

# THE POTENTIAL USES OF X-RAY FELS IN NUCLEAR STUDIES

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## INTRODUCTION

The intent of this study is to theoretically extend the territory of quantum optics to the fields of nuclear physics [1–3] and x-ray optics [4]. Typically, nuclear transition have energies above 10 keV and a very narrow linewidth, rendering optical lasers unsuitable to control nuclei. However, the novel X-ray Free Electron Laser (XFEL) [5, 6] delivers very bright hard x-rays providing completely new opportunities to study the light-nucleus interaction. Encouraged by this coming revolution, in the first part of this work the technique of stimulated Raman adiabatic passage and the two  $\pi$ -pulse method are investigated in the context of controlling the nuclear population with two Lorentz boosted XFEL pulses. In the second part of this work we consider another promising aspect of novel coherent x-ray sources, namely that the spot size of a tightly focused hard x-ray beam can be essentially smaller than a single atom. Thus, using hard x-ray photons as the information carriers for the future photonic circuits may lead to sub-nm architectures. As the first step, the coherent control of single hard x-ray photons with the help of the 14.4 keV transition of the  $^{57}\text{Fe}$  Mössbauer nucleus is addressed in what follows.

## NUCLEAR COHERENT POPULATION TRANSFER

Coherent control of nuclear states remains so far challenging [7–9]. Typically, the traditional way of shifting nuclei from one internal quantum state to another is by incoherent photon absorption, i.e., incoherent  $\gamma$ -rays (usually bremsstrahlung) illuminate the nuclear sample and excite the nuclei to some high-energy states. Subsequently, some of the excited nuclei may decay to the target state by chance, according to the corresponding branching ratio. This kind of method is rather passive, and its efficiency is low. Encouraged by the development of the XFEL, a promising setup for nuclear coherent population transfer in a three-level system using the quantum optics technique of stimulated Raman adiabatic passage (STIRAP) [10] has been proposed [11, 12]. Two overlapping x-ray laser pulses drive two nuclear transitions and allow for coherent population transfer directly between the nuclear states of interest without loss via incoherent processes. This would enable actively manipulating the nuclear state by using coherent hard x-ray photons and lay an important milestone in the new developing field of nuclear quantum optics [13].

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Efficient control of the nuclear population dynamics in a three-level system as the one shown in Fig. 1 (also referred to as  $\Lambda$ -type system) is in particular interesting due to its association to level schemes necessary for isomer depletion. Nuclear metastable 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 isomers, has received a lot of attention in the last one and a half decades, especially related to the fascinating prospects of nuclear batteries [14]. As shown in Fig. 1, by shining only the pump radiation pulse, depletion occurs when the nuclear population in isomer state  $|1\rangle$  is excited to a higher triggering level  $|3\rangle$  whose spontaneous decay to other lower levels, e.g., state  $|2\rangle$  is no longer hindered by the long-lived isomer. However, such nuclear state control is achieved by incoherent processes (spontaneous decay) and its efficiency is therefore low. In this summary we consider the efficient coherent nuclear population transfer setup proposed in Ref. [11, 12]. Two x-ray laser pulses, the pump and the Stokes pulse, drive the two nuclear transitions  $|1\rangle \rightarrow |3\rangle$  and  $|2\rangle \rightarrow |3\rangle$ , respectively. Since most of the nuclear transition energies are higher than the energies of the currently available coherent x-ray photons, an accelerated nuclear target is envisaged, i.e., a nuclear beam produced by particle accelerators [13]. This allows for a match of the x-ray photon and nuclear transition frequency in the nuclear rest frame.

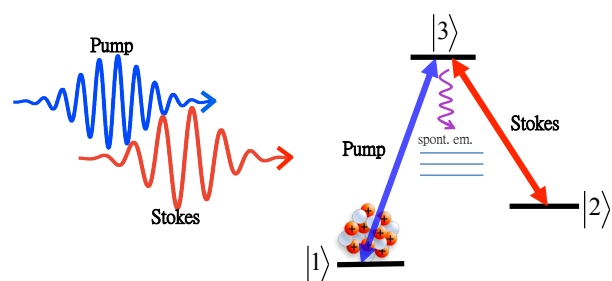


Figure 1: The  $\Lambda$ -level scheme. The blue arrow illustrates the pump pulse, and the red arrow depicts the Stokes pulse. All initial population is in state  $|1\rangle$ .

### STIRAP versus $\pi$ Pulses

The interaction of a  $\Lambda$ -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 Fig. 1. In STIRAP, at first the Stokes laser creates a superposition

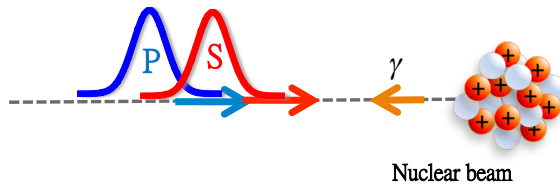


Figure 2: Two-color scheme (copropagating-beams) in the lab frame. The angular frequency of pump laser  $\omega_p$  is different from that of the Stokes laser  $\omega_s$ . The nuclear beam is accelerated such that  $\gamma(1 + \beta)\omega_{p(s)} = ck_{31(2)}$  is fulfilled. Here  $\gamma$  is the relativistic factor,  $\beta$  is the nuclear velocity in the unit of speed of light  $c$  and  $k_{31(2)}$  is the wave number of  $|1\rangle \rightarrow |3\rangle$  ( $|2\rangle \rightarrow |3\rangle$ ) transition.

of the two unpopulated states  $|2\rangle$  and  $|3\rangle$ . Subsequently, the pump laser couples the fully occupied  $|1\rangle$  and the pre-built coherence of the two empty states. If the two fields are sufficiently slowly varying and fulfill the adiabaticity condition, the dark (trapped) state

$$|D\rangle = \frac{\Omega_S(t)}{\sqrt{\Omega_p^2(t) + \Omega_S^2(t)}}|1\rangle - \frac{\Omega_p(t)}{\sqrt{\Omega_p^2(t) + \Omega_S^2(t)}}|2\rangle \quad (1)$$

is formed and evolves with the time-dependent pump and Stokes Rabi frequencies  $\Omega_p(t)$  and  $\Omega_S(t)$ , respectively [10]. Obviously, one can control the populations in the states  $|1\rangle$  and  $|2\rangle$  via temporally adjusting the laser parameters, e.g., the laser electric field strengths of pump and Stokes.

Another option for achieving the coherent population transfer is utilizing so-called  $\pi$ -pulses. Using the scheme in Fig. 1, let us consider the interaction of a two-level system with a single-mode laser, driving the  $|1\rangle \rightarrow |3\rangle$  transition by the pump laser (for now we temporarily neglect state  $|2\rangle$  and the Stokes laser). The resulting state of this system is

$$|\psi\rangle = \cos\left(\frac{1}{2}\int_{-\infty}^t \Omega_p(\tau) d\tau\right)|1\rangle + \sin\left(\frac{1}{2}\int_{-\infty}^t \Omega_p(\tau) d\tau\right)|3\rangle. \quad (2)$$

Obviously, the complete coherent population transfer happens when

$$\int_{-\infty}^t \Omega_p(\tau) d\tau = n\pi \quad (3)$$

for  $n$  odd. Because of this particular case,  $\Omega_p(\tau)$  is called a  $\pi$ -pulse if  $\int_{-\infty}^{\infty} \Omega_p(\tau) d\tau = \pi$ . In the scheme in Fig. 1, one can shine a pump  $\pi$ -pulse and subsequently a Stokes  $\pi$ -pulse on the target to coherently channel all population from state  $|1\rangle$  to state  $|2\rangle$  via the intermediate state  $|3\rangle$ . This technique is termed as two  $\pi$ -pulses method.

### Three-beam Setup

We study the collider system depicted in Fig. 2, composed of an accelerated nuclear beam that interacts with

two incoming XFEL pulses. The nuclear excitation energies are typically higher than the designed photon energy of the XFEL and SXFEL. The accelerated nuclei can interact with two Doppler-shifted x-ray laser pulses as showed in Fig. 2. The two laser frequencies and the relativistic factor  $\gamma$  of the accelerated nuclei have to be chosen such that in the nuclear rest frame both one-photon resonances are fulfilled. Copropagating laser pulses should have different frequencies in the laboratory frame in order to match the nuclear transition energies.

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 Source (LCLS) in Stanford, USA and of the European XFEL are on the order of 1 fs, much shorter than the pulse duration of 100 fs [5, 6]. The SXFEL, considered as an upgrade for both facilities, will deliver completely transversely and temporally coherent pulses, that can reach 0.1 ps pulse duration and about 10 meV bandwidth [15]. Recently, a self-seeding scheme successfully produced a near Fourier-transform-limited x-ray pulses with 0.4 – 0.5 eV bandwidth at 8 – 9 keV photon energy [16]. Another option is the XFEL that will provide coherence time on the order of the pulse duration  $\sim 1$  ps, and meV narrow bandwidth [17]. We consider here the laser photon energy for the pump laser fixed at 25 keV for the XFEL and 12.4 keV for the SXFEL. The relativistic factor  $\gamma$  is given by the one-photon resonance condition:

$$E_3 - E_1 = \gamma(1 + \beta)\hbar\omega_p, \quad (4)$$

where  $E_j$  is the energy of state  $|j\rangle$  for  $j \in \{1, 2, 3\}$ ,  $\beta$  is the nuclear velocity in the unit of speed of light,  $\hbar$  is the reduced Planck constant and  $\omega_p$  is the pump laser angular frequency. The angular frequency of the Stokes x-ray laser  $\omega_s$  can be then determined. For copropagating pump and Stokes beams, the photon energy of the Stokes laser is smaller than that of the pump laser since  $E_2 > E_1$ .

Our numerical results [11, 12] show that an XFEL (SXFEL) laser peak intensity of around  $10^{18} - 10^{23}$  W/cm<sup>2</sup> together with a nuclear beam with  $\gamma$  factor between 1 and 72 is sufficient to achieve 100% nuclear coherent population transfer for several species of nuclei.

## COHERENT CONTROL OF SINGLE X-RAY PHOTONS

The photon as flying qubit is anticipated to be the fastest information carrier and to provide the most efficient computing implementation. However, with the extension of Moores law to the future, quantum photonic circuits must meet the bottleneck of the diffraction limit, i.e., a few hundred nm for the optical region. Forwarding optics and quantum information to shorter wavelengths in the x-ray region has the potential of shrinking computing elements in future photonic devices such as the quantum photonic circuit. This is strongly related to the development and availability of coherent x-ray sources. The realization of a short

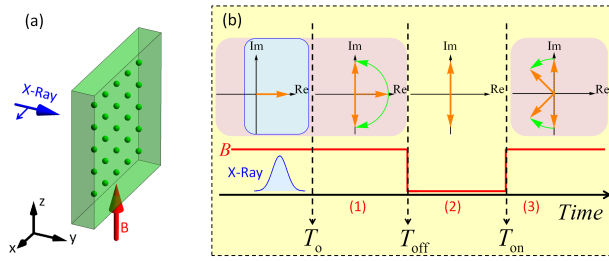


Figure 3: (a) NFS setup. The linear polarized x-ray pulse propagates in the  $y$  direction, and  $B$  is the external magnetic field initially parallel to the  $z$  axis. (b) Time dependence of the hyperfine magnetic field  $B$  and the corresponding dynamics of the nuclear currents (rotating arrows). The dynamics will be surveyed in three temporal domains: (1)  $T_0 < t < T_{\text{off}}$ ; (2)  $T_{\text{off}} < t < T_{\text{on}}$ ; (3)  $T_{\text{on}} < t$ .

wavelength quantum photonic circuit requires mastery of x-ray optics and powerful control tools of single-photon wave packet amplitude, frequency, polarization, and phase. Efficient coherent photon storage for photon delay lines and x-ray phase modulation, preferably even for single photon wave packets, are the next milestones to be reached.

### Nuclear Forward Scattering

Moving towards the interactions in the x-ray regime, new physical systems also come into play; e.g., nuclei with low-lying collective states naturally arise as candidates for x-ray quantum optics studies. Nuclear quantum optics may be rendered experimentally possible by the advent and commissioning of XFEL facilities. Coherent control tools based on nuclear cooperative effects are known also from nuclear forward scattering (NFS) [18, 19] experiments with third-generation synchrotron light sources. The underlying physics here relies on the delocalized nature of the nuclear excitation produced by coherent XFEL or synchrotron radiation (SR) light, i.e., the formation of so-called nuclear excitons. The typical NFS setup involves a solid-state target containing  $^{57}\text{Fe}$ . An x-ray pulse with meV bandwidth (either SR or coherent XFEL light) tuned on the 14.413 keV nuclear transition from the ground state to the first excited state shines perpendicular to the nuclear sample, as shown in Fig. 3 (a). SR typically produces at most one excited nucleus per pulse, thus providing a reliable single excitation and single released photon scenario. An externally applied magnetic field  $B$  parallel to the  $z$  axis induces the nuclear hyperfine splitting of the ground and excited  $^{57}\text{Fe}$  nuclear states of spins  $I_g = 1/2$  and  $I_e = 3/2$ , respectively. Depending on the x-ray polarization, different hyperfine transitions will be driven. In this study, we consider the x-ray field linearly polarized parallel to the  $x$  axis driving the two  $\Delta m = m_e - m_g = 0$  magnetic dipole transitions, where  $m_e$  and  $m_g$  denote the projections of the excited and ground state nuclear spins on the quantization axis, respectively.

### Coherent Storage and Phase Modulation of Single X-ray Photons

In the second part of this summary, a scheme of coherent control of x-ray single-photon wave packets is proposed [20]. We theoretically show that by switching off and on again the magnetic field in the considered nuclear sample, coherent storage of photons in the keV regime can be achieved for 10 - 100 ns. Fig. 3 (b) illustrates the time evolution of our photon storage scheme. The external magnetic field  $B$ , depicted by the red line, is present before the x-ray pulse impinges on the target at  $T_0$ . At  $T_{\text{off}}$  the  $B$  field is turned off and later turned back on at  $T_{\text{on}}$ . The rotating orange arrows depict the time evolution of the induced nuclear transition currents. Initially, the ensemble of  $^{57}\text{Fe}$  nuclei is excited by the x-ray pulse at  $T_0$ . Subsequently, the purely real currents are abruptly built. In the time interval (1), the two currents start to rotate in opposite directions on the complex plane with the factor of  $\exp(\pm i\Delta_B t)$  caused by the magnetic field until  $T_{\text{off}}$  when  $B$  is turned off. The corresponding phase gain is  $\pm i\Delta_B \tau$ , where  $\Delta_B$  is the hyperfine energy shift of interacting nuclear transitions and  $\tau = T_{\text{off}} - T_0$ . Within the time interval (2), the nuclear quantum state is frozen with the factor of  $\exp(\pm i\Delta_B \tau)$  since the hyperfine field has vanished. Therefore choosing  $\Delta_B \tau = N\pi/2$  with odd  $N$ , the emission of single photons is suppressed due to the destructive interference of two induced nuclear currents, i.e., the storage of single photons. During the time interval (3), the presence of the magnetic field allows the single photon signals emerge again. Corresponding phase modulation of the stored single-photon wave packet can be accomplished if the retrieving magnetic field is rotated by 180 degree.

## CONCLUSION

Nuclear quantum optics - there is a vast land remaining unexplored, which may lead to breakthrough for controlling atomic nuclei with coherent light sources [21]. Also, promising applications such as nuclear energy storage, a gamma-ray laser based on nuclear transitions or a nuclear-based frequency standard may become reality.

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