

Toward Nonlinear Optics with Mössbauer Nuclei Using X-Ray Cavities

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Strong excitation of nuclear resonances, particularly of Mössbauer nuclei, has been a longstanding goal and the advance of novel x-ray sources is promising new options in this regard. Here, we map out the necessary experimental conditions for the more general goal of entering the nonlinear optics regime with nuclei and compare with available technology. In particular, we present a comprehensive theory of nonlinear nuclear excitation in thin-film x-ray cavities by focused x-ray pulses. We thereby identify cavity geometries with broad resonances that allow one to boost the nuclear excitation even at moderately tight focusing and offer the possibility to mitigate radiation damage.

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In this Letter, we revisit an old challenge: is it possible to fully excite an ensemble of atomic nuclei using externally applied electromagnetic fields? This question relates to the goal of building a γ -ray laser (“graser”), with inverted nuclei as a gain medium, which was proposed long ago after the laser-maser principle [1]. The discovery of the Mössbauer effect, which allows for the recoilless absorption and emission of γ -ray photons from certain nuclei [2,3], further fueled this research. Despite interim hope to circumvent difficulties by utilizing lasing without inversion [4], most approaches to grasing were deemed unfeasible by the late 1990s [5]. Nevertheless, recent technological and conceptual developments prompt us to reconsider the challenge.

First, advances in x-ray source technology such as x-ray free-electron lasers (XFEL) [6,7] have promoted the study of high-energy nonlinear effects in atoms, analogous to the revolutionary development at lower photon energies following the invention of conventional laser sources [8,9]. Various nonlinear x-ray processes have already been observed involving electronic transitions [10–14]. Also lasing on an inner-shell electronic transition based on XFEL-generated inversion has been demonstrated [15]. This raises the question of whether the source advances provide new avenues for the excitation of nuclei.

Second, Mössbauer nuclei by now have evolved into a promising platform for quantum optics. In a series of experiments, key concepts of quantum optics [16–22] and coherent control [23–30] have been demonstrated with nuclei as the matter part. However, while these experiments exploit coherence and interference effects and demonstrate a surprising level of control, they still operate in the linear optics regime [31,32].

These two developments also motivate a shift in focus away from the goal of reaching full inversion and toward more general nonlinear optics effects, further fueled by recent experiments from the optical domain [33–37] demonstrating that effects such as photon correlations can already appear far below full excitation. First experiments with Mössbauer nuclei at XFELs [38,39] explore this direction, e.g., by scattering multiple photons per shot [38]. Still these experiments do not reach beyond linear optics, which remains an open challenge.

On the theoretical side, a number of schemes have been proposed to achieve higher excitation. For example, it has been suggested to overcome discouraging first estimates [40] using additional accelerations of the target nuclei [41], nondipole transitions [42], or collective effects in the light-matter interaction [43,44]. Furthermore, there are multiple approaches to boost the excitation of the nuclei by a suitable tailoring of their electromagnetic environment. As one option, front coupling in one-dimensional x-ray waveguides used for imaging applications [45,46] has recently been implemented with nuclei [47,48], and tapered variants have been suggested as a candidate for nuclear inversion [49,50]. Alternatively, x-ray thin-film planar cavities containing layers of Mössbauer nuclei are well established as a platform for nuclear quantum optics in the linear regime [51,52]. The field enhancement inside such

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structures has been considered [16,31,53–55] and optimized [46,56]. However, the latter works consider the state-of-the-art probing using highly collimated x-rays, which is unfavorable for strong excitation. Focusing to enhance the nuclear excitation has been proposed [57], but so far, rigorous modeling of the excitation dynamics in planar waveguides driven by focused x-ray beams and a corresponding road map toward nonlinear excitation are missing.

In this Letter, we utilize a comprehensive theory developed in a companion paper [58] to study the excitation dynamics of Mössbauer nuclei in planar cavities driven by focused x-ray pulses (see Fig. 1). We find that the rigorous modeling of the focusing leads to optimum cavity structures that qualitatively differ from well-established design paradigms for collimated x-ray beams. With varying size of the x-ray focus, we identify a transition between standing-wave interference and ballistic reflections, with significant field enhancement down to moderately tight focusing. We calculate the necessary source parameters to enter the nonlinear regime of significantly excited nuclei and demonstrate its feasibility by comparing with currently available conditions and planned upgrades.

We start with the nuclear excitation dynamics. Unlike in the low-excitation regime [55,59], the modeling of dissipative quantum dynamics of many interacting few-level systems at higher excitation remains a challenging problem of much current interest [60–62]. However, x-ray pulses delivered by accelerator-based sources are orders of magnitude shorter (typically ps to fs scale) than any other relevant timescale in the nuclear dynamics (e.g., ns for ^{57}Fe). This allows one to separate the excitation from the subsequent decay dynamics. The former then essentially

reduces to a single-particle problem (see [58,63] for details), since dissipative and coupling processes are ineffective over the short excitation timescales.

The single-nucleus dynamics can straightforwardly be solved numerically for arbitrary input fields. However, to gain analytical insight, we further assume resonant and Fourier-limited x-ray pulses, an idealization of pulses generated via hard x-ray self-seeding [64–66]. Then, the pulse area theorem [67–69] relates the nuclear state after the x-ray pulse to the Fourier transform of the x-ray field at the nuclear resonance frequency $\mathbf{E}_{\text{drive}}(\mathbf{r}_{\text{nuc}}, \omega_{\text{nuc}})$ as [58,63,70]

$$\Phi = \frac{4\pi}{\hbar} |\mathbf{d} \cdot \mathbf{E}_{\text{drive}}(\mathbf{r}_{\text{nuc}}, \omega_{\text{nuc}})|. \quad (1)$$

Here, Φ is the so-called pulse area, and $\mathbf{d}$, $\mathbf{r}_{\text{nuc}}$, and ω_{nuc} are the electric dipole moment (an effective quantity for magnetic transitions), the position, and the transition frequency of the nuclei, respectively. The nuclear excitation—which is directly related to the magnitude of many nonlinear processes—is then obtained as $\sigma^z = -\cos(\Phi)$, where σ^z is the expectation value of the transition’s Pauli operator.

An intuitive interpretation is gained by rewriting Eq. (1) [58] as

$$\Phi = \frac{\chi_\sigma}{w_0} \chi_{\text{source}} \xi_{\mathcal{E}}, \quad (2)$$

where χ_σ is a transition characteristic that solely depends on the nuclear properties such as the dipole moment. w_0 is the beam waist size in the focal plane, characterizing the strength of the focusing. The incident x-ray pulses are characterized by their pulse energy E_{pulse} via $\chi_{\text{source}} := \sqrt{E_{\text{pulse}}/b_r}$, where b_r is the relative bandwidth such that $\Delta\nu = b_r\nu$ is the absolute frequency bandwidth. Finally, $\xi_{\mathcal{E}}$ is an enhancement factor due to the cavity. Without cavity it is equal to unity. The goal of the cavity environment is to increase $\xi_{\mathcal{E}}$. From the criterion $\Phi = \pi$ for reaching inversion, we define a *necessary* source characteristic for a given nucleus, cavity, and focusing as

$$\chi_{\text{source}}^{\text{nec}} = \pi \frac{w_0}{\chi_\sigma \xi_{\mathcal{E}}}. \quad (3)$$

For a given χ_{source} , one can then express the achievable nuclear excitation as $\sigma^z = -\cos(\pi\chi_{\text{source}}/\chi_{\text{source}}^{\text{nec}})$.

The crucial position-dependent enhancement factor $\xi_{\mathcal{E}}(\mathbf{r})$ is obtained by calculating the propagation of focused x-ray pulses in the cavity medium using Maxwell’s equations. In the companion paper [58], an efficient semianalytical algorithm specialized for this purpose is developed, which forms the basis of the results below. This formalism solves the wave propagation in the cavity for a focused beam accounting for interferences between the reflections at each interface. Equation (3) is also derived in detail, including a discussion of physical limitations.

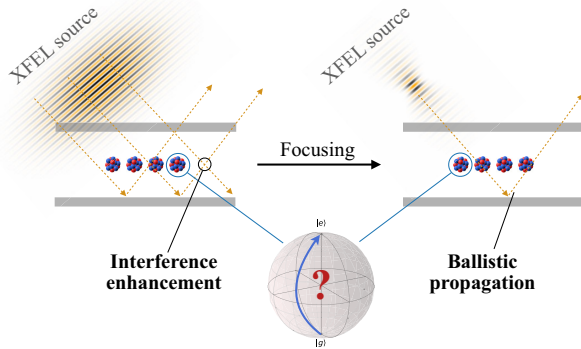


FIG. 1. Sketch of the setup, featuring a focused x-ray pulse (not to scale) incident on a thin-film cavity doped with narrow resonances, such as Mössbauer nuclei. By focusing the incident x-ray pulse, one can concentrate the radiation on fewer nuclei, potentially increasing their excitation toward inversion (illustrated by the Bloch sphere). However, this comes at the cost of reducing the cavity enhancement from interference. We show that using suitably designed cavities, this limitation can be overcome in intermediate focusing regimes.

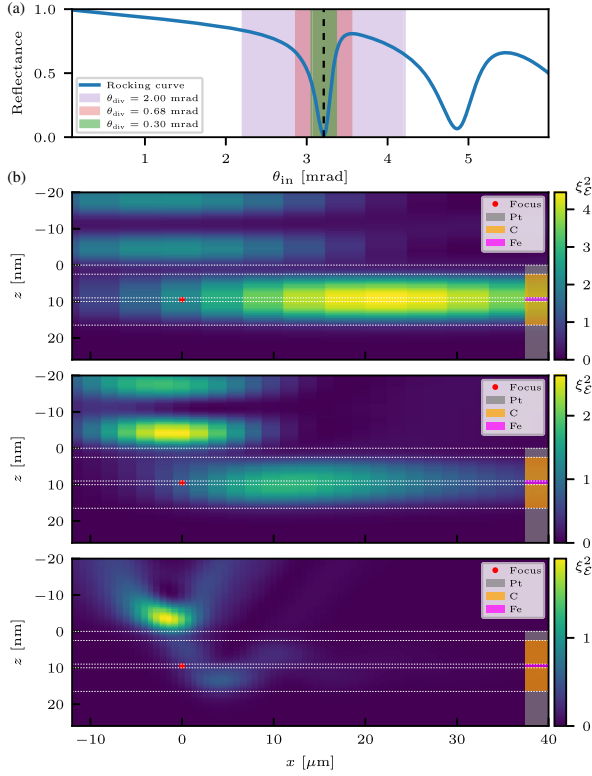


FIG. 2. Results for an example cavity following standard design criteria. (a) Electronic reflection as a function of incidence angle (rocking curve). The shaded areas indicate three different input beam divergence ranges considered in panel (b); see legend. (b) Spatially resolved cavity enhancement factor ξ_E^2 in and around the example cavity (cladding boundary at $z = 0$) for the beam divergences $\theta_{\text{div}} = 0.3, 0.685$, and 2.0 mrad (top to bottom), where the middle value corresponds to 40 nm spot size. The incident beam propagates from the top left to the bottom right and encounters the cavity starting from $z = 0$. The colored areas and white horizontal lines indicate the location of the cavity layers (see legend). The red dot indicates the free-space beam focus, which is placed at the depth of the nuclear ensemble.

We start with the discussion of a standard cavity design [16,71] in Fig. 2. Panel (a) depicts the electronic reflectance as a function of grazing incidence angle (rocking curve), featuring two cavity resonances. Following standard design criteria as well as commonly used cladding and spacer materials, the first resonance is relatively narrow and critically coupled [16,46,56], i.e., the resonant electronic reflectance vanishes [72].

Panel (b) shows the spatially resolved cavity enhancement factor inside and outside the cavity for three different x-ray focusing strengths (top to bottom). The corresponding beam divergences $\theta_{\text{div}} = 0.3, 0.685$, and 2.0 mrad are indicated as shaded regions in (a). x is the projection of the pulse propagation direction onto the waveguide surface, and z the surface normal.

At low focusing (panel b, top), the field enhancement inside the cavity is clearly visible, arising from the

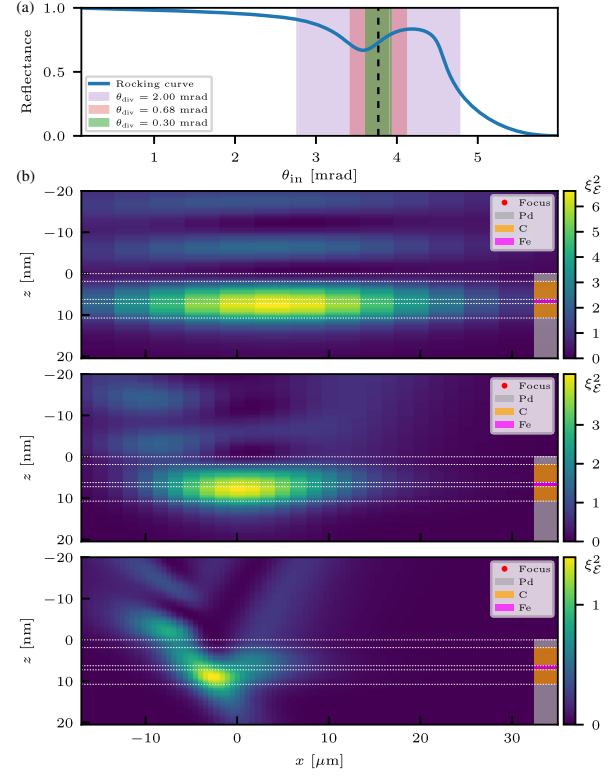


FIG. 3. Analogous to Fig. 2, but for a cavity optimized for a focal spot size of 40 nm. Note that the free-space focus is located above the plot limits at 33.2 nm as a result of the optimization. Parameters of the optimized cavity are summarized in [71].

constructive interference of multiple x-ray reflections. As desired, the intensity maximum is at the position of the nuclei.

Toward higher focusing [panel (b), top to bottom] the intensity enhancement reduces. On the one hand, with increasing beam divergence, a growing part of the incident wave vector components is off-resonant and cannot contribute to the standing-wave enhancement. Also, the focused wave packets become smaller such that the spatial overlap of the different reflections diminishes, again impeding the constructive interference leading to the intensity enhancement. The latter effect is visible in the highly focused case [(b), bottom], and implies a practical lower spot-size limit for the focusing. In addition to the enhancement reduction, most of the wave packet is reflected already at the outer cladding. Thus, the intensity at the nuclei's positions is in fact lower than it would be in the absence of the cavity.

From these results, we conclude that standard cavity designs are unsuitable in combination with focusing. However, these limitations can be overcome using alternative cavity designs. To this end, we choose a moderately strong focusing spot size of 40 nm and optimized the cavity layer structure for maximum intensity enhancement at the position of the nuclei (for details on the optimization,

see [71]) in Fig. 3. In comparison to the standard cavity in Fig. 2, we observe two qualitative differences. First, the rocking curve in panel (a) features a relevant resonance that is rather weak and surprisingly broad. Second, the optimized cavity [structure see Fig. 3, panel (a)] enables a significant intensity enhancement even at higher focusing [panel (b), top to bottom]. This improvement is due to the larger acceptance angle of broader cavity resonances.

After discussing the qualitative aspects of focusing on the nuclear excitation dynamics, we now turn to quantitative predictions for the Mössbauer isotopes ^{57}Fe , ^{187}Os , ^{169}Tm , and ^{119}Sn . We determine the necessary source parameter $\chi_{\text{source}}^{\text{nec}}$ to reach nuclear inversion as a function of the free-space beam spot size w_0 for each of these isotopes. For comparison, we further indicate the source conditions available at the European XFEL [73–76] and the projected conditions at the planned oscillator upgrade [77] (detailed data tables are provided in [58]).

Results are shown in Fig. 4. As a reference, panel (a) illustrates the case without cavity environment, for which $\chi_{\text{source}}^{\text{nec}} \propto \omega_{\text{nuc}}$. In the relevant hard x-ray energy range, nanofocusing setups at EuXFEL beamlines currently

offer spot sizes down to about 40 nm [75] (vertical dashed line). At this focusing strength, present-day beam conditions at EuXFEL are still far below the requirements for full inversion. However, XFEL conditions [77] already approach the required χ_{source} values and significant σ^z can be reached, indicating the onset of the nonlinear regime.

Panel (b) shows corresponding results with waveguide enhancements. We optimized the cavity structure for maximum excitation enhancement at each spot size using the algorithm developed in the companion paper [58]. The optimization considers the thicknesses of the top cladding layer and the two carbon spacer layers in a five-layer stack with the resonant isotope (^{57}Fe as a concrete example in Figs. 2 and 3) in the center, a selection of cladding materials, as well as the incidence angle and the focal spot depth. Further details on the computational approach to the optimization are provided in [71]. We see that the cavity enhancement indeed allows one to reduce the source requirements, such that the projected XFEL parameters even exceed the requirements for inversion with ^{187}Os and ^{169}Tm . The improvement by the cavity is quantified in panel (c), which shows the ratios of the required source

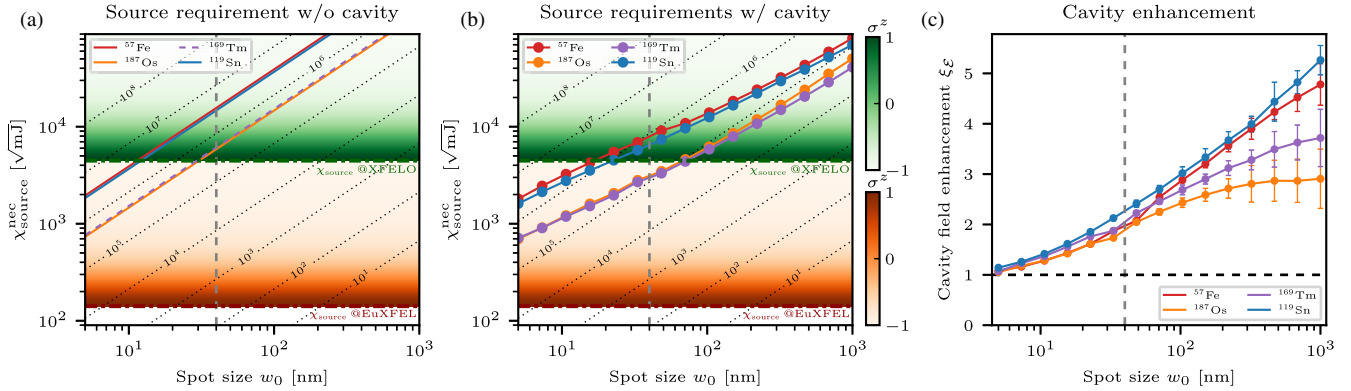


FIG. 4. Parameter maps characterizing achievable conditions (including achievable excitation and radiation damage for different sources and isotopes) with (a) and without (b) cavity. Inversion conditions: the plots do not only show the result for a given x-ray source and isotope, but instead allow to read off the excitation for any given conditions. To this end, the y axis depicts the necessary source requirement $\chi_{\text{source}}^{\text{nec}} = \sqrt{E_{\text{pulse}}^{\text{nec}}/b_r^{\text{nec}}}$, with E_{pulse} the pulse energy and b_r the relative bandwidth. For a given isotope and spot size w_0 (x axis), one can then compute the $\chi_{\text{source}}^{\text{nec}}$ via Eq. (3). Examples for the isotopes ^{57}Fe , ^{187}Os , ^{169}Tm , and ^{119}Sn are shown as colored lines in the parameter space (see legend). These lines indicate how large the source parameter of a given source would have to be to reach full inversion at each spot size. In (a), the results are based on analytic formulas. In (b), the cavity environment was optimized at the spot sizes indicated by the dots, using the formalism developed in [58]. Actual excitation: to obtain the actual achievable excitation, one has to compare to available source parameters χ_{source} . The case of the EuXFEL (red) and the EuXFEL with the upcoming oscillator upgrade (green) are shown as horizontal dash-dotted lines. At the point where these lines cross the $\chi_{\text{source}}^{\text{nec}}$ lines—as well as the areas below (for lower $\chi_{\text{source}}^{\text{nec}}$)—for each isotope, full inversion is predicted to be feasible. Above these lines, lower excitation is achieved according to $\sigma^z = -\cos(\pi\chi_{\text{source}}/\chi_{\text{source}}^{\text{nec}})$, which is shown as a white-to-green and white-to-orange color map for the EuXFEL and EuXFELo, respectively. Again, these color maps characterize all possible isotope conditions for the given source. The actual excitation value for a given isotope then has to be read off at the location of the $\chi_{\text{source}}^{\text{nec}}$ line. Radiation damage: an additional quantity of interest is the x-ray fluence per pulse bandwidth at the sample (a) or cavity surface (b), which allows to estimate the radiation damage on the targets for the given conditions. Similarly to the color maps of the excitation, this quantity is shown as dotted contour lines on the parameter map (numbers on the contours in units of $(\mu\text{J}/\mu\text{m}^2 \text{ meV})$). Actual fluences are obtained by multiplying this value with the absolute source bandwidth. Cavity enhancement: panel (c) additionally shows the cavity enhancement factor for the isotopes and optimized cavities in (b). The error bars quantify the dependence of the enhancement on the precise optimized structure (see main text), quantifying manufacturing imperfections.

parameters with and without cavity. It also includes error bars that quantify the dependence of the enhancement on imperfections in the optimum cavity structure. For this, we randomly modified the top-layer and the two spacer-layer thicknesses with imperfections drawn from a Gaussian distribution of 0.3 nm width. Repeating this 250 times yields a histogram of enhancements, the standard deviation of which quantifies the stability of the cavity enhancement against manufacturing imperfections, and is the quantity shown as error bar in panel (c). We see that enhancements in the pulse area are possible even at substantial focusing. Note that away from full inversion, which presently is experimentally most relevant, the excitation scales quadratically with the pulse area, such that nonlinear effects are enhanced quadratically.

A further important practical question is whether the target can survive possible radiation damage. To this end, Fig. 4 also shows the fluences per pulse bandwidth on the sample surface. Damage thresholds of materials in the x-ray regime have been much investigated (see, e.g., [78,79]) and typical values in combination with Fig. 4 indicate that experiments may be feasible depending on the level of monochromatization. Alternatively, the small focal spot can be scanned across the cavity surface from pulse to pulse. In any case, the focusing strength plays a crucial role with regards to practical feasibility: a stronger focus is necessary for strong excitation, but simultaneously increases radiation damage. Figure 4 shows that cavities can further help in this regard, since in free space, the fluence contours have the same slope as the excitation contours, but for the optimized cavities, the slope of the latter is changed.

In summary, we have shown that optimized thin-film cavities provide means to significantly boost Mössbauer nuclei toward the nonlinear optics regime even for tight focusing. The enhancement can be interpreted as a longitudinal compression of the pulse via multipath interference in the cavity. In this sense, cavities are complementary to focusing techniques, which provide a transversal compression of the light field. We find to achieve significant enhancements down to 40 nm focusing, special optimized cavities structures are necessary. With planned upgrades to currently available sources, the resulting enhancement enables excitation of nuclei into the nonlinear regime. In particular for the isotopes ^{187}Os and ^{169}Tm , inversion is predicted to be achievable and ^{57}Fe —the workhorse of Mössbauer science—is boosted to significantly nonlinear excitation fractions. Our results thereby outline a road map for further source developments and experimental efforts. Progressing beyond the state-of-the-art linear regime [16–22,24,28] would then allow one to access an unexplored parameter regime, including the impact of the electronic, phononic, and thermal environment on the nuclei, and potentially enable the creation of correlated x-ray photons [35–37].

The software and figures in this Letter made use of the NUMPY [80], SCIPY [81], and MATPLOTLIB [82] software packages.

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- [1] G. C. Baldwin, J. C. Solem, and V. I. Gol’danskii, Approaches to the development of gamma-ray lasers, *Rev. Mod. Phys.* **53**, 687 (1981).
 - [2] R. L. Mössbauer, Kernresonanzfluoreszenz von gammastrahlung in ^{191}Ir , *Z. Phys.* **151**, 124 (1958).
 - [3] R. L. Mössbauer, Recoilless nuclear resonance absorption of gamma radiation, *Science* **137**, 731 (1962).
 - [4] O. Kocharovskaya, Amplification and lasing without inversion, *Phys. Rep.* **219**, 175 (1992).
 - [5] G. C. Baldwin and J. C. Solem, Recoilless gamma-ray lasers, *Rev. Mod. Phys.* **69**, 1085 (1997).
 - [6] C. Pellegrini, A. Marinelli, and S. Reiche, The physics of x-ray free-electron lasers, *Rev. Mod. Phys.* **88**, 015006 (2016).
 - [7] I. Georgescu, The first decade of XFELs, *Nat. Rev. Phys.* **2**, 345 (2020).
 - [8] A. L. Schawlow and C. H. Townes, Infrared and optical masers, *Phys. Rev.* **112**, 1940 (1958).
 - [9] W. E. Lamb, W. P. Schleich, M. O. Scully, and C. H. Townes, Laser physics: Quantum controversy in action, *Rev. Mod. Phys.* **71**, S263 (1999).
 - [10] Nonlinear X-Ray Spectroscopy for Materials Science, edited by I. Matsuda and R. Arafune, Springer Series in Optical Sciences Vol. 246 (Springer, Singapore, 2023), pp. XIV.
 - [11] N. Rohringer, X-ray raman scattering: A building block for nonlinear spectroscopy, *Phil. Trans. R. Soc. A* **377**, 20170471 (2019).
 - [12] S. Sofer, E. Strizhevsky, A. Schori, K. Tamasaku, and S. Schwartz, Quantum enhanced x-ray detection, *Phys. Rev. X* **9**, 031033 (2019).
 - [13] S. Nandi *et al.*, Observation of rabi dynamics with a short-wavelength free-electron laser, *Nature (London)* **608**, 488 (2022).
 - [14] G. Doumy *et al.*, Nonlinear atomic response to intense ultrashort x rays, *Phys. Rev. Lett.* **106**, 083002 (2011).
 - [15] N. Rohringer, D. Ryan, R. A. London, M. Purvis, F. Albert, J. Dunn, J. D. Bozek, C. Bostedt, A. Graf, R. Hill, S. P. Hau-Riege, and J. J. Rocca, Atomic inner-shell x-ray laser at 1.46 nanometres pumped by an x-ray free-electron laser, *Nature (London)* **481**, 488 (2012).
 - [16] R. Röhlberger, K. Schlage, B. Sahoo, S. Couet, and R. Ruffer, Collective lamb shift in single-photon superradiance, *Science* **328**, 1248 (2010).

- [17] R. Röhlberger, H.-C. Wille, K. Schlage, and B. Sahoo, Electromagnetically induced transparency with resonant nuclei in a cavity, *Nature (London)* **482**, 199 (2012).
- [18] K.P. Heeg, H.-C. Wille, K. Schlage, T. Guryeva, D. Schumacher, I. Uschmann, K.S. Schulze, B. Marx, T. Kämpfer, G.G. Paulus, R. Röhlberger, and J. Evers, Vacuum-assisted generation and control of atomic coherences at x-ray energies, *Phys. Rev. Lett.* **111**, 073601 (2013).
- [19] K.P. Heeg, C. Ott, D. Schumacher, H.-C. Wille, R. Röhlberger, T. Pfeifer, and J. Evers, Interferometric phase detection at x-ray energies via Fano resonance control, *Phys. Rev. Lett.* **114**, 207401 (2015).
- [20] K.P. Heeg, J. Haber, D. Schumacher, L. Bocklage, H.-C. Wille, K.S. Schulze, R. Loetzsch, I. Uschmann, G.G. Paulus, R. Ruffer, R. Röhlberger, and J. Evers, Tunable subluminal propagation of narrow-band x-ray pulses, *Phys. Rev. Lett.* **114**, 203601 (2015).
- [21] J. Haber, K.S. Schulze, K. Schlage, R. Loetzsch, L. Bocklage, T. Guryeva, H. Bernhardt, H.-C. Wille, R. Ruffer, I. Uschmann, G.G. Paulus, and R. Röhlberger, Collective strong coupling of x-rays and nuclei in a nuclear optical lattice, *Nat. Photonics* **10**, 445 EP (2016).
- [22] J. Haber, X. Kong, C. Strohm, S. Willing, J. Gollwitzer, L. Bocklage, R. Ruffer, A. Pálffy, and R. Röhlberger, Rabi oscillations of x-ray radiation between two nuclear ensembles, *Nat. Photonics* **11**, 720 (2017).
- [23] P. Helistö, I. Tittonen, M. Lippmaa, and T. Katila, Gamma echo, *Phys. Rev. Lett.* **66**, 2037 (1991).
- [24] Y.V. Shvyd'ko, T. Hertrich, U. van Bürck, E. Gerdau, O. Leupold, J. Metge, H.D. Rüter, S. Schwendy, G.V. Smirnov, W. Potzel, and P. Schindelmann, Storage of nuclear excitation energy through magnetic switching, *Phys. Rev. Lett.* **77**, 3232 (1996).
- [25] P. Schindelmann, U. van Bürck, W. Potzel, G.V. Smirnov, S.L. Popov, E. Gerdau, Y.V. Shvyd'ko, J. Jäschke, H.D. Rüter, A.I. Chumakov, and R. Ruffer, Radiative decoupling and coupling of nuclear oscillators by stepwise Doppler-energy shifts, *Phys. Rev. A* **65**, 023804 (2002).
- [26] F. Vagizov, V. Antonov, Y.V. Radeonychev, R.N. Shakhmuratov, and O. Kocharovskaya, Coherent control of the waveforms of recoilless γ -ray photons, *Nature (London)* **508**, 80 (2014).
- [27] S. Sakshath, K. Jenni, L. Scherthan, P. Würtz, M. Herlitschke, I. Sergeev, C. Strohm, H.-C. Wille, R. Röhlberger, J.A. Wolny, and V. Schünemann, Optical pump—nuclear resonance probe experiments on spin cross-over complexes, *Hyperfine Interact.* **238**, 89 (2017).
- [28] K.P. Heeg, A. Kaldun, C. Strohm, P. Reiser, C. Ott, R. Subramanian, D. Lentrodt, J. Haber, H.-C. Wille, S. Goerttler, R. Ruffer, C.H. Keitel, R. Röhlberger, T. Pfeifer, and J. Evers, Spectral narrowing of x-ray pulses for precision spectroscopy with nuclear resonances, *Science* **357**, 375 (2017).
- [29] K.P. Heeg, A. Kaldun, C. Strohm, C. Ott, R. Subramanian, D. Lentrodt, J. Haber, H.-C. Wille, S. Goerttler, R. Ruffer, C.H. Keitel, R. Röhlberger, T. Pfeifer, and J. Evers, Coherent x-ray-optical control of nuclear excitons, *Nature (London)* **590**, 401 (2021).
- [30] L. Bocklage, J. Gollwitzer, C. Strohm, C.F. Adolff, K. Schlage, I. Sergeev, O. Leupold, H.-C. Wille, G. Meier, and R. Röhlberger, Coherent control of collective nuclear quantum states via transient magnons, *Sci. Adv.* **7**, eabc3991 (2021).
- [31] R. Röhlberger, *Nuclear Condensed Matter Physics with Synchrotron Radiation*, Springer Tracts in Modern Physics Vol. 208 (Springer, Berlin, Heidelberg, 2005).
- [32] W. Sturhahn, Conus and phoenix: Evaluation of nuclear resonant scattering data, *Hyperfine Interact.* **125**, 149 (2000).
- [33] M.T. Manzoni, D.E. Chang, and J.S. Douglas, Simulating quantum light propagation through atomic ensembles using matrix product states, *Nat. Commun.* **8**, 1743 (2017).
- [34] S. Mahmoodian, M. Čepulkovskis, S. Das, P. Lodahl, K. Hammerer, and A.S. Sørensen, Strongly correlated photon transport in waveguide quantum electrodynamics with weakly coupled emitters, *Phys. Rev. Lett.* **121**, 143601 (2018).
- [35] A.S. Prasad, J. Hinney, S. Mahmoodian, K. Hammerer, S. Rind, P. Schneeweiss, A.S. Sørensen, J. Volz, and A. Rauschenbeutel, Correlating photons using the collective nonlinear response of atoms weakly coupled to an optical mode, *Nat. Photonics* **14**, 719 (2020).
- [36] M. Cordier, M. Schemmer, P. Schneeweiss, J. Volz, and A. Rauschenbeutel, Tailoring photon statistics with an atom-based two-photon interferometer, *Phys. Rev. Lett.* **131**, 183601 (2023).
- [37] C. Liedl, F. Tebbenjohanns, C. Bach, S. Pucher, A. Rauschenbeutel, and P. Schneeweiss, Observation of super-radiant bursts in a cascaded quantum system, *Phys. Rev. X* **14**, 011020 (2024).
- [38] A.I. Chumakov, A.Q.R. Baron, I. Sergueev, C. Strohm, O. Leupold, Y. Shvyd'ko, G.V. Smirnov, R. Ruffer, Y. Inubushi, M. Yabashi, K. Tono, T. Kudo, and T. Ishikawa, Superradiance of an ensemble of nuclei excited by a free electron laser, *Nat. Phys.* **14**, 261 (2018).
- [39] Y. Shvyd'ko *et al.*, Resonant x-ray excitation of the nuclear clock isomer ^{45}Sc , *Nature (London)* **622**, 471 (2023).
- [40] S. Matinyan, Lasers as a bridge between atomic and nuclear physics, *Phys. Rep.* **298**, 199 (1998).
- [41] T.J. Bürvenich, J. Evers, and C.H. Keitel, Nuclear quantum optics with x-ray laser pulses, *Phys. Rev. Lett.* **96**, 142501 (2006).
- [42] A. Pálffy, J. Evers, and C.H. Keitel, Electric-dipole-forbidden nuclear transitions driven by super-intense laser fields, *Phys. Rev. C* **77**, 044602 (2008).
- [43] K.P. Heeg, X-ray quantum optics with Mössbauer nuclei in thin-film cavities (2014), [10.11588/heidok.00017869](https://arxiv.org/abs/10.11588/heidok.00017869).
- [44] A. Junker, A. Pálffy, and C.H. Keitel, Cooperative effects in nuclear excitation with coherent x-ray light, *New J. Phys.* **14**, 085025 (2012).
- [45] T. Salditt and M. Osterhoff, X-ray focusing and optics, in *Nanoscale Photonic Imaging*, edited by T. Salditt, A. Egner, and D.R. Luke (Springer International Publishing, Cham, 2020), pp. 71–124.
- [46] F. Schenk, *Optimization of Resonances for Multilayer X-Ray Resonators*, Göttingen Series in X-Ray Physics Vol. 003 (Universitätsverlag Göttingen, Göttingen, 2011).
- [47] L.M. Lohse, P. Andrejić, S. Velten, M. Vassholz, C. Neuhaus, A. Negi, A. Panchwanee, I. Sergeev, A. Pálffy, T. Salditt, and R. Röhlberger, Collective nuclear excitation dynamics in mono-modal x-ray waveguides, [arXiv:2403.06508](https://arxiv.org/abs/2403.06508).

- [48] L. M. Lohse and P. Andrejić, Nano-optical theory of planar x-ray waveguides, *Opt. Express* **32**, 9518 (2024).
- [49] H.-Y. Chen, S. Hoffmann, and T. Salditt, X-ray beam compression by tapered waveguides, *Appl. Phys. Lett.* **106**, 194105 (2015).
- [50] Y.-H. Chen, P.-H. Lin, G.-Y. Wang, A. Pálffy, and W.-T. Liao, Transient nuclear inversion by x-ray free electron laser in a tapered x-ray waveguide, *Phys. Rev. Res.* **4**, L032007 (2022).
- [51] R. Röhlsberger, J. Evers, and S. Shwartz, Quantum and nonlinear optics with hard x-rays, in *Synchrotron Light Sources and Free-Electron Lasers: Accelerator Physics, Instrumentation and Science Applications*, edited by E. Jaeschke, S. Khan, J. R. Schneider, and J. B. Hastings (Springer International Publishing, Cham, 2014), pp. 1–28.
- [52] R. Röhlsberger and J. Evers, Quantum optical phenomena in nuclear resonant scattering, in *Modern Mössbauer Spectroscopy*, edited by Y. Yoshida and G. Langouche (Springer, Singapore, 2021), pp. 105–171.
- [53] R. Röhlsberger, K. Schlage, T. Klein, and O. Leupold, Accelerating the spontaneous emission of x rays from atoms in a cavity, *Phys. Rev. Lett.* **95**, 097601 (2005).
- [54] K. P. Heeg and J. Evers, X-ray quantum optics with Mössbauer nuclei embedded in thin-film cavities, *Phys. Rev. A* **88**, 043828 (2013).
- [55] D. Lentrodt, K. P. Heeg, C. H. Keitel, and J. Evers, Ab initio quantum models for thin-film x-ray cavity QED, *Phys. Rev. Res.* **2**, 023396 (2020).
- [56] O. Diekmann, D. Lentrodt, and J. Evers, Inverse design approach to x-ray quantum optics with Mössbauer nuclei in thin-film cavities, *Phys. Rev. A* **105**, 013715 (2022).
- [57] K. P. Heeg, C. H. Keitel, and J. Evers, Inducing and detecting collective population inversions of mössbauer nuclei, [arXiv:1607.04116](https://arxiv.org/abs/1607.04116).
- [58] D. Lentrodt, C. H. Keitel, and J. Evers, companion paper, Excitation of narrow x-ray transitions in thin-film cavities by focused pulses, *Phys. Rev. A* **112**, 013711 (2025).
- [59] A. Asenjo-Garcia, J. D. Hood, D. E. Chang, and H. J. Kimble, Atom-light interactions in quasi-one-dimensional nanostructures: A Green's-function perspective, *Phys. Rev. A* **95**, 033818 (2017).
- [60] C. D. Mink and M. Fleischhauer, Collective radiative interactions in the discrete truncated Wigner approximation, *SciPost Phys.* **15**, 233 (2023).
- [61] J. Schachenmayer, A. Pikovski, and A. M. Rey, Many-body quantum spin dynamics with Monte Carlo trajectories on a discrete phase space, *Phys. Rev. X* **5**, 011022 (2015).
- [62] M. Moreno-Cardoner, D. Goncalves, and D. E. Chang, Quantum nonlinear optics based on two-dimensional Rydberg atom arrays, *Phys. Rev. Lett.* **127**, 263602 (2021).
- [63] D. Lentrodt, Ab initio approaches to x-ray cavity QED (2021), [10.11588/heidok.00030671](https://arxiv.org/abs/10.11588/heidok.00030671).
- [64] J. Amann *et al.*, Demonstration of self-seeding in a hard-x-ray free-electron laser, *Nat. Photonics* **6**, 693 (2012).
- [65] I. Nam *et al.*, High-brightness self-seeded x-ray free-electron laser covering the 3.5 keV to 14.6 keV range, *Nat. Photonics* **15**, 435 (2021).
- [66] S. Liu *et al.*, Cascaded hard x-ray self-seeded free-electron laser at megahertz repetition rate, *Nat. Photonics* **17**, 984 (2023).
- [67] S. L. McCall and E. L. Hahn, Self-induced transparency by pulsed coherent light, *Phys. Rev. Lett.* **18**, 908 (1967).
- [68] J. Eberly, Area theorem rederived, *Opt. Express* **2**, 173 (1998).
- [69] B. W. Shore, *Manipulating Quantum Structures Using Laser Pulses* (Cambridge University Press, Cambridge, England, 2011).
- [70] L. Allen and J. Eberly, *Optical Resonance and Two-level Atoms*, Dover Books on Physics and Chemistry (Dover, New York, 1987).
- [71] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/1lcz-r8d3> for details on the numerical algorithm to compute the field enhancement of focused x-ray beams in thin-film cavities.
- [72] R. Röhlsberger, H. Thomas, K. Schlage, E. Burkel, O. Leupold, and R. Rüffer, Imaging the magnetic spin structure of exchange-coupled thin films, *Phys. Rev. Lett.* **89**, 237201 (2002).
- [73] T. Tschentscher, C. Bressler, J. Grünert, A. Madsen, A. P. Mancuso, M. Meyer, A. Scherz, H. Sinn, and U. Zastrau, Photon beam transport and scientific instruments at the European XFEL, *Appl. Sci.* **7**, 592 (2017).
- [74] A. Madsen, J. Hallmann, G. Ansal di, T. Roth, W. Lu, C. Kim, U. Boesenberg, A. Zozulya, J. Möller, R. Shayduk, M. Scholz, A. Bartmann, A. Schmidt, I. Lobato, K. Sukharnikov, M. Reiser, K. Kazarian, and I. Petrov, Materials imaging and dynamics (MID) instrument at the European x-ray free-electron laser facility, *J. Synchrotron Radiat.* **28**, 637 (2021).
- [75] A. Madsen, J. Hallmann, T. Roth, and G. Ansal di, Technical design report—scientific instrument MID, https://www.xfel.eu/sites/sites_custom/site_xfel/content/e35165/e46561/e46883/e46942/e46945/xfel_file46946/TR-2013-005_TDR_MID_eng.pdf (2013), accessed: 2021-07-25.
- [76] M. Nakatsutsumi, K. Appel, G. Priebe, I. Thorpe, A. Pelka, B. Muller, and T. Tschentscher, Technical design report—scientific instrument HED, https://www.xfel.eu/sites/sites_custom/site_xfel/content/e35165/e46561/e46886/e46954/e46959/xfel_file46960/TR-2014-001_TDR_HED_eng.pdf (2014), accessed: 2021-07-25.
- [77] B. Adams *et al.*, Scientific opportunities with an x-ray free-electron laser oscillator, [arXiv:1903.09317](https://arxiv.org/abs/1903.09317).
- [78] R. Follath, T. Koyama, V. Lipp, N. Medvedev, K. Tono, H. Ohashi, L. Patthey, M. Yabashi, and B. Ziaja, X-ray induced damage of B₄C-coated bilayer materials under various irradiation conditions, *Sci. Rep.* **9**, 2029 (2019).
- [79] J. Kim, A. Nagahira, T. Koyama, S. Matsuyama, Y. Sano, M. Yabashi, H. Ohashi, T. Ishikawa, and K. Yamauchi, Damage threshold of platinum/carbon multilayers under hard x-ray free-electron laser irradiation, *Opt. Express* **23**, 29032 (2015).
- [80] C. R. Harris *et al.*, Array programming with NUMPY, *Nature (London)* **585**, 357 (2020).
- [81] P. Virtanen *et al.* (SciPy 1.0 Contributors), SCIPY 1.0: Fundamental algorithms for scientific computing in Python, *Nat. Methods* **17**, 261 (2020).
- [82] J. D. Hunter, MATPLOTLIB: A 2d graphics environment, *Comput. Sci. Eng.* **9**, 90 (2007).

End Matter

End Matter—To supplement the discussion in the main text, Fig. 5 shows analogous results to Fig. 4 for a fixed cavity. That is instead of optimizing the cavity structure for each spot size, the behavior of the cavity optimized for a spot size of 40 nm is investigated. In this case, the

cavity enhancement levels off more rapidly, such that the necessary source parameter scales the same way as the radiation damage starting at a spot size of around 70 nm. This observation further illustrates how optimized cavities can be useful to mitigate radiation damage.

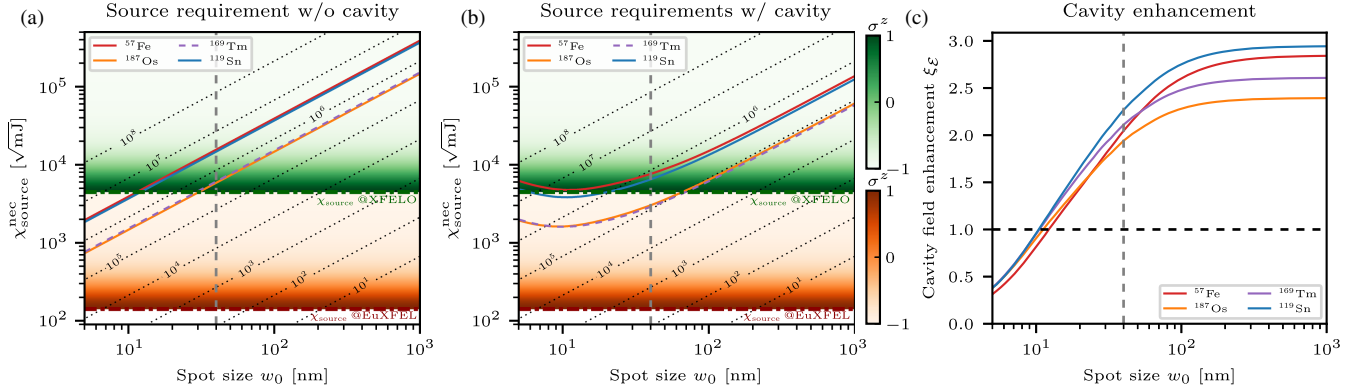


FIG. 5. Source requirements for a specified cavity. This figure is analogous to Fig. 4, involving the same panels (a)–(c) but uses a single cavity geometry optimized for a spot size of 40 nm focusing instead of at each spot size in order to illustrate the behavior of the cavity structures discussed in the main text.