|3⟩$. Subsequently, the pump laser couples the fully occupied $|1⟩$ and the pre-built coherence of the 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 [5].

Typically, the A-level scheme is not closed, i.e. the population in $|3⟩$ will not only decay to $|1⟩$ and $|2⟩$ but also to other low energy levels through spontaneous radiative decay or by other decay mechanisms such as internal conversion or alpha decay. This open feature of $|3⟩$ speaks against direct pumping, allowing us to identify two situations: (i) the lifetime of $|3⟩$ is longer than the pulse duration. Since the nucleus can stay in $|3⟩$ long enough, apart from STIRAP, also NCPT via sequential isolated pulses such as π pulses such as STIRAP, at first the Stokes laser creates a superposition of the two unpopulated states $|2⟩$ and $|3⟩$. A first π pulse can pump the nuclei from $|1⟩$ to $|3⟩$, followed by a second Stokes π pulse that drives the $|3⟩ \rightarrow |2⟩$ decay. The latter scenario lacks the robustness of STIRAP, having a sensitive dependence on the laser intensities. (ii) the lifetime of $|3⟩$ is shorter than the pulse duration. Because of the high decay rate of $|3⟩$, 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 XFEL 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. Copropagating laser pulses (with $θ_p = 0$ in Fig. 1(b)) should have different frequencies in the laboratory frame in order to match the nuclear 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 ($θ_p \neq 0$, as shown in Fig. 1(b).)

In general, situation (i) is related to nuclear excitations of tens up to hundreds of keV, such that $γ \lesssim 10$. These low-lying levels have however energy widths of about 1 eV 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 [3]. For case (ii), the required $γ$ for driving MeV transitions is on the order of $20 – 100$. Typically, such transitions have widths ($\sim 1$ eV) larger than the bandwidth of the XFEL 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 a number of nuclei with suitable transitions for both (i) and (ii) regimes is presented in Tables 1 and 2.

We study the dynamics of the system depicted in Fig. 1(a) in the nuclear rest frame. This is governed by the master equation for the density matrix $\hat{ρ}$ [14] that reads $\frac{d\hat{ρ}}{dt} = i [\hat{H}, \hat{ρ}] + \hat{ρ}_{relax}$, with the interaction Hamiltonian

$$\hat{H} = -\frac{\hbar}{2} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \Omega_p^* \\ \Omega_p & \Omega_S & \Omega_{S\delta} \end{pmatrix},$$

and the relaxation matrix $\hat{ρ}_{relax}$ that includes the spontaneous decay. The initial conditions are $ρ_j(0) = δ_{j1}$, $j = 1, 2, 3$. In the expression above, $Δ_{pS} = γ [1 + β cos θ] ω_{pS} - c k_{312} γ$ is the laser detuning, where $γ$ and $β$ denote the relativistic factors, $γ = 1/\sqrt{1 - β^2}$, $c$ is the speed of light, $ω_{pS}$ is the pump (Stokes) laser angular frequency and $k_{31}$ and $k_{32}$ are the wave numbers of the corresponding transitions. The angle $θ$ is zero for the pump laser and $θ = θ_p$ for the Stokes laser. The slowly varying effective Rabi frequencies $Ω_{pS}(t)$ of the nuclear rest frame for nuclear transitions of electric ($e$) or magnetic ($μ$) multipolarity $L$ are given by [3–8]

$$Ω_{pS}(t) = \frac{4 \sqrt{π}}{\hbar} \left( γ [1 + β cos θ] ω_{pS} (L + 1) B(e/μ L) \right)^{1/2} c e L \times \left[ \frac{k_{31}^2 (2L + 1)!}{\sqrt{2π e^2 L}} \right] \exp \left\{ - \left[ γ [1 + β cos θ] (1 - τ_{pS}) \right]^2 \right\}.$$

Here we have expressed the nuclear multipole moment with the help of the reduced transition probabilities $B(e/μ L)$ following the approach developed in [8]. This allows for a unified treatment of the laser-nucleus interaction for both dipole-allowed ($L = 1$) and dipole-forbidden nuclear transitions. All the laser quantities have been transformed in Eq. 3 into the nuclear rest frame, leading to the angular frequency $γ [1 + β cos θ] ω_{pS}$, bandwidth $γ [1 + β cos θ] Γ_{pS}$, pulse duration $Γ_{pS}$, and laser peak intensity $Γ_{pS}$. Furthermore, the effective laser intensity has been taken into account $I_{relax} = I_{pS} Γ_{pS}$, with $Γ$ the nuclear transition width and $Γ_{pS}$ the laser bandwidth. Further notations used in Eq. 2 are $e_0$ the vacuum permittivity, $h$ the reduced Planck constant, and $τ_{pS}$ the temporal peak position of the pump (Stokes) laser, respectively.

In the following we address the laser beam parameter requirements. The most important prerequisite for nuclear STIRAP is the temporal coherence of the x-ray lasers. The coherence time of the existent XFEL at the Linac Coherent Light
The set of laser parameters is obtained by a careful analysis of the pulse duration of 100 fs, much shorter than the pulse duration of 1 ps and 10 meV bandwidth [12, 15, 16]. Another option is the XFELO that will provide coherence time on the order of the pulse duration ~1 ps, and meV narrow bandwidth [9]. 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 \( E_3 - E_1 = \gamma(1 + \beta)\omega_0 \). The frequency of the Stokes x-ray laser can then be determined depending on the geometry of the setup. For copropagating pump and Stokes beams (implying a two-color XFELO), 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 Fig. [1]b. The angle \( \theta_s \) 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 values of \( \gamma, E_3, \) and \( \theta_s \) for NCPT for the nuclear systems under consideration are given in Table [1].

### Table 1: Nuclear Parameters

| Nucl. | \( E_3 \) | \( E_2 \) | \( \epsilon/\mu L \) | \( B(\epsilon/\mu L) \) (wsu) |
|-------|-------|-------|----------------|------------------|
| \( ^{185}\text{Re} \) | 284 | 125 | 6.4 \times 10^{-3} | 3.7 \times 10^{-4} |
| \( ^{97}\text{Tc} \) | 657 | 324 | 5 \times 10^{-2} | 6.7 \times 10^{-5} |
| \( ^{154}\text{Gd} \) | 1241 | 123 | 4.4 \times 10^{-2} | 4.9 \times 10^{-2} |
| \( ^{168}\text{Er} \) | 1786 | 79 | 3.2 \times 10^{-3} | 9.1 \times 10^{-3} |

For regime (i) that allows NCPT via both \( \pi \) pulses and STIRAP, we considered the lowest three nuclear levels of \( ^{185}\text{Re} \). In the crossed-beam setup, NCPT is achieved at lower intensities via sequential \( \pi \) pulses. At the exact \( \pi \)-pulse value of the pump intensity, a peak in the nuclear population transfer for \( ^{185}\text{Re} \) can be observed, at \( I_p = 6 \times 10^{25} \text{ W/cm}^2 \) (Fig. [2]a) and \( I_p = 6 \times 10^{22} \text{ W/cm}^2 \) (Fig. [2]b). With increasing \( I_p \) in the crossed-beam setup (Fig. [2]a), the \( ^{185}\text{Re} \) nuclei are only partially excited to state \( |2\rangle \) and the NCPT yield starts to oscillate. The amplitude and frequency of the oscillations are varying as a result of our pulse delay optimization procedure. At sufficient intensities in the pulse overlap regime STIRAP becomes preferable as compared to the \( \pi \) pulses mechanism due to the lack of oscillations. The plateau at 100% population transfer indicates that NCPT via STIRAP alone is reached. In the two-color copropagating beams scheme (Fig. [2]c,d), the pulse shape of pump and that of Stokes are the same in the nuclear rest frame. This renders STIRAP more efficient and thus preferable compared to the single-color setup, as the STIRAP plateau can be reached with lower laser intensities.

For case (ii), we present our results for \( ^{154}\text{Gd} \) and \( ^{168}\text{Er} \), that require stronger nuclear acceleration with \( \gamma \) factors between 24 and 72 and fs pulse delays. The \( ^{154}\text{Gd} \) ground state population starts to be coherently channeled at about \( I_p = 10^{17} \text{ W/cm}^2 \) using XFELO and \( I_p = 10^{19} \text{ W/cm}^2 \) using SXFEL parameters, respectively. Up to \( I_p = 10^{19} \text{ W/cm}^2 \) (XFELO) and \( I_p = 10^{21} \text{ W/cm}^2 \) (SXFEL), more than 95% of the nuclei reach \( |2\rangle \).

In this case \( \pi \) pulses cannot provide the desired NCPT due to the fast spontaneous decay of state \( |3\rangle \) in neither copropagating-nor cross-beam setups. The calculated intensities necessary for complete NCPT are within the designed intensities of the XFELO sources. Considering the operating and designed peak power of 20-100 GW [4, 12, 15, 16] 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 \( 10^{17} - 10^{18} \text{ W/cm}^2 \) for XFELO [3] and \( 10^{21} - 10^{22} \text{ W/cm}^2 \) for SXFEL [1].

One of the most relevant applications of NCPT is isomer pumping or depletion. In Fig. [2] we present our results for NCPT in \(^{97}\text{Tc} \) nuclei starting from the \( E_1 = 96.57 \text{ keV} \) iso-
Figure 2: NCPT for several nuclei as a function of the pump laser intensity using SXFEL (a,c) and XFEL (b,d) parameters. For the crossed-beams setup (a) and (b), the Stokes laser intensities were chosen $I_S = 0.02I_p$ for $^{185}$Re, $I_S = 0.34I_p$ for $^{168}$Er, $I_S = 0.81I_p$ for $^{154}$Gd and $I_S = 20.82I_p$ for $^{97}$Tc, respectively, according to the $\pi$ pulse intensity ratios $I_p/I_S$. The inset in (b) depicts the wave-front form necessary to extend the spatial overlap region of the laser and ion beams where STIRAP may occur. In the two-color setup (c) and (d), $I_S = 0.03I_p$ for $^{185}$Re, $I_S = 0.39I_p$ for $^{168}$Er, $I_S = 0.90I_p$ for $^{154}$Gd and $I_S = 35.93I_p$ for $^{97}$Tc. All detunings are $\delta_p = \delta_S = 0$. See discussion in the text and Tables 1 and 2 for further parameters.
and ion bunches shows that the copropagating laser beams setup is more advantageous. Using LHC beam size parameters \([21]\) and a 10 \(\mu\)m radius of the XFEL focusing spot, we estimate that for copropagating laser beams up to 3 \(\times\) \(10^5\) nuclei meet the laser focus per bunch and laser pulse, while for crossed laser beams this number reduces to 80 for the smallest overlap volume \(\text{at } \theta_3 = 90^\circ\). The extreme temporal and spatial fine-tuning required to match the overlaps of a bunched ion beam with the two laser beams in the crossed-beam setup is however at present challenging. A continuous ion beam, on the other hand, has the disadvantage of much lower ion density at the overlap with the pump and Stokes beam and of no possibility to control when the ions pass through the overlap region. Furthermore, the necessary time delay between pump and Stokes and the adiabaticity condition \(\Omega_{pl}\Delta\tau \gg 1\) \([5]\) for STIRAP will be in this case only fulfilled for ions at the diagonal line of the overlap area. In order to maintain the pulse delay and the adiabaticity condition for the whole overlap region with the nuclear beam, a special laser pulse front as presented in the inset Fig. 2(b) is required. With optical lasers, such a design can be achieved with the help of dispersive glass or specially-shaped mirrors, that could also be developed for x-rays \([23]\). We conclude therefore that for a number of technical and conceptual difficulties, the two-color copropagating beams scheme might have better chances to be realized experimentally in the near future.

X-ray coherent light sources are not available today at the few large ion acceleration facilities. At present a new materials research center MaRIE (Matter-Radiation Interactions in Extreme) providing both a fully coherent XFEL with photon energy up to 100 eV and accelerated charged-particle beams is envisaged in the USA \([24]\). In addition, the photonuclear physics pillar of the Extreme Light Infrastructure (ELI) in construction in Romania can provide simultaneously a compact XFEL as well as ion acceleration reaching up to 4-5 GeV \([25]\). At ELI, the combination of coherent gamma-rays and acceleration of the nuclear target are already under consideration for nuclear resonance fluorescence experiments \([25]\). Furthermore, ELI is also envisaged to deliver coherent gamma rays with energies of few MeV \([25]\), which could be used for direct photoexcitation of giant dipole resonances \([26]\).

Table-top solutions for both ion acceleration and x-ray coherent light would facilitate the experimental realization of isomer depletion in NCPT and nuclear batteries. Table-top x-ray undulator sources are already operational \([27]\), with a number of ideas envisaging compact x-ray FELs \([28, 29]\). Rapid progress spanning five orders of magnitude increase in the achieved light brightness within only two years has been reported \([30, 31]\). In conjunction with the crystal cavities designed for the XFELO, 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 exciting forecast of compact shaped-foil-target ion accelerators \([32]\) and radiation pressure acceleration \([33]\) together with microlens beam focusing \([3-4]\) are likely to provide a viable table-top solution to be used together with the existing large-scale XFELs.

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. The challenge for the experimental realization of NCPT and the future of nuclear batteries thus rely especially on the development of x-ray coherent sources and their conjuncture with ion accelerators, perhaps making use of high-precision table-top solutions for lasers and ion accelerators to be flexibly used at any location around the globe.

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