Cooperative effects in nuclear excitation with coherent x-ray light

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*New Journal of Physics* 14 (2012) 085025 (21pp)
Received 16 February 2012
Published 29 August 2012
Online at [http://www.njp.org/](http://www.njp.org/)
doi:10.1088/1367-2630/14/8/085025

**Abstract.** The interaction between super-intense coherent x-ray light and nuclei is studied theoretically. One of the main difficulties in driving nuclear transitions arises from the very narrow nuclear excited state widths, which limit the coupling between lasers and nuclei. In the context of direct laser–nucleus interaction, we consider the nuclear width broadening that occurs when in solid targets, when the excitation caused by a single photon is shared by a large number of nuclei, forming a collective excited state. Our results show that cooperative effects mostly contribute with a modest increase to the nuclear excited state population except in the case of \(^{57}\)Fe, where the enhancement can reach almost two orders of magnitude. Additionally, an update is given of previous estimates of the nuclear excited state population and signal photons for x-ray lasers interacting with solid-state and ion beam nuclear targets taking into account the experimental advances of x-ray coherent light sources. The presented values are an improvement by orders of magnitude and are encouraging as to the future prospects of nuclear quantum optics.

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1. Introduction

For a long time the direct interaction of photons with nuclei was generally considered too small to be relevant, primarily based on estimates about the magnitude of interaction matrix elements as in [1]. Nevertheless, the first interesting effects were observed in Mössbauer spectroscopy experiments, where despite low excitation rates, single gamma-photon electromagnetically induced transparency (EIT) was observed [2] and coherent control schemes were suggested [3]. Nuclear excitation experiments conducted at synchrotron radiation (SR) facilities evolving from the direction of Mössbauer spectroscopy have shown that interesting coherent phenomena such as quantum beats and photon echoes [4] may occur even without exciting a large part of the nuclear sample [5, 6]. Furthermore, it was observed that resonant scattering of SR on a nuclear ensemble, such as identical nuclei in a crystal lattice that allows for recoil-less nuclear transitions, may occur via an excited state that is excitonic in nature [6–8]. The decay of this collective nuclear excited state then occurs coherently in the forward direction, giving rise to nuclear forward scattering (NFS), and in the case of nuclei in a crystal also at Bragg angles [7–9]. The correlation of nuclear excitation amplitudes in the excitonic state furthermore leads to a speedup of the nuclear decay, also called coherent decay, which becomes considerably faster than the spontaneous de-excitation (characterized by the natural lifetime of a single nucleus). The most recent NFS experiments with SR used this feature to measure the collective Lamb shift in nuclei and to demonstrate EIT with resonant nuclei in a cavity [10, 11]. The aforementioned cooperative effects, although concerning nuclear excitation, have nevertheless been historically more related to solid-state physics, and their impact on the interaction between lasers and nuclei has so far been disregarded.

With the advent and commissioning of new light sources of higher power, brightness and temporal and transverse coherence, the driving of nuclear transitions with photons was set on the new course of nuclear quantum optics. The direct interaction between nuclei and coherent radiation from x-ray free electron lasers (XFEL) was proposed for the study of phenomena well known from atomic systems such as coherent trapping, electromagnetic-induced transparency or optical measurements of nuclear properties such as transition frequency and dipole moment [12]. This first pioneering work was followed by further studies of the resonant laser–nucleus interaction involving nuclear electric dipole-forbidden transitions [13, 14], which had been until then traditionally disregarded in presumed analogy to atomic quantum
optics. Nuclear coherent population transfer in a stimulated Raman adiabatic passage reminiscent of atomic quantum optics has also been investigated [15], as well as the direct laser-driven quantum nuclear control in a theoretical framework [16]. In all these works, the narrow bandwidth of nuclear transitions is a limiting issue. With values being of the order of $10^{-5}$–$10^{-10}$ eV for a single nucleus, their size lies tremendously below current XFEL energy bandwidths, suppressing the nuclear interaction with the radiation. Basically, the amount of nuclear excitation is dependent on the number of resonant laser photons, and unlike the typical situation in atomic quantum optics, in the case of nuclei the laser bandwidth is orders of magnitude larger than the resonance. Control of the nuclear bandwidth would very much help the matter.

In this work, we consider the impact of cooperative excitation on the efficiency of nuclear quantum optics applications. In solid-state targets consisting of Mössbauer nuclei, the excitation caused by a single photon may be shared by a large number of scattering centers, forming a collective excited state, sometimes with a significantly higher decay width than that of a single nucleus. This phenomenon may be exploited to enhance the widths of nuclear transitions and, to the best of our knowledge, has never been included so far in calculations of the laser–nucleus interaction. We show that for certain cases this enhancement of the nuclear transition width can increase by a factor proportional to the number of nuclei in the sample, and thus with the sample thickness within the laser Rayleigh length. We also study the physical limitations of the previous statement. Furthermore, a comparison between the cases with and without cooperative effects is made for the excited state population of the $^{57}_{26}$Fe isotope at different sizes of the photon beam focal spot and target thickness.

The use of solid-state targets that allow for cooperative effects for laser–nucleus interaction is restricted to nuclear transitions with energies below the laser photon energy available. At present, this value lies in the range of tens of keV. The first operational XFEL worldwide, the Linac Coherent Light Source (LCLS) at SLAC [17–19], has provided, since 2009, laser beams with a photon energy of approximately 10 keV (a tunability up to 1.2 Å wavelength was reported [18]). Beam diagnosis on the second and third harmonics of the primary beam has been performed [20]. The second operational XFEL worldwide, the SPring–8 Angstrom Compact free electron Laser (SACLA) in Japan, recently achieved the shortest wavelength of 0.634 Å x-rays (a photon energy of approximately 19.5 keV) [21, 22]. The European XFEL, at present still in construction at DESY in Hamburg, Germany, will achieve a 24.8 keV photon energy, corresponding to a wavelength of 0.5 Å [23, 24].\(^2\) Apart from the photon energy, for nuclear quantum optics applications, a crucial feature of the laser light pulses is the temporal coherence, i.e. the lack of phase jumps throughout the pulse duration. Here XFELs have the potential to bring a significant improvement compared to SR, besides the fact that they are also considerably brighter. To this day, even XFELs are not able to provide fully coherent laser pulses, but there are already several proposals on how to solve this problem in the near future [25–28]. Ideas include providing a single-pass XFEL with coherent seeding radiation (Seeded XFEL [25–27]) or designing cavities with the help of diamond mirrors [29, 30] to allow for multiple passes of the light through the electron beam (XFEL Oscillator [28]).

With these new developments for temporally coherent XFEL pulses and given the tremendous progress in x-ray beam focusing (recently reduced to a focal diameter of 7 nm [31]),

\(^2\) At present, it is speculated that the maximum photon wavelength of 1 Å specified in the technical design report is an understatement and even 0.5 Å will be achieved at the European XFEL, corresponding to a maximum photon energy of $E_{\text{max}} \approx 24.8$ keV.
our previous estimates for the magnitude of excited state population and signal rates based on experimental parameters dating back to 2007 and 2008 are in need of revision. Here we also give an update of the values presented in [13, 14] for both excitation of nuclei in solid-state targets and excitation of nuclei in ion beams. The latter will be useful when nuclear transitions with excitation energies higher than the available XFEL photon energies are required. In this case, moderate target acceleration has been proposed to bridge the gap between x-ray photon frequencies and nuclear transition energies [12]. We find that the excited state population values are enhanced in comparison with the older estimates by several orders of magnitude in many cases, especially for solid-state targets. For a focus of 7 nm, for instance, complete nuclear population inversion could be reached. Further experimental developments in this field may clear the way for unprecedented possibilities involving the direct interaction of radiation fields with nuclei. In particular, nuclear Rabi oscillations raise hope of the coherent control of nuclear excited state population, thus having the potential to open the entire field of nuclear quantum optics.

This paper is organized as follows. In section 2 we give a brief introduction to the theoretical framework for laser–nucleus interaction and cooperative effects in nuclei. The optical Bloch equations and the form of the laser–nucleus interaction Hamiltonian for higher multipole transitions are derived. Furthermore, we discuss the effective laser parameters entering the calculation. Section 2.3 introduces the collective nuclear excitation for Mössbauer nuclei in a lattice and investigates the origin of line width broadening of excited state transitions due to collective effects. In section 3, we present our numerical results for a number of nuclei with transition energies between 1 and 100 keV. We conclude the paper with a short summary and outlook.

2. Theory

In this section, the density matrix formalism is applied to a two-level nuclear system which is resonantly driven by a super-intense electromagnetic field following the outline presented in [13]. We use a semi-classical description, where the nucleus is considered as a hyperfine-split quantum two-level system and the electromagnetic field is treated classically. The effective laser parameters entering the calculation are introduced. In section 2.3, we introduce the cooperative effects that appear in solid-state targets. The nuclear width enhancement is studied by means of an iterative wave equation for the electric field of the scattered radiation.

2.1. Density matrix formalism for nuclei

We consider a nuclear two-level system consisting of the ground state $|g\rangle$ and the excited state $|e\rangle$. In the presence of an intrinsic or external magnetic field, the two levels will be split into several ground state magnetic sublevels $|I_g M_g \rangle$ and excited state ones $|I_e M_e \rangle$, where $I$ denotes the nuclear spin quantum number and $M$ its projection on the quantization axis. All nuclei are assumed to initially populate the ground state sublevels. Starting from time $t = 0$ they are irradiated with the intensity $I(t)$. The dynamics of the density matrix $\hat{\rho}$ is determined by [32]

$$i\hbar \frac{\partial}{\partial t} \hat{\rho} = [\hat{H}_0 + \hat{H}_I, \hat{\rho}] + \mathcal{L} \hat{\rho},$$

where $\hat{H}_0$ denotes the unperturbed nuclear Hamilton operator, $\hat{H}_I$ is the interaction Hamilton operator characterizing the effect of the irradiating laser electric field and $\mathcal{L}$ represents the

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Lindblad operator describing the various spontaneous relaxation channels. The matrix elements describing the system are denoted by \( \rho_{ij}(M_i, M_j) \) with \( i, j \in \{e, g\} \) where the respective magnetic sublevels for the ground and excited states are indicated by the magnetic spin quantum numbers in parentheses. We obtain the optical Bloch equations

\[
\frac{\partial}{\partial t} \rho_{gg}(M_g) = -\frac{2}{\hbar} \text{Im} \left( \sum_{M_e} \langle I_e, M_e | \hat{H}_1 | I_g, M_g \rangle e^{i\omega_k t} \rho_{ge}(M_g, M_e) \right) + \sum_{M_e} \gamma(M_g, M_e) \rho_{ee}(M_e),
\]

\[
\frac{\partial}{\partial t} \rho_{ee}(M_e) = \frac{2}{\hbar} \text{Im} \left( \sum_{M_g} \langle I_e, M_e | \hat{H}_1 | I_g, M_g \rangle e^{i\omega_k t} \rho_{ge}(M_g, M_e) \right) - \rho_{ee}(M_e) \sum_{M_g} \gamma(M_g, M_e),
\]

\[
\frac{\partial}{\partial t} \rho_{ge}(M_g, M_e) = i\Delta \rho_{ge} + \frac{i}{\hbar} \langle g | \hat{H}_1 | e \rangle e^{-i\omega_k t} \left( \rho_{ge} - \rho_{ee} \right) - \frac{\gamma(M_g, M_e)}{2} \rho_{ge}(M_g, M_e) - \gamma_{dec} \rho_{ge}(M_g, M_e),
\]

where \( \omega_k \) is the laser frequency, \( \Delta = \omega_0 - \omega_k \) is the detuning of the laser frequency with respect to the transition frequency and \( \gamma(M_g, M_e) \) is the (partial) decay rate corresponding to transitions between definite excited and ground state magnetic sublevels. The partial decay rates are related to the clebsch–gordan coefficients \( \langle j_1m_1, j_2m_2 | jm \rangle \) [33]

\[
\gamma(M_g, M_e) = \frac{2L_e + 1}{2L + 1} \left( \langle I_e M_g, I_e - M_e | LM \rangle \right)^2 \Gamma_0,
\]

where \( L \) and \( M \) denote the photon multipolarity and its total angular momentum projection, respectively, and \( \Gamma_0 \) is the total decay rate of the excited state. In equation (2) another decay term \( \gamma_{dec} \) that fastens the decay of the off-diagonal elements has been introduced. This stands for the decoherence rate that takes into account a possibly limited coherence time of the laser used in the experiment. The interaction Hamiltonian between the nucleus and the electromagnetic field can be written as

\[
\hat{H}_1 = -\frac{1}{c} \int \vec{j}(\vec{r}) \vec{A}(\vec{r}, t) \, d^3r,
\]

where \( c \) is the speed of light, \( \vec{j}(\vec{r}) \) denotes the nuclear charge current and \( \vec{A}(\vec{r}, t) \) represents the vector potential of the electromagnetic field. Usually, nuclear transitions have a specific multipolarity or present weak multipole mixing. When decomposing the vector potential into spherical waves and additionally assuming only one multipolarity, we obtain for the interaction Hamiltonian matrix element [13]

\[
\langle I_e M_e | \hat{H}_1 | I_g M_g \rangle \sim \mathcal{E}_k e^{-i\omega_k t} \sqrt{2\pi} \sqrt{\frac{L + 1}{L}} \frac{k^{L-1}}{(2L + 1)!!} \langle I_e M_e, I_g - M_g | L - \sigma \rangle \times \sqrt{2I_g + 1} \sqrt{B(\lambda L, I_g \rightarrow I_e)},
\]

where \( \mathcal{E}_k \) is the electric field amplitude, \( k \) denotes the photon wave number, \( \sigma \) represents the photon polarization and \( B(\lambda L, I_g \rightarrow I_e) \) is the reduced nuclear transition probability. Equation (5) is valid for both magnetic and electric multipole transitions. The calculation of the reduced transition probability requires knowledge of the nuclear wave function within a nuclear model. In order to keep a large degree of generality and to be independent of any particular theoretical model, it is more appropriate to take the experimental value of the reduced
transition probability, listed for example in online databases such as [34]. The reduced transition probability is also connected to the radiative decay rate by the expression [33]

\[ \Gamma_{\text{rad}} = \frac{2L+2}{\epsilon_0 L ((2L+1)))^2} \left( \frac{E_{\gamma}}{\hbar c} \right)^{2L+1} B(\lambda L, I_e \rightarrow I_g) , \]  

(6)

where \( \epsilon_0 \) is the vacuum permittivity and \( E_{\gamma} \) is the transition energy.

2.2. Effective laser parameters

According to equation (5), the electric field amplitude of the laser light needs to be specified in order to obtain the interaction Hamiltonian matrix element. This quantity has to be related to the laser parameters typically given in technical design reports of XFELs. The interaction Hamiltonian matrix element depends on the effective electric field amplitude, i.e. only photons resonant with the nuclear transition contribute to the laser–nucleus interaction. Nuclear transition widths are typically very small (10^{-5}–10^{-10} eV), whereas current XFELs achieve bandwidths of the order of 1 eV. Accordingly, only a small fraction of all photons within the XFEL pulse meet the resonance condition. To calculate the effective electric field \( \mathcal{E}_k \), we need to know the total peak intensity \( I_p \) of the photon beam and the number of photons from the laser which are resonant with the nuclear transition.

The total photon flux \( \Phi_{\text{tot}} \) can be obtained from the peak power \( P_p \) and the photon energy \( E_{\text{ph}} \), \( \Phi_{\text{tot}} = P_p / E_{\text{ph}} \). The total number of photons per pulse is related to the pulse duration \( T_p \) by \( N_{\text{tot}} = \Phi_{\text{tot}} T_p \). With the width of the nuclear transition \( \Gamma_0 \), the bandwidth of the laser \( BW \) and transition energy \( E_{\gamma} \), the resonant photon flux reads as

\[ \Phi_{\text{res}} = \Phi_{\text{tot}} \frac{\Gamma_0}{BW E_{\gamma}} . \]  

(7)

The number of resonant photons per pulse is then the product of the resonant photon flux \( \Phi_{\text{res}} \) and the pulse duration \( T_p \), \( N_{\text{res}} = \Phi_{\text{res}} T_p \). The total peak intensity \( I_p \) can be determined from the focal spot \( A_{\text{loc}} \) and the peak power \( P_p \),

\[ A_{\text{loc}} = \pi \left( \frac{d_{\text{loc}}}{2} \right)^2 , \quad I_p = \frac{P_p}{A_{\text{loc}}}, \]  

(8)

where \( d_{\text{loc}} \) is the focal diameter. Subsequently, the effective electric field can be calculated from the effective intensity via

\[ I_{\text{ef}} = I_p \frac{N_{\text{res}}}{N_{\text{tot}}} , \quad \mathcal{E}_{\text{ef}} = \sqrt{\frac{2 I_{\text{ef}}}{\epsilon_0 c}} . \]  

(9)

As is apparent in equation (7), the nuclear transition width \( \Gamma_0 \) is a crucial quantity that limits the number of resonant photons within the laser bandwidth and thus the strength of the field intensity and electric field amplitude. Typically, for neutral atoms, the nuclear width can be written as the sum of the radiative and internal conversion (IC) decay rates. In the case of a solid-state target, the collective nature of the nuclear excitation can lead to a broadening of the nuclear bandwidth, as discussed in the following.
2.3. Collective effects

New generation lasers such as the XFEL have brought with them a tremendous improvement with respect to brightness, coherence and spectral bandwidth in the keV regime. However, nuclear line widths are still orders of magnitude smaller than those we can achieve with even the best light sources today. Therefore, an important goal concerning laser–nucleus interaction is not only an improvement of laser bandwidths, but one is also seeking potential mechanisms that could increase the natural line widths of nuclei.

The possibility to use collective effects for line width enhancement in the interaction of light with a sample of identical nuclei relies on the recoilless absorption or emission of x-ray photons, i.e. the Mössbauer effect. For nuclei in a solid-state target, the photon momentum can be transferred to the crystal lattice as a whole rather than to a single nucleus. Typically, recoilless transitions involve an excited level with lifetimes in the range of 10 ps and energies between 5 and 180 keV. Longer (shorter) lifetimes than indicated lead, according to the Heisenberg uncertainty principle, to too narrow (broad) emission and absorption lines, which no longer effectively overlap. Even for samples of Mössbauer nuclei, the probability of recoilless absorption and emission is mostly less than 1, and can be approximated in the Debye model [35]

\[ f_{LM} = \exp \left[ -\frac{2E_R}{k_B\theta_D} \left( 1 + 4 \frac{T^2}{\theta_D^2} \int_0^{\frac{\theta_D}{T}} \frac{x \, dx}{e^x - 1} \right) \right], \quad (10) \]

by what is called the Lamb–Mössbauer factor \( f_{LM} \). In the above expression, \( k_B \) is the Boltzmann constant, \( \theta_D \) is the Debye temperature, \( T \) represents the solid-state target temperature and \( E_R = \frac{\hbar^2 e^2}{2M} \) denotes the recoil energy. We assume throughout this work that the solid-state target is at room temperature \( T = 300 \text{ K} \), as this does not have a significant effect on our results.

A single nucleus that is excited by an XFEL pulse can decay back to the ground state either via radiative decay or IC with a finite constant rate \( \Gamma \). If nuclei are, however, bound inside a crystal lattice, the excitation generated by the absorption of a photon will not be localized at a single nucleus, but rather spread out across a large number of nuclei all sharing and contributing to this so-called excitonic state [7]. Obviously, this is only possible via coherent photon scattering, meaning that a nucleus absorbing a photon decays back to its initial state upon re-emission. By any incoherent process such as IC, nuclear recoil or spin-flip, it would be possible to reveal the source’s location and therefore tell which nucleus was excited. The decay of the excitonic state occurs not only via the known radiative and IC channels, but also via a time-dependent coherent channel. The coherent decay channel at the time of creation of the exciton contributes to the width of the nuclear state and thus in turn to the number of photons in the laser pulse resonant with the nuclear transition.

There are generally two approaches to deduce the time-dependent coherent decay rate. One approach calculates the response function \( G(t) \) as done in [8] to obtain the time-dependent intensity. The other case [36, 37] is briefly presented in the following. Starting from Maxwell’s equations, one obtains [36]

\[ \left( \nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \vec{E} = \frac{4\pi}{c} \frac{\partial}{\partial t} \vec{J}, \quad (11) \]

where \( \vec{E} \) is the electric field vector of the laser light and \( \vec{J} \) denotes the nuclear source current. In the slowly varying envelope approximation [32] and assuming the light propagating in the
$\frac{\partial}{\partial z} \vec{E} = -\frac{2\pi}{c} \vec{J}.

(12)

The electric field amplitude of the radiation re-emitted coherently in a forward direction in second-order perturbation theory can be determined via the wave equation

$$\frac{\partial \vec{E}(z, t)}{\partial z} = -\sum_{l} K_l \vec{J}_l(t) \int_{-\infty}^{t} \vec{J}_l^\dagger(\tau) \cdot \vec{E}(z, \tau) \, d\tau,$n

(13)

where $\vec{J}_l(t)$ denotes the nuclear transition current matrix element for the transition specified by the index $l$ that runs over all possible transitions between hyperfine states. The coefficients $K_l$ characterize the transition and were defined in [36]. Assuming an initial laser pulse $\vec{E}(t) = \vec{E}_0 \delta(t)$ which is short on the time scale of nuclear dynamics, equation (13) can be solved iteratively, such that the electric field can be written as a sum

$$\vec{E}(z, t) = \sum_{n=0}^{\infty} \vec{E}_n(z, t),

(14)

where each summation term represents a multiple scattering order. The intensity behind the sample is given by $I(t) = |\vec{E}(d, t)|^2$, with $d$ being the target thickness. Assuming only one transition being driven by the laser pulse and disregarding hyperfine splitting, the intensity can be obtained analytically [7, 36],

$$I(t) = \frac{E_0^2 \xi^2}{\tau} e^{-\tau} \left[J_1(\sqrt{4\xi \tau})\right]^2,$n

(15)

where $\tau = \Gamma_0 t$ is the dimensionless time coordinate and $\Gamma_0$ denotes the transition’s total decay rate (radiative + IC). The notation $\xi = \sigma_R N d / 4$ is used for the so-called dimensionless thickness parameter, where

$$\sigma_R = 2\pi \frac{2I_e + 1}{2I_g + 1} \left(\frac{\hbar c}{E_0}\right)^2 \frac{1}{1 + \alpha} f_{LM},

(16)

is the radiative nuclear resonance cross section and $N$ is the number density of Mössbauer nuclei in the sample. Furthermore, $\alpha$ denotes the IC coefficient. In the limit of small times $\tau \lesssim 1/\xi$, the Bessel function $J_1$ can be expanded in a Taylor series and we approximately obtain

$$I(t) = \frac{E_0^2 \xi^2}{\tau} e^{-\xi t},

(17)

immediately after excitation. This clearly shows that the coherent decay is faster compared to the spontaneous incoherent $e^{-t}$ decay. Figure 1 illustrates the relation between different types of decay. The solid red line in figure 1 shows the coherently scattered radiation intensity versus time for the $^{57}_{26}$Fe isotope. The dotted black line in figure 1 indicates the normal $e^{-t}$ decay, whereas the dashed green line corresponds to the enhanced $e^{-\xi t}$ decay. For transitions between nuclear levels with hyperfine splitting, the intensity of the scattered light is modulated by the quantum beat due to interference between several unresolved hyperfine transitions. For our numerical analysis, however, the hyperfine splitting can be neglected due to the very short laser pulse length compared to the time scale of the quantum beats.

From equation (17), we can also determine the enhancement factor of the decay rate,

$$\Gamma \simeq (\xi + 1) \Gamma_0,

(18)
where $\Gamma_0$ is the spontaneous, isolated nucleus decay rate including the radiative and IC channel and $\Gamma$ additionally includes the enhancement due to collective effects. Since the effective thickness can take very large values (an effective thickness of $\xi = 100$ corresponds to the actual sample thickness of only approximately $d = 20 \, \mu m$ in the case of $^{57}\text{Fe}$), the enhancement factor can be substantial. However, obviously the effect cannot persist for arbitrary large values of the effective thickness $\xi$. The actual length $l_c$ in the sample thickness over which the coherent excitation can occur is limited by scattering and absorption processes, i.e. the photoelectric effect. Due to these processes the photon beam loses intensity corresponding to a characteristic photo-absorption length $1/\mu$, unique for every material and usually of the order of several $\mu m$ up to tens of $\mu m$. The effective thickness $\xi$ is therefore limited by the finite length in space over which the x-ray photons penetrate unperturbed inside the solid-state target. Typically, it is assumed that the length over which the coherent excitation can occur is of the order of the characteristic photo-absorption length. For instance, the enhancement for the 14.4 keV resonance in $^{57}\text{Fe}$ in NFS geometry is limited by photo-absorption to $10^3$ [38]. The effect of absorption in the crystal and of laser focusing (so far we implicitly assumed that the laser beam focus extends over the whole crystal thickness) will be addressed in detail in the next section.

3. Numerical results

In this section we present our numerical results on the excited state population after a single radiation pulse and expected signal photon rate for several nuclear transitions. We draw a line at transition energies of $E_\gamma = 25$ keV and assume that for all nuclei below this value the XFEL can deliver photons of the necessary energy such that a solid-state target can be used. For these cases, we investigate the magnitude of the cooperative effects and the corresponding limitations. Our choice of 25 keV transition energy is related to the predicted maximum photon energy value for

Figure 1. Intensity of coherently scattered light in equation (15) (solid red line), incoherent natural decay $e^{-\tau}$ (upper dotted black line) and enhanced decay in equation (17) (lower dashed green line) for the $^{57}\text{Fe}$ isotope (without hyperfine splitting) as a function of time over the duration of one excited nuclear state lifetime. The nuclear sample is assumed to have the effective thickness $\xi = 10$. 

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Table 1. Laser beam parameters of LCLS [18, 19, 43], SACLA [22, 44], European XFEL [23] and XFELO [28, 45]: the maximum photon energy $E_{\text{max}}$, bandwidth BW, pulse duration $T_p$, coherence time $T_{\text{coh}}$ (except for SACLA), peak power $P_{\text{peak}}$, peak and average brilliance $B$ and pulse repetition rate. The numbers for the European XFEL and XFELO correspond to their expected performance, while the ones for LCLS and SACLA are experimentally confirmed values.

| Parameter   | LCLS | SACLA | European XFEL | XFELO   |
|-------------|------|-------|---------------|---------|
| $E_{\text{max}}$ (eV) | 10 332 | 19 556 | 24 800 [24] | 25 000 |
| BW          | $2\times10^{-3}$ | $<10^{-3}$ | $8\times10^{-4}$ | $1.6\times10^{-7}$ |
| $T_p$ (fs)  | 70–100 | 1000 | 10000 | 10000 |
| $T_{\text{coh}}$ (fs) | 2 | – | 0.2 | 1000 |
| $P_{\text{peak}}$ (W) | $1.5\times10^{10}$ | $10^{10}$ | $2\times10^{10}$ | $4.1\times10^{9}$ |
| $B_{\text{peak}}$ a | $2\times10^{33}$ | $10^{33b}$ | $5.4\times10^{33}$ | $10^{35}$ |
| $B_{\text{average}}$ a | $6\times10^{21}$ | $10^{20b}$ | $1.6\times10^{25}$ | $1.5\times10^{27}$ |
| Rep. rate (Hz) | $3\times10$ | 10 | $4\times10^4$ | $10^8$ |

a The unit of brilliance is photons per (s mm$^2$ mrad$^2$ 0.1% BW).

b Not yet experimentally reported; values from the technical design report [44].
To solve the optical Bloch equations, equations (2), we assume as initial conditions that the system is in the ground state and that its population is equally distributed among the corresponding magnetic sublevels. This is well justified because the hyperfine energy splitting is much lower than the thermal energy at room temperature. We can subsequently numerically calculate the population of each magnetic sublevel of the excited state after one pulse length $T_p$ from the Bloch equations (2) using the interaction matrix element (5). The nuclear parameters $L_e$, $I_g$, the transition energy and multipolarity and the reduced nuclear transition probability $B(\lambda L, I_g \rightarrow I_e)$ are taken from nuclear databases such as [34]. The effective field intensity $E_{ef}$ is estimated using the relevant laser parameters according to the procedure described in section 2.2. The total excited state population is then the sum over all excited state magnetic sublevels

$$\rho_{ee} = \sum_{M_e=-I_e}^{I_e} \rho_{ee}(M_e).$$

After the radiation pulse, the excited nuclei decay back to their ground state either via the emission of a photon or, when possible, an IC electron. The re-emitted photons can be measured in a fluorescence experiment and in first approximation we obtain the signal photon rate

$$S = \rho_{ee} N_{fv} f_1 \frac{1}{1 + \alpha},$$

where $N_{fv}$ represents the number of atoms located inside the focal volume of the beam, $f_1$ is the pulse repetition rate of the laser and the factor $1/(1 + \alpha)$ includes the IC decay channel. For bare ions, the IC channel is closed and $\alpha = 0$ in the expression above.

### 3.1. Solid-state targets

In this section, we study several isotopes with transition energies below 25 keV, namely the cases of $^{57}$Fe, $^{72}$Ge, $^{83}$Rb, $^{119}$Sn, $^{134}$Cs, $^{137}$La, $^{149}$Sm, $^{153}$Sm, $^{167}$Tm, $^{169}$Tm, $^{171}$Tm, $^{181}$Tm, $^{187}$Os, $^{193}$Pt, $^{201}$Hg and $^{205}$Pb. The XFEL pulses shine on nuclei in a solid-state target. We calculate the population in the excited level after a single laser pulse which is resonantly driving the nuclear transition and the expected signal photon rate.

The enhancement of the nuclear width due to collective effects is increasing the number of resonant laser photons and therefore the percentage of excited state population. The key quantity to be determined is the effective thickness up to which the collective enhancement may occur. Two important parameters have to be taken into account: (i) the laser focal length within which the excitonic state can form and (ii) the spatial limitation of the excitonic state due to absorption and scattering of electrons which is approximated as the characteristic photo-absorption length. With x-ray focusing of the order of 100 nm, the focal length is given by twice the Rayleigh length via $L_{foc} = \frac{\pi d}{4} \left( \frac{\lambda}{d} \right)^2 \approx 1.6 \times 10^{-4} \text{ m}$ for a wavelength of 1 Å [46]. In table 2 we present relevant data for nuclear samples on the Lamb–Møllsauer factor $f_{LM}$, the characteristic photo-absorption length $1/\mu$, the focal length $L_{foc}$ and the dimensionless thickness parameter $\xi$ for $d = 1/\mu$. We find that the Lamb–Møllsauer factor becomes smaller for increasing transition energies. The characteristic photo-absorption length varies between 0.2 and 65 $\mu$m and is always smaller than the corresponding focal length for the considered laser focus of 100 nm. The dimensionless thickness parameter varies between values close to zero and reaches a maximum of $\xi = 87$ for $^{57}$Fe. The advantages of the $^{57}$Fe isotope (which is by far
Table 2. For each isotope, the Lamb–Mössbauer factor \( f_{\text{LM}} \), the characteristic photo-absorption length \( 1/\mu \), the focal length \( L_{\text{foc}} \) for a focus of \( 1 \mu \text{m} \) and the dimensionless thickness parameter \( \xi \) for \( d = 1/\mu \) are presented. The isotopes are ordered by their transition energy.

| Nuclide | \( E_\gamma \) (keV) | \( f_{\text{LM}} \) | \( 1/\mu \) (\( \mu \text{m} \)) | \( L_{\text{foc}} \) (\( \mu \text{m} \)) | \( \xi \) |
|---------|----------------|----------------|----------------|----------------|---------|
| \(^{201}\text{Hg}\) | 1.565 | 0.98 | 0.341 | 19.8 | 0.003 59 |
| \(^{193}\text{Pt}\) | 1.642 | 1 | 0.239 | 20.8 | 0.323 |
| \(^{205}\text{Pb}\) | 2.329 | 0.95 | 1.12 | 29.5 | 2.98 \times 10^{-7} |
| \(^{151}\text{Sm}\) | 4.821 | 0.92 | 3.96 | 61.1 | 0.218 |
| \(^{171}\text{Tm}\) | 5.036 | 0.95 | 1.96 | 63.8 | 0.249 |
| \(^{83}\text{Rb}\) | 5.260 | 0.2 | 18.3 | 66.6 | 0.556 |
| \(^{181}\text{Ta}\) | 6.238 | 0.94 | 1.79 | 79 | 2.53 |
| \(^{169}\text{Tm}\) | 8.410 | 0.85 | 7.56 | 107 | 1.66 |
| \(^{187}\text{Os}\) | 9.756 | 0.95 | 4.33 | 124 | 1.34 |
| \(^{167}\text{Tm}\) | 10.400 | 0.78 | 2.9 | 132 | 0.155 |
| \(^{137}\text{La}\) | 10.560 | 0.5 | 7.23 | 134 | 0.384 |
| \(^{134}\text{Cs}\) | 11.244 | 0.000 15 | 29.3 | 142 | 0.000 239 |
| \(^{73}\text{Ge}\) | 13.284 | 0.75 | 12.5 | 168 | 0.0764 |
| \(^{57}\text{Fe}\) | 14.413 | 0.76 | 21.9 | 183 | 86.7 |
| \(^{149}\text{Sm}\) | 22.507 | 0.16 | 33.7 | 285 | 0.472 |
| \(^{119}\text{Sn}\) | 23.871 | 0.082 | 64.6 | 302 | 6.86 |

the most widely used Mössbauer nucleus so far) become clear with the observation that it has both a large Lamb–Mössbauer factor and its crystal lattice allows for considerable effective thickness parameters \( \xi \).

Considering the case of \(^{57}\text{Fe}\), we proceed to investigate first the magnitude of collective effects for the excited state population. The Mössbauer transition of iron has a large Lamb–Mössbauer factor \( (f_{\text{LM}} \simeq 0.8) \). We study the dependence of the excited state population \( \rho_{ee} \) on the focal diameter \( d_{\text{foc}} \) considering a constant number of XFEL photons in the beam focus. A larger focus is thus related to the counteracting effects of lower intensity and larger collective nuclear width enhancement. In turn, a smaller focus limits the possible contribution of collective effects on the nuclear transition width but simultaneously allows for higher laser intensity. We use here for exemplification the XFELO laser parameters.

Figure 2 shows the excited state population \( \rho_{ee} \) as a function of the focal diameter \( d_{\text{foc}} \) on a logarithmic scale for the \(^{57}\text{Fe}\) isotope. We cover focal diameters from 7 nm (the best focus achieved in [31]) up to 100 nm. The sample thickness \( d \) is limited on the upper side due to both photo-absorption and beam focusing, i.e. we always need to choose the smaller one of the two. For a better visualization, in figure 2 the \( d_{\text{foc}} \)-axis is separated into two regions by a dashed vertical line, corresponding to the focal diameter at which the focal length is equal to the characteristic photo-absorption length of iron \( (L_{\text{foc}} = 1/\mu) \). As a comparison, we also present calculated data for the nuclear excited state population when no collective effects would occur.

According to figure 2 we may distinguish two regimes:

- \( d = L_{\text{foc}} < 1/\mu \): the sample thickness \( d \) is limited on the upper side by the focal length.

According to its definition, the dimensionless thickness parameter \( \xi \) depends linearly on
Figure 2. Excited state population $\rho_{ee}$ of $^{57}$Fe versus focal diameter $d_{foc}$ with (black crosses) and without (red triangles) cooperative effects taken into account. The dashed vertical line indicates the focal diameter where $d = L_{foc} = 1/\mu$.

the sample thickness $d = L_{foc}$, and the focal length $L_{foc}$ depends on the square of the focal diameter: $\xi \propto d_{foc}^2$. The dataset including collective effects (black crosses) is hardly affected by an increase of $d_{foc}$, because the line width enhancement due to the coherent decay compensates for the decreasing photon flux. For the case without collective effects (red triangles), $\rho_{ee}$ decreases significantly on the same interval. We find that at the best focus both datasets differ by a factor of 4.5, while at the vertical dashed line ($d = L_{foc} = 1/\mu$) the discrepancy already reaches a value of 88. This reflects the fact that at very small focal diameters not many nuclei are located inside the focal volume and thus collective effects become less important. For all isotopes for small excited state populations (as in our case), the cooperative effect enhancement factor for $\rho_{ee}$ is approximately $\xi + 1$, see equation (18).

- $d = \frac{1}{\mu} < L_{foc}$: the sample thickness $d$ is limited on the upper side by the (constant) characteristic photo-absorption length, which is about 22 $\mu$m for the $^{57}$Fe isotope. In this case, we obtain a constant dimensionless thickness parameter of $\xi \approx 87$. Hence, collective effects do not increase in magnitude any further when proceeding to larger focal diameters, and the corresponding dataset (black crosses) decreases parallel to the dataset without collective effects to the right of the vertical dashed line. We can thus deduce the maximum collective effect enhancement factor on the excited state population $\rho_{ee}$, which is limited due to the characteristic photo-absorption length. In the case of $^{57}$Fe this factor is $\xi(d = 1/\mu) + 1 \approx 88$.

The most interesting differences between the cases with and without collective effects occur at very small focal diameters of the order of nm up to several tens of nm. In [31], a reduction of the x-ray beam focusing up to diameters of the order of 7 nm was reported. However, in this case such a tiny focus occurs along with a tremendous loss in radiation intensity, i.e. only a small part of the photons in the pulse are focused while the rest are lost. At present, a 100 nm focus spot size achieved with Kirkpack–Baez mirrors is expected to be possible for 95% of the photons in the pulse [23]. As an alternative, diffractive lenses for x-rays [47–49] should be able to focus $5 \times 10^{10}$ photons on an $80 \times 80$ nm$^2$ focal spot. For our calculations in the following we
Table 3. The excited state population $\rho_{ee}$ and the signal photon rate $S$ for a sample of nuclei with transition energies below 25 keV are presented for four sets of laser parameters. Seeding, i.e. perfect coherence of the pulse, is considered for the European XFEL, LCLS and SACLAC. XFELQ is designed from the start to provide fully coherent pulses. The horizontal lines in the LCLS/SACLAC column indicate the border between currently accessible photon energies and isotopes with higher-lying first excited states. The numbers in parentheses denote the power of ten to multiply with.

| Nuclide | $E_{\gamma}$ (keV) | $\lambda L$ | European XFEL | LCLS | SACLAC | XFELQ |
|---------|--------------------|------------|---------------|-------|--------|-------|
|         | $\rho_{ee}$ $S$ (s$^{-1}$) | $\rho_{ee}$ $S$ (s$^{-1}$) | $\rho_{ee}$ $S$ (s$^{-1}$) | $\rho_{ee}$ $S$ (s$^{-1}$) |
| $^{201}$Hg | 1.565 M1 1.26(−8) 1.16 | 2.54(−8) 1.75(−3) | 6.35(−12) 1.46(−7) | 4.28(−7) 9.86(2) |
| $^{193}$Pt | 1.642 M1 8.15(−5) 1.83(5) | 1.64(−4) 2.76(2) | 4.11(−8) 2.3(−2) | 2.77(−3) 1.55(8) |
| $^{205}$Pb | 2.329 E2 2.04(−18) 5.34(−14) | 4.11(−18) 8.08(−17) | 1.03(−21) 6.73(−21) | 6.93(−17) 4.54(−11) |
| $^{151}$Sm | 4.821 M1 5.53(−8) 2.34(3) | 1.11(−7) 3.54 | 2.83(−11) 3.0(−4) | 1.91(−6) 2.02(6) |
| $^{171}$Tm | 5.036 M1 5.68(−6) 9.76(4) | 1.15(−5) 1.48(2) | 2.9(−9) 1.24(−2) | 1.95(−4) 8.4(7) |
| $^{83}$Rb | 5.260 M1 1.16(−5) 6.76(6) | 2.34(−5) 1.02(4) | 1.4(−8) 2.04 | 9.47(−4) 1.38(10) |
| $^{181}$Ta | 6.238 E1 5.15(−11) 2.23(1) | 1.04(−10) 3.37(−2) | 2.72(−14) 2.94(−6) | 1.84(−9) 1.98(4) |
| $^{169}$Tm | 8.410 M1 1.11(−5) 3.93(6) | 2.24(−5) 5.94(3) | 6.2(−9) 5.48(−1) | 4.18(−4) 3.69(9) |
| $^{187}$Os | 9.756 M1 8.1(−6) 2.79(6) | 1.63(−5) 4.22(3) | 4.2(−9) 3.62(−1) | 2.83(−4) 2.44(9) |
| $^{167}$Tm | 10.400 M1 1.49(−5) 8.21(5) | 3.01(−5) 1.24(3) | 7.81(−9) 1.07(−1) | 5.27(−4) 7.24(8) |
| $^{137}$La | 10.560 M1 4.02(−9) 2.38(3) | 8.1(−9) 3.6 | 2.6(−12) 3.84(−4) | 1.75(−7) 2.59(6) |
| $^{134}$Cs | 11.244 M1 1.78(−8) 1.53(4) | 3.6(−8) 2.32(1) | 2.35(−11) 5.06(−3) | 1.59(−6) 3.41(7) |
| $^{73}$Ge | 13.284 E2 1.01(−13) 1.56(−2) | 2.03(−13) 2.35(−5) | 5.21(−17) 2.01(−9) | 3.51(−12) 1.35(1) |
| $^{57}$Fe | 14.413 M1 2.03(−6) 1.23(8) | 4.08(−6) 1.86(5) | 1.34(−9) 2.04(1) | 9.03(−5) 1.37(11) |
| $^{149}$Sm | 22.507 M1 1.17(−7) 1.23(6) | 2.35(−7) 1.85(3) | 1.61(10) 4.23(−1) | 1.09(−5) 2.85(9) |
| $^{119}$Sn | 23.871 M1 1.17(−6) 1.43(8) | 2.35(−6) 1.26(5) | 6.33(−9) 1.93(2) | 4.27(−4) 1.3(12) |

have assumed that the full photon number remains in the beam focus and considered the more modest value of 100 nm for the focal spot size.

Taking into account the laser parameters of several XFEL facilities and a 100 nm focus spot size, we have calculated the population in the excited level after a single laser pulse $\rho_{ee}$ and the expected signal photon rate $S$ for the considered isotopes with transition energies below 25 keV. The collective enhancement of the nuclear transition width has been taken into account. For the case of full temporal coherence, we have considered the coherence time to be equal to the pulse duration. The difference between the results with partial and full temporal coherence account for almost six orders of magnitude for LCLS and European XFEL parameters and confirm the crucial importance of the longitudinal coherence of the pulses. The difference of the values of the excited state population $\rho_{ee}$ and the signal rate $S$ is smaller for SACLAC because we have assumed in the first place a long coherence time for the no-seeding case, which is probably optimistic. The SACLAC results are overall smaller than those for the other parameter sets mostly due to a very short pulse (10 fs)}
Table 4. The excited state population $\rho_{ee}$ and the signal photon rate $S$ this time without seeding, using the actual performance parameters for LCLS and SACLA and the present design values for the European XFEL. The horizontal lines in the LCLS/SACLA column indicate the border between currently accessible photon energies and isotopes with higher-lying first excited states. The numbers in parentheses denote the power of ten to multiply with.

| Nuclide | $E_y$ (keV) | $\lambda L$ | European XFEL | LCLS | SACLA |
|---------|-------------|-----------|---------------|------|-------|
| $^{201}$Hg | 1.565 | M1 | 1.67(−14) | 1.54(−6) | 9.09(−14) | 6.28(−9) | 5.58(−15) | 1.29(−10) |
| $^{193}$Pt | 1.642 | M1 | 1.08(−10) | 2.42(−1) | 5.88(−10) | 9.89(−4) | 3.62(−11) | 2.03(−5) |
| $^{205}$Pb | 2.329 | E2 | 2.7(−24) | 7.08(−20) | 1.47(−23) | 2.9(−22) | 9.54(−25) | 6.26(−24) |
| $^{151}$Sm | 4.821 | M1 | 7.32(−14) | 3.1(−3) | 3.99(−13) | 1.27(−5) | 2.49(−14) | 2.64(−7) |
| $^{171}$Tm | 5.036 | M1 | 7.52(−12) | 1.29(−1) | 4.1(−11) | 5.29(−4) | 2.55(−12) | 1.1(−5) |
| $^{83}$Rb | 5.260 | M1 | 1.54(−11) | 8.95 | 8.4(−11) | 3.66(−2) | 1.24(−11) | 1.8(−3) |
| $^{181}$Ta | 6.238 | E1 | 6.82(−17) | 2.95(−5) | 3.72(−16) | 1.21(−7) | 2.42(−17) | 2.61(−9) |
| $^{169}$Tm | 8.410 | M1 | 1.47(−11) | 5.2 | 8.02(−11) | 2.13(−2) | 5.46(−12) | 4.82(−4) |
| $^{187}$Os | 9.756 | M1 | 1.07(−11) | 3.7 | 5.85(−11) | 1.51(−2) | 3.7(−12) | 3.18(−4) |
| $^{167}$Tm | 10.400 | M1 | 1.98(−11) | 1.09 | 1.08(−10) | 4.44(−3) | 6.88(−12) | 9.45(−5) |
| $^{137}$La | 10.560 | M1 | 5.32(−15) | 3.15(−3) | 2.9(−14) | 1.29(−5) | 2.29(−15) | 3.38(−7) |
| $^{134}$Cs | 11.244 | M1 | 2.36(−14) | 2.03(−2) | 1.29(−13) | 8.3(−5) | 2.07(−14) | 4.45(−6) |
| $^{72}$Ge | 13.284 | E2 | 1.34(−19) | 2.06(−8) | 7.29(−19) | 8.42(−11) | 4.63(−20) | 1.78(−12) |
| $^{57}$Fe | 14.413 | M1 | 2.68(−12) | 1.63(2) | 1.46(−11) | 6.67(−1) | 1.18(−12) | 1.79(−2) |
| $^{149}$Sm | 22.507 | M1 | 1.54(−13) | 1.62 | 8.42(−13) | 6.64(−3) | 1.42(−13) | 3.72(−4) |
| $^{119}$Sn | 23.871 | M1 | 1.55(−12) | 1.89(2) | 8.43(−12) | 7.73(−1) | 5.57(−12) | 1.7(−1) |

and the currently small repetition rate of 10 Hz. The high repetition rate of the future European XFEL, on the other hand, makes this facility particularly attractive for high signal rates. Seeding simulations have already been performed for this facility, but unfortunately mostly aiming at a shorter pulse duration (7 fs) [27]. Optimal for nuclear quantum optics applications, however, are longer pulses, as may be available someday at a future XFEL facility.

For better visualization, we plot the excited state population and the signal rate for the European XFEL parameters with seeding and a pulse duration of 100 fs in figure 3. We refrain from plotting the same figures for SACLA, LCLS and XFEL since all points only have a constant offset factor that can be looked up in tables 3 and 4. The highest excited state populations are obtained for the $^{193}$Pt isotope: $\rho_{ee} = 8.15 \times 10^{-5}$ (European XFEL), $\rho_{ee} = 1.64 \times 10^{-4}$ (LCLS), $\rho_{ee} = 2.77 \times 10^{-3}$ (XFEL) and $\rho_{ee} = 4.11 \times 10^{-8}$ (SACLA). The highest signal photon rates are obtained for the $^{119}$Sn isotope, namely $1.43 \times 10^5$ s$^{-1}$ (European XFEL), $S = 2.16 \times 10^5$ s$^{-1}$ (LCLS), $S = 1.30 \times 10^{12}$ s$^{-1}$ (XFEL) and $S = 1.93 \times 10^2$ s$^{-1}$ (SACLA). Since LCLS has a lower pulse repetition rate than the European XFEL, the signal photon rates are lower even though the excited state population obtained with the LCLS parameters is actually higher. Were the total number of photons per pulse to be successfully focused on the 7 nm focal spot [31], the achieved intensity would allow for an excited state population as high as $\rho_{ee} = 1$ for XFEL laser parameters. One may therefore speculate that
Figure 3. The excited state population $\rho_{ee}$ (left) and the signal photon rate $S$ (right) versus transition energy $E_\gamma$ obtained for the European XFEL parameters considering full temporal coherence for the isotopes in Table 3.

the experimental realization of nuclear Rabi oscillations is related to the future improvement of x-ray optics devices.

Finally, we conclude this part with two important observations regarding the approximations used in our calculations and possible experimental issues. Firstly, the formation of the exciton and its coherent decay perturb both the time signature of the spectrum as well as the ratio of emitted photons to emitted IC electrons. In this respect, the signal rate expression in equation (20) is a rather poor approximation. Following the iteration procedure described in section 2.3, the time-dependent collective width of the nuclear excited states should be accurately calculated [50] (also taking into account the coherence of the laser) and correspondingly the radiative nuclear decay expected in fluorescence experiments.

The second observation is meant to counter the effects of the first one and relates to the ablation or melting of the target. Due to the very strong intensities of the XFEL, far larger than previous available values in SR experiments, solid-state samples may be completely depleted of electrons and, as a consequence of Coulomb repulsion, explode on a time scale shorter than that of the nuclear decay. As a consequence, the decay of the excited nuclei proceeds from a state very much different from the original target. While studies on target damage and nuclear x-ray fluorescence have not been performed so far, we expect that (i) through Coulomb explosion the excitonic state is destroyed and no coherent decay is present and (ii) the nuclear decay proceeds with the spontaneous decay rate known from single nuclei. Presumably, the IC channel is restored by electron recombination into the highly charged ion on a time scale faster than that of the nuclear decay. The destruction of the sample would make multiple exposure of the same nuclei to the XFEL pulse impossible, but successive burns on a rotating or tape station system solid-state nuclear target offer a scenario for the laser–nuclei interaction experiment. The expression for the signal photon rate (20) is therefore suitable for describing this situation.

Nuclear fluorescence experiments similar to NFS ones can be performed with solid-state targets. The main requirement for the targets is to have a high Lamb–Mössbauer factor $f_{LM}$, i.e. a large fraction of recoilless nuclear transitions occurring in the sample. This can be achieved either in pure crystals or in polycrystalline foils such as the ones used in nuclear lighthouse effect experiments [51]. In principle, the nuclei of interest can also be implanted.
as dopants in a hard crystalline host with a conveniently high Debye temperature, in order to increase the value of \( f_{\text{LM}} \), as was suggested in [50]. However, estimating the recoilless fraction of absorption and emission in nuclear transitions for the impurities in hard crystalline host materials requires dedicated calculations. In contrast to NFS experiments with SR, the sample is likely to be destroyed by the pulse, such that the fluorescence photons from the coherent decay will no longer be emitted in the forward direction alone. Furthermore, one may expect a high background from the electronic plasma created by the x-ray pulse. Time gating techniques such as those used in NFS experiments [52] may prove themselves useful also here. A good signal-to-background ratio is in this case crucial to experimentally confirm for the first time the direct laser–nucleus interaction.

### 3.2. Accelerated nuclear targets

Here we provide an update of previous values [13, 14] for the case of x-ray pulses attempting to drive nuclear transitions with higher excitation energies than those available directly from the laser. To bridge the gap between photon and nuclear transition frequencies, target acceleration has been proposed [12]. The target nuclei can be accelerated to relativistic speeds towards the photon beam. Due to the relativistic Doppler shift the photon energy becomes higher in the nuclear rest frame than it is in the laboratory frame. Hence, one accelerates the nuclei to a specific velocity \( v \) relative to the laboratory system to achieve an overlap between photon energy and transition energy in the nuclear rest frame (subscript \( n \)),

\[
\omega_n = \omega \sqrt{\frac{1 + \beta}{1 - \beta}},
\]

where \( \beta = \frac{v}{c} \).

The laser frequency in the laboratory frame \( \omega \) and in the nuclear rest frame \( \omega_n \) are related by the relativistic Doppler shift. The electric field amplitude of the laser is transformed as

\[
E_n = E \left( 1 + \beta \right) \gamma,
\]

where \( \gamma = \frac{1}{\sqrt{1 - \beta^2}} \).

Furthermore, both the natural bandwidth of the laser \( \text{BW} \) and the relative uncertainty in the gamma factor \( \frac{\Delta \gamma}{\gamma} \) influence the bandwidth of the laser in the nuclear rest frame. We assume that both photon energy in the laboratory frame and the gamma factor follow a Gaussian distribution, such that the bandwidth transformed into the nuclear rest frame can be determined by Gaussian error propagation and is approximately given by

\[
\text{BW}_n \simeq \sqrt{(\text{BW})^2 + \left( \frac{\Delta \gamma}{\gamma} \right)^2}.
\]

The transformed laser bandwidth then enters the calculation of the flux of resonant photons in equation (7).

We analyze the cases of \(^{131}\text{Xe}, ^{153}\text{Sm}, ^{153}\text{Eu}, ^{160}\text{Tb}, ^{165}\text{Ho}, ^{173}\text{Yb}, ^{183}\text{W}, ^{192}\text{Ir}, ^{223}\text{Ra} \) and \(^{195}\text{Pt} \) having transition energies up to 100 keV. The maximum photon energy of LCLS is no longer assumed to be 25 keV as in section 3.1, but rather 10.3 keV as stated in table 1 corresponding to the photon energies available at present. Similarly, we have considered 19.5 keV photon energy for the SACLA XFEL. As in [13, 14], we consider here perfect temporal coherence of the laser pulses, which corresponds to XFEL seeding. The target consists now of bare nuclei without the surrounding electrons so that neither collective effects nor IC plays a role and only the radiative decay rate \( \Gamma_{\text{rad}} \) (6) is taken into account. In the few cases where the laser-driven nuclear transition does not connect the ground state to the first excited state but to a higher level, we neglect the weak non-resonant coupling between the x-ray pulse and the intermediate lower-lying nuclear states, which remain unpopulated.
The largest signal rates, namely for the XFELO, are also illustrated in figure 4. Excited state population after one radiation pulse and the signal photon rate for European XFEL, LCLS, SACLA and XFELO parameters. Acceleration of the target nuclei is considered. The numbers in parentheses denote the power of ten to multiply with. See text for further explanations.

Table 5. The excited state population $\rho_{ee}$ and the signal photon rate $S$ for nuclei with transition energies above 25 keV for European XFEL, LCLS, SACLA and XFELO parameters. Acceleration of the target nuclei is considered. The numbers in parentheses denote the power of ten to multiply with. See text for further explanations.

| Nuclide | $E_\gamma$ (keV) | $\lambda L$ | European XFEL $\rho_{ee}$ ($s^{-1}$) | LCLS $\rho_{ee}$ ($s^{-1}$) | SACLA $\rho_{ee}$ ($s^{-1}$) | XFELO $\rho_{ee}$ ($s^{-1}$) |
|---------|-----------------|-------------|--------------------------------------|----------------|----------------|----------------|
| $^{153}\text{Sm}$ | 35.843 | E1 | 2.13 (−7) | 2.1 (−3) | 2.47 (−6) | 7.6 (−6) | 1.11 (−8) | 1.14 (−8) | 4.30 (−6) | 1.07 |
| $^{183}\text{W}$ | 46.484 | M1 | 5.55 (−9) | 5.47 (−5) | 6.43 (−8) | 1.98 (−7) | 1.55 (−9) | 1.59 (−9) | 1.12 (−7) | 2.78 (−2) |
| $^{223}\text{Ra}$ | 50.128 | E1 | 5.29 (−9) | 5.22 (−5) | 6.13 (−8) | 1.88 (−7) | 2.62 (−10) | 2.68 (−10) | 1.07 (−7) | 2.65 (−2) |
| $^{160}\text{Tb}$ | 64.110 | E1 | 3.99 (−10) | 3.94 (−6) | 4.63 (−9) | 1.42 (−8) | 2.30 (−11) | 2.36 (−11) | 8.05 (−9) | 2.00 (−3) |
| $^{171}\text{Yb}$ | 78.647 | M1 | 2.88 (−8) | 2.84 (−4) | 3.34 (−7) | 1.03 (−6) | 6.69 (−9) | 6.85 (−9) | 5.81 (−7) | 1.45 (−1) |
| $^{131}\text{Xe}$ | 80.185 | M1 | 8.73 (−10) | 8.62 (−6) | 1.01 (−8) | 3.11 (−8) | 6.50 (−11) | 6.67 (−11) | 1.76 (−8) | 4.38 (−3) |
| $^{191}\text{Ir}$ | 84.275 | E1 | 2.30 (−10) | 2.27 (−6) | 2.67 (−9) | 8.2 (−9) | 1.08 (−11) | 1.11 (−11) | 4.64 (−9) | 1.15 (−3) |
| $^{165}\text{Ho}$ | 94.700 | M1 | 3.14 (−7) | 3.10 (−3) | 3.64 (−6) | 1.12 (−5) | 3.75 (−8) | 3.85 (−8) | 6.33 (−6) | 1.57 |
| $^{157}\text{Eu}$ | 97.429 | E1 | 3.10 (−8) | 3.06 (−4) | 3.59 (−7) | 1.10 (−6) | 1.17 (−9) | 1.20 (−9) | 6.24 (−7) | 1.55 (−1) |
| $^{195}\text{Pt}$ | 98.882 | M1 | 2.07 (−9) | 2.04 (−5) | 2.40 (−8) | 7.38 (−8) | 4.72 (−10) | 4.83 (−10) | 4.18 (−8) | 1.04 (−2) |

We proceed similarly to the case of a solid-state target in section 3.1, but leave out IC and collective effects when determining the transition width and include the relativistic boost of all laser parameters before proceeding to the calculation of the electric field. Table 5 shows the excited state population after one radiation pulse and the signal photon rate for European XFEL, LCLS, SACLA and XFELO laser parameters. The results for the set of parameters which yields the largest signal rates, namely for the XFELO, are also illustrated in figure 4.

The highest excited state populations involve the $^{165}_{67}\text{Ho}$ isotope. One reaches $\rho_{ee} = 3.14 \times 10^{-7}$ (European XFEL), $\rho_{ee} = 3.64 \times 10^{-6}$ (LCLS), $\rho_{ee} = 3.75 \times 10^{-8}$ (SACLA) and $\rho_{ee} = 6.33 \times 10^{-6}$ (XFELO). Our results are roughly three (European XFEL), four (XFELO), five (LCLS) and two (SACLA) orders of magnitude larger compared to previous estimates in [13]. The rather modest results obtained for the SACLA parameters can be again traced back to the present very short pulse (10 fs) and low repetition rate of 10 Hz.

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In general, nuclear transitions widths are larger for higher excitation energies. Furthermore, the boosting of the electric field amplitude enhances the radiation intensity in the nuclear rest frame. Therefore, we would expect that in comparison to solid-state targets the disappearance of the internal conversion decay channel and of the collective effects is compensated for. However, this is not the case. The low excited state populations in the case of target acceleration can be traced back to the relative uncertainty in the relativistic $\gamma$ factor, a property of the ion accelerator producing the beam that causes the bandwidth of the photons to be higher than the actual laser bandwidth in the nuclear rest frame, see equation (21). Ideally, the relative uncertainty $\Delta \gamma / \gamma$ would be smaller than the laser bandwidth. This unfortunately is not the case and the main advantages of the seeded XFEL and XFELO, the low bandwidth of about $10^{-7}$, close to the Fourier width of the laser pulses, cannot be exploited. We thus obtain overall lower excited state populations than in section 3.1. Moreover, ion beams have rather small densities of the order of $\rho = 10^{11}$ ions cm$^{-3}$ [53], which results in significantly lower signal photon rates than for solid-state targets.

On the one hand, ion beams are less problematic than solid-state targets because they contain no electrons; therefore no radiation is absorbed due to the photoelectric effect as in crystals, also causing no background in the photon detector. Furthermore, they are comparatively easy to treat theoretically because they consist of bare nuclei without an electron shell. On the other hand, a large accelerator is needed to produce an ion beam of high quality and unfortunately such a facility does not exist in conjunction with an XFEL today. Furthermore, particle densities in ion beams are very low and high densities as in a solid are useful in obtaining the large number of excited nuclei that we want to measure. In summary, accelerated nuclear targets in conjunction with XFEL radiation are a gedanken experiment at the moment; nevertheless, both types of facilities already exist, and might even be combined in the near future, for instance in the framework of MaRIE [39].

4. Conclusions

The interaction between nuclei and intense x-ray laser fields has been revisited to include nuclear solid-state effects arising from the delocalization of the excitation over a large number of identical nuclei. The formation of this nuclear exciton leads to a broadening of the nuclear width, which in turn increases the number of laser photons in resonance and the amount of excitation. The necessary conditions and limitations, mainly related to scattering and absorption in solid states and the laser Rayleigh length, were investigated. We find that by far the most promising candidate for laser–nucleus interaction is the well-known $^{57}$Fe Mössbauer isotope, for which cooperative effects can increase the coupling of the 14.4 keV nuclear transition to the XFEL by roughly two orders of magnitude. Additionally, we have also provided an update of previous values for laser–nucleus interaction in the accelerated ion beam target setup. These values are, however, less promising than the results for solid-state targets. After a long history of use in Mössbauer spectroscopy, with the operation of XFEL machines to produce photons of suitable energy, $^{57}$Fe might also be the first isotope to open the field of nuclear quantum optics. Experimentally, nuclear quantum optics has the potential to play an important role in future XFEL applications, as long-term objectives involve exciting goals such as the preparation of nuclei in excited states, nuclear branching ratio control and isomer triggering.
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