

Superradiant Parametric Mössbauer Radiation Source

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(Received 1 January 2025; accepted 8 May 2026; published 15 July 2026)

Future applications of the spectrally narrow x-ray resonances in Mössbauer nuclei require x-ray sources with exceptionally high peak and average spectral flux. Here, we introduce a superradiant parametric Mössbauer radiation (SPMR) source, which utilizes the scattering of spatially microstructured electron bunches produced in x-ray free-electron laser (XFEL) accelerators on crystals. The spatial structuring leads to a coherent add-up of the radiation from different electrons, thereby enhancing the Mössbauer radiation by many orders of magnitude. We find that the performance is optimized at qualitatively different operation conditions than considered so far. For a focused electron beam with parameters based on the European XFEL facility, our approach predicts the generation of more than 900 SPMR photons per pulse within one linewidth of the $^{57}_{26}\text{Fe}$ Mössbauer resonance. This opens up new opportunities for precision metrology, and for hard x-ray pump-probe spectroscopy techniques via the parallel use of the bunched XFEL electron and photon beams.

DOI: [10.1103/PhysRevLett.137.035001](https://doi.org/10.1103/PhysRevLett.137.035001)

Introduction—Ongoing progress in the development of accelerator-based x-ray sources [1–6] is opening access to as-yet unexplored parameter regimes in Mössbauer science [7,8], which bear potential for a broad range of applications [9–15]. So far, multiexcitation effects [16,17] and the excitation of ultranarrow Mössbauer resonances [18,19] have been experimentally demonstrated at (self-seeded) x-ray free-electron lasers (XFELs). However, efficient excitation of Mössbauer resonances remains challenging, since their transition linewidths are orders of magnitude narrower than the spectral widths of the x-ray pulses, such that only a tiny pulse fraction interacts with the nuclei. Concepts under development such as the self-seeded XFELs [20–24] and the x-ray free-electron laser oscillator (XFELo) [25–28] promise a smaller relative pulse bandwidth, thereby increasing the resonant photon component. Nevertheless, source improvement remains a challenge for future Mössbauer applications.

An alternative source approach involves parametric x-ray radiation (PXR) [29], based on relativistic electrons passing through crystals (see Ref. [30] for related work). PXR is a versatile x-ray source [31,32] with many applications [33–37]. It is governed by the electronic crystal susceptibilities, which characterize the interaction between the

electrons and the periodically arranged atoms. Analogously, parametric Mössbauer radiation (PMR) can be generated via nuclear susceptibilities, with interesting applications for Mössbauer science [38–41]. Electrons can excite Mössbauer isotopes with higher nuclear transition energies beyond the reach of existing XFEL facilities. Also, particular crystals allow for the generation of highly monochromatized x-rays favorable for Mössbauer applications. To maximize the photon yield, the so-called grazing-extremely asymmetric diffraction (EAD) geometry was suggested [38,42,43], which minimizes the propagation length of the generated radiation through the attenuating crystal. Nevertheless, despite their promising concept, the PMR sources suggested so far are too weak to be of practical interest.

Recently, a qualitative enhancement of PXR was predicted by advancing from incoherent electrons to spatially microbunched beams generated via self-amplified spontaneous emission (SASE) in XFELs [44–46]. The spatial structuring on the radiation wavelength scale leads to constructive interference between the radiation generated from different electrons, amplifying PXR by many orders of magnitude. This prompts the question, if a superradiant enhancement could advance PMR to a Mössbauer source of practical interest.

In this Letter, we generalize the concept of superradiant amplification to PMR. The resulting superradiant parametric Mössbauer radiation (SPMR) source exceeds regular PMR in the relevant performance measures by many orders of magnitude, to a point where SPMR becomes practically relevant and comparable to undulator radiation from self-seeded XFELs. However, the structuring of the electron beam alone is not sufficient for these results. In particular,

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we show that the grazing EAD geometry considered so far is not favorable. Instead, the performance is optimized in a qualitatively different geometry, where the crystal diffraction considered so far is strongly attenuated, the electron beam is moving above the crystal rather than inside, and where the competing mechanism of specular diffraction dominates over the Bragg diffraction inside the crystal for the Mössbauer photon generation. For this optimization of geometry, we utilize a comprehensive theoretical framework for PMR and SPMR described in all detail in a companion paper [47]. It applies to arbitrary emission directions, including the grazing EAD case where approximations commonly applied break down. Our calculations predict the generation of more than 900 SPMR photons per pulse within a linewidth of the ^{57}Fe Mössbauer isotope, using the enriched α -iron crystal and assuming a focused electron beam with parameters based on the European XFEL facility. This renders SPMR a competitive source for the exploration of Mössbauer science beyond the reach of synchrotron facilities [11,16,18].

Qualitative description—PXR arises from the scattering of the virtual-photon fields of relativistic charged particles on crystallographic lattice planes. The virtual photons of the frequency ω have the wave vector $\mathbf{k}_v = \omega \mathbf{v}/v^2$, with the electron velocity $\mathbf{v}$ [29]. The crystallographic lattice plane and its orientation determine the reciprocal lattice vector $\mathbf{g}$, which in principle allows for a tuning of the PXR emission frequency over a broad range. In contrast, the core challenge of SPMR is the stringent requirement set by the precisely defined Mössbauer resonance frequency ω_0 : the Bragg diffraction, the superradiant enhancement of radiation, and the Mössbauer resonance must all coincide at the same frequency. This severely limits the parameter space available for optimizations.

Superradiant x-ray emission may occur in XFEL undulators when the electron bunch is short as compared to the radiation wavelength [48], analogous to the condition for Dicke superradiance. Alternatively, a larger electron bunch can be structured into multiple independent microbunches, see Fig. 1. If each microbunch remains short relative to the radiation wavelength, and the separation d_0 between adjacent microbunches satisfies $\Delta\phi = k_v d_0 = 2n\pi$ with $k_v = |\mathbf{k}_v|$, then all electrons are still in equivalent positions with respect to a single emission mode [47,48]. Thus, superradiance occurs, with the intensity scaling with the total electron number squared [44,48]. The condition on $\Delta\phi$ translates to discrete emission harmonics $\omega_s^{(n)} = 2\pi v n/d_0$ ($n \in \{1, 2, \dots\}$), of which one harmonic has to coincide with the Mössbauer resonance frequency $\omega_s^{(n)} = \omega_0$ for SPMR to occur. These considerations fix the electron beam parameters.

Next, we describe the global optimization of SPMR regarding the scattering geometries. If the crystallographic lattice plane and the reciprocal lattice vector $\mathbf{g}$ are selected for maximal reflection, then only the geometric

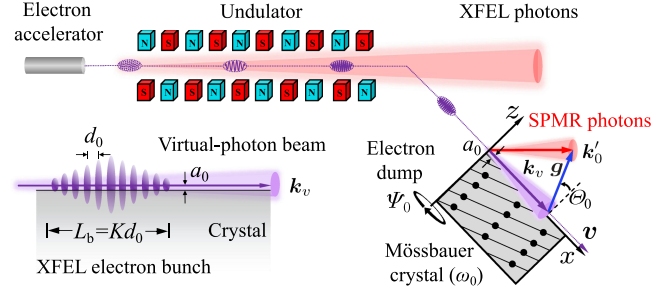


FIG. 1. Schematic setup for SPMR. Electron bunches exiting from an XFEL undulator interact with a crystal containing Mössbauer nuclei with the resonance frequency ω_0 . Due to the scattering of the electron virtual-photon field by the crystal, intense pulses of SPMR photons are generated under the Bragg condition $\mathbf{k}_v + \mathbf{g} = \mathbf{k}'_0$. Since the electron bunches of length $L_b = Kd_0$ are spatially structured by the SASE mechanism into K microbunches with distance d_0 , the photon generation is superradiantly enhanced. The generation of SPMR is governed by the competition between the virtual-photon fields of the electrons distributed outside and inside the crystal with the central virtual-photon wave vector $\mathbf{k}_v = (\omega/v)\mathbf{e}_x$, corresponding to the specular diffraction and the Bragg diffraction, respectively. This competition identifies an optimum geometry which crucially differs from the previously considered grazing EAD condition.

configuration remains to optimize SPMR. The diffraction geometry is determined by the transverse electron beam distance from the crystal surface, a_0 , and by four angles $(\vartheta_0, \varphi_0, \Theta_0, \Psi_0)$; see Fig. 1. The latter determine the Mössbauer emission wave vector $\mathbf{k}'_0 = -k_0(\sin \vartheta_0 \cos \varphi_0, \sin \vartheta_0 \sin \varphi_0, \cos \vartheta_0)$ with $k_0 = \omega_0/c$, and $\mathbf{g} = g(\sin \Theta_0 \cos \Psi_0, \sin \Theta_0 \sin \Psi_0, \cos \Theta_0)$. The angles ϑ_0 , φ_0 , and Ψ_0 , are fixed by the PXR Bragg condition [29]

$$\mathbf{k}_v + \mathbf{g} = \mathbf{k}'_0, \quad \frac{\omega_0}{v^2} \mathbf{v} + \mathbf{g} = \frac{\omega_0}{c} \mathbf{n}, \quad (1)$$

where $\mathbf{n} = \mathbf{k}'_0/k_0$. In the grazing EAD geometry considered for the regular PXR [42], SPXR [44], and PMR [38] so far, the crystal surface cutting angle is manually chosen as $\Theta_0 = 45^\circ$, and electron beam positions a_0 below the crystal surface were considered. In the following, as a key result, we show that optimizing Θ_0 and a_0 leads to a qualitatively different emission regime with strongly enhanced SPMR intensity.

Theoretical approach—We model the XFEL electron bunch with N_e electrons moving along the crystal surface with grazing-incidence velocity $\mathbf{v} = v\mathbf{e}_x$ (see Fig. 1) using Gaussian distributions [44,49,50],

$$\rho(\theta_e) = \frac{1}{\pi\sigma_a^2} e^{-\theta_e^2/\sigma_a^2}, \quad (2)$$

$$\mathcal{D}(\mathbf{r}_0) = \frac{1}{\pi\sigma_t^2} e^{-r_t^2/\sigma_t^2} \sum_{l=0}^K \frac{1}{\sqrt{\pi}\sigma_x} e^{-(x_0 - ld_0)^2/\sigma_x^2}. \quad (3)$$

Here, $\mathbf{r}_0$ is the initial electron position and $\boldsymbol{\theta}_e$ the orientation of the velocity. The parameters σ_t , σ_a , and σ_x characterize the transverse size and angular spread of the electron bunch, as well as the fluctuations around the microbunch period d_0 , respectively. Further, r_t is the transverse position of electrons, and $L_b = Kd_0$ the XFEL electron bunch length.

The number of PXR photons at the far-field position $\mathbf{r}$ with s polarization ($s = \sigma$ and π) emitted in the solid angle $d\Omega$ around the direction $\mathbf{n} = \mathbf{r}/r$ and in the frequency interval $d\omega$ around the emission frequency ω is [44,47]

$$\frac{\partial^2 N_{n,\omega s}}{\partial \omega \partial \Omega} = \frac{e_0^2 \omega}{16\pi^3 \hbar \epsilon_0 c^3} \left| \sum_a \int_0^L dt \mathbf{E}_{gs}^\pm \cdot \mathbf{v} e^{ik_{gs}^\pm \cdot \mathbf{r}_a(t)} e^{i\omega t} \right|^2. \quad (4)$$

Here L is the crystal length, and $+$ ($-$) denotes the crystal outside (inside). After integrating over single-electron trajectories $\mathbf{r}_a(t)$, the amplitudes of all electrons coherently sum up, showing the potential for a scaling with N_e^2 . We convert the sum over all electrons to integrals over the $n_e = N_e/K$ electrons within each microbunch $\sum_a \rightarrow \sum_l n_e \int \mathcal{D}(\mathbf{r}_0) \rho(\boldsymbol{\theta}_e) d^2 \boldsymbol{\theta}_e d^3 \mathbf{r}_0$. The electron distribution shape then decides if a superradiant N_e^2 scaling appears. Rather than directly evaluating the scattered x-rays, the problem is simplified by considering the inverse scattering process of incident x-rays impinging in the reverse direction onto the source-free crystal [29,47]. In this “reverse picture,” $\mathbf{E}_{gs}^\pm$ in Eq. (4) are the electric fields of the crystal diffracted wave inside the crystal and the specularly reflected diffracted wave outside the crystal, and their respective wavevectors $\mathbf{k}_{gs}^\pm$ are parallel to the crystal surface but antiparallel to the electron velocity in the grazing EAD geometry [29,42,43] (also see End Matter). To treat the general diffraction geometries considered here, we extend the approximate grazing EAD approach [42] to four-beam boundary interactions within x-ray dynamical diffraction theory [51–54] in a companion paper [47]. Since the Mössbauer crystal serves as the source, the incident wave vector in the reverse picture is $\mathbf{k}_0 = -\mathbf{k}'_0$, and the diffraction is quantified by the crystal susceptibility $\chi_g^{(s)}(\omega) = \chi_{gn}^{(s)}(\omega) + \chi_{ge}^{(s)}$, comprising nuclear (n) and electronic (e) scatterings [47].

Crystal and electron beam parameters—We apply our approach to a ^{57}Fe α -iron crystal of length $L = 1$ cm with nuclear resonance energy 14.4 keV and natural linewidth 4.7 neV, including splitting due to the hyperfine magnetic field $B_{\text{hf}} \approx 33$ T, which results in six Zeeman transitions. In order to obtain x-ray scattering predominantly from the nuclear resonances [38,41], we choose the intense (011) reflection with the Bragg angle $\theta_B \approx 12.26^\circ$ [55], which has already been employed in Mössbauer spectroscopy of iron crystals [56–58]. We consider electron parameters based on the EuXFEL facility [50,59,60], with electron energy $E_e = 17.5$ GeV and bunch charge $Q = 1$ nC,

corresponding to $N_e = 6.25 \times 10^9$ electrons per bunch. The bunch length and emittance are $L_b = 23$ μm and $\epsilon = \sigma_a \sigma_t \approx 2 \times 10^{-11}$ m · rad, respectively [60]. Within the undulator, typically $\sigma_t = 20$ μm and $\sigma_a = 1$ μrad . Since the SPMR setup likely would be placed downstream from the undulators, we also consider the possibility to focus the electron beam [61,62] to a smaller transverse size $\sigma_t = 2$ μm with $\sigma_a = 10$ μrad . The quality of the microbunching affects the degree of superradiant enhancement. We chose $\sigma_x = 0.01$ nm in the calculations, for which superradiance can be achieved [48].

Geometric optimization—We now turn to the optimization of the remaining free parameters Θ_0 and a_0 . We first examine in Fig. 2(a) the dependence of the total number of SPMR photons N_{SPMR} (on and off resonance with the nuclei) generated per electron bunch (pulse) on the cutting angle Θ_0 and the transverse distance a_0 of the electron beam to the crystal surface. The dashed line at $a_0 = 0$ forms the boundary between vacuum and crystal. Strikingly, we find that N_{SPMR} can be enhanced

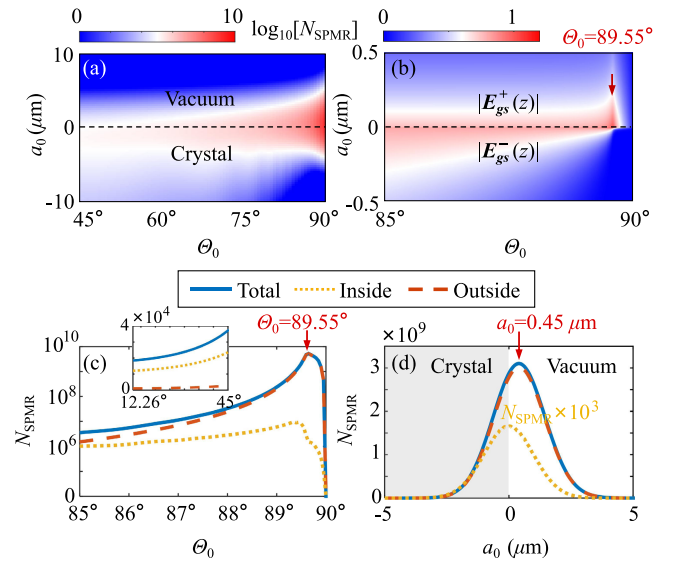


FIG. 2. (a) Contour plot of the total number of SPMR N_{SPMR} per electron bunch with logarithmic scale as a function of cutting angle of the crystal surface Θ_0 and electron distance from the crystal surface a_0 . (b) Diffracted field distribution $\mathbf{E}_{gs}^\pm$ normalized to the incident field amplitude $\mathbf{E}_{0s}$ in the reverse picture. (c) and (d) show the sections through (a) at $a_0 = 0$ and $\Theta_0 = 89.55^\circ$, respectively. Next to the total electric field (blue solid lines), also the individual contributions of crystal diffraction (inside field $\mathbf{E}_{gs}^-$, yellow dotted lines) and specular reflected diffraction (outside field $\mathbf{E}_{gs}^+$, red dashed lines) to N_{SPMR} are indicated. The inset in (c) shows the case of $\Theta_0^{\text{min}} = 12.26^\circ \leq \Theta_0 \leq 45^\circ$ for $a_0 = 0$. In (d), the contribution of the field $\mathbf{E}_{gs}^-$ is enlarged as $N_{\text{SPMR}} \times 10^3$. Results are given for the (011) reflection of the ^{57}Fe α -iron crystal with the electron beam size $\sigma_t = 2$ μm . In the chosen geometry, only the $s = \sigma$ -polarization perpendicular to the diffraction plane $\mathbf{v} \times \mathbf{g}$ contributes to the total photon number.

significantly by increasing Θ_0 , away from the grazing EAD geometry with $\Theta_0 = 45^\circ$, and by displacing the electron beam slightly above the crystal surface.

To understand this qualitatively different optimum geometry, we consider the spatial distribution of the diffracted fields $E_{gs}^\pm$ in Fig. 2(b). The crystal diffracted field E_{gs}^- dominates over a wide range of (Θ_0, a_0) values. However, its amplitude decreases rapidly as Θ_0 approaches 90° . This is due to the increase of the travel distance of the parametric beam through the crystal with Θ_0 . By contrast, the specularly reflected diffraction increases with Θ_0 . Eventually, it becomes the dominant radiation channel and overcomes the increased absorption inside the crystal. Subsequently, in the limit of $\Theta_0 \rightarrow 90^\circ$, both fields vanish. The amplitude of the specularly reflected diffracted wave E_{gs}^+ is further influenced by the total external reflection on the crystal, leading to a maximum at high Θ_0 . We denote this optimum geometry as the *grazing-specular diffraction* (SD) geometry. In this regime, the field E_{gs}^+ receives contributions from a larger lateral range of a_0 , and the spatial phase- and amplitude-distributions of $E_{gs}^\pm$ determine how the transverse electron-beam profile contributes to N_{SPMR} .

This interpretation of the grazing SD geometry is also reflected in the Θ_0 and a_0 dependencies of N_{SPMR} . Figures 2(c) and 2(d) show the contributions of the crystal diffraction and the specularly reflected diffraction to N_{SPMR} separately. In Fig. 2(c), at higher Θ_0 , N_{SPMR} is dominated by the specularly reflected diffraction, and is peaked at $\Theta_0 \approx 89.55^\circ$. In contrast, at lower cutting angles including the grazing EAD geometry, the crystal diffracted wave dominates [see the inset of Fig. 2(c)]. In Fig. 2(d), the dependence of N_{SPMR} on the transverse electron distance a_0 is shown for the optimum $\Theta_0 = 89.55^\circ$. As expected, the crystal diffraction contributes most at the crystal surface where the absorption is minimized. However, its peak value is lower than the contribution of the specularly reflected diffraction by 3 orders of magnitude (note the scaling of this curve in the figure). The maximum of $N_{\text{SPMR}} \approx 3 \times 10^9$ photons generated per bunch is located slightly outside of the crystal ($a_0 \approx 0.45 \mu\text{m}$), arising almost exclusively from the specularly reflected diffraction, emitted at a small exit angle of about 0.19° to the crystal surface. We note that the grazing-diffraction geometry was employed in an earlier study of PXR [54], however, without the global geometry optimization employed here and without comparison to the grazing EAD geometry, which was assumed to be optimal in the recent work on PMR [38], PXR [42], and SPXR [44,45]. The full geometry dependence of SPMR is discussed in detail in Ref. [47].

Mössbauer photon number—Next, we analyze the SPMR performance in the optimum grazing SD geometry. The emission spectrum $n_{\text{SPMR}}(\omega_x) = \partial N_{\text{SPMR}} / \partial \omega_x$ around the Mössbauer resonance, where $\omega_x = (\omega - \omega_0) / (\gamma_0 / 2)$ is a dimensionless frequency, is shown in Fig. 3.

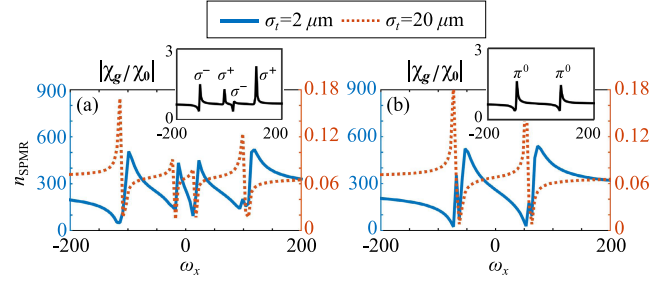


FIG. 3. SPMR emission spectra for different orientations of B_{hf} : (a) $B_{\text{hf}} \parallel (\mathbf{v} \times \mathbf{g})$ is perpendicular to the diffraction plane, and (b) $B_{\text{hf}} \perp (\mathbf{v} \times \mathbf{g})$ is parallel to the diffraction plane and perpendicular to the Mössbauer wave vector $\mathbf{k}_0$, $B_{\text{hf}} \perp \mathbf{k}_0$. Results are shown for different transverse electron beam diameters, keeping the emittance constant: $\sigma_t = 2 \mu\text{m}$ with the optimum geometry parameters $(\Theta_0, a_0) = (89.55^\circ, 0.45 \mu\text{m})$ (blue solid lines), and $\sigma_t = 20 \mu\text{m}$ with optimum geometry $(\Theta_0, a_0) = (89.92^\circ, 0.46 \mu\text{m})$ (red dotted lines). The insets show the ratio $|\chi_g(\omega)/\chi_0(\omega)|$. Results are for the (011) reflection of the ^{57}Fe α -iron crystal.

To demonstrate the superradiant enhancement, we compare SPMR with the results of PMR based on incoherent electrons delivered by the MAMI facility [38]. The relevant figures of merit are the total numbers of Mössbauer resonant photons generated per electron bunch (pulse) N_{pulse} , per second N_{sec} , and per electron $\bar{N} = N_{\text{pulse}}/N_e$. These values are calculated by summing over a bandwidth of one natural linewidth γ_0 within each hyperfine line. The corresponding photon number N_{γ_0} per pulse and γ_0 , averaged over the respective values of all hyperfine lines, relates to the spectral flux. Note that N_{sec} and N_{γ_0} are of relevance for experiments requiring a high average [18] or peak (see Refs. [11,17] and references therein) resonant flux, respectively. Results are given in Table I based on the bunch rates of 27000 Hz for EuXFEL [59] and 2.45 GHz for the incoherent 100 μA electron beam at MAMI [38,63]. For the narrower XFEL electron beam diameter ($\sigma_t = 2 \mu\text{m}$), a spectral flux of 935 SPMR photons/(pulse $\cdot \gamma_0$) is predicted, exceeding PMR by eleven orders of magnitude. N_{sec} exceeds the PMR value by 7 orders of magnitude, due to the $\sim 10^5$ times higher number of pulses per second at MAMI, while $\bar{N}$ is 4 orders of magnitude higher than in PMR due to the superradiant enhancement. Also at lower cutting angles $\Theta_0 = 45^\circ$, the SPMR source outperforms PMR in all the above quantities except $\bar{N}$, with N_{pulse} exceeding the PMR value by 6 orders of magnitude. The angle $\Theta_0 = 45^\circ$ represents the geometry considered for SPXR before, and the corresponding values also act as references for evaluating the enhancement gained by optimizing the geometry to $\Theta_0 = 89.55^\circ$. The SPMR generation is maximized in a range of distances from the crystal surface due to the finite transverse electron field extend. Therefore, like in regular PMR, the SPMR performance depends on the electron beam diameter σ_t .

TABLE I. Numbers of Mössbauer photons per bunch (pulse) (N_{pulse}), per second (N_{sec}), and per electron ($\bar{N}$) summed over one linewidth γ_0 of each hyperfine line. N_{γ_0} is the corresponding number per γ_0 averaged over the hyperfine lines, relating to the spectral flux. SPMR results are calculated for $\mathbf{B}_{\text{hf}} \parallel (\mathbf{v} \times \mathbf{g})$ with parameters based on the EuXFEL facility. The geometry (Θ_0, a_0) is optimized for $\sigma_t = 20 \mu\text{m}$ and $\sigma_t = 2 \mu\text{m}$, respectively. PMR results are based on Ref. [38] for a single linewidth around ω_0 , without hyperfine splitting, with parameters for the MAMI facility. All values correspond to the (011) reflection of the ^{57}Fe α -iron crystal.

	SPMR		PMR	
σ_t	20 μm	2 μm	2 μm	1.9 μm
Θ_0	89.92°	89.55°	45°	45°
a_0	0.46 μm	0.45 μm	0	0
N_{pulse}	0.92	3.74×10^3	1.15×10^{-2}	5.71×10^{-9}
N_{sec}	2.48×10^4	1.01×10^8	3.11×10^2	14
$\bar{N}$	1.47×10^{-10}	5.98×10^{-7}	1.84×10^{-12}	2.24×10^{-11}
N_{γ_0}	0.23	9.35×10^2	2.87×10^{-3}	5.71×10^{-9}

To illustrate this, N_{SPMR} is shown as a function of σ_t in End Matter Fig. 4(b). The value of N_{SPMR} increases with decreasing σ_t until most electrons are within the optimum distance range. Superradiant enhancement persists at larger beam sizes, which relaxes the requirements for a demonstration of SPMR. In Table I, we consider $\sigma_t = 20 \mu\text{m}$ representative for an electron beam within the XFEL undulator. Also for this larger beam size, N_{pulse} and N_{sec} exceed the PMR results by 8 and 3 orders of magnitude, respectively.

The narrow-band SPMR radiation is accompanied by a spectrally broad SPXR background, analogous to the background at established accelerator-based sources. Methods established at existing sources, such as high-resolution monochromatization, x-ray polarimetry, or time gating, could also be used to remove the SPXR background. High-speed shutters (see, e.g., Ref. [64]) could block the SPXR emitted on fs-ps timescales from the SPMR on ns time scales, thereby opening up the possibility of generating a background-free SPMR source. Alternatively, crystals with pure nuclear reflexes could suppress the electronic background [38].

Conclusion—We have investigated the superradiant parametric generation of Mössbauer radiation. Compared with conventional PMR, we found that SPMR can improve all relevant figures of merit by many orders of magnitude. This improvement is a qualitative step, since it for the first time renders SPMR competitive with the direct photon generation at accelerator-based x-ray sources [24]. For example, 935 SPMR photons/(pulse $\cdot \gamma_0$) represent a spectral flux of 2.0×10^{11} photons/(pulse $\cdot \text{eV}$), which corresponds to a self-seeded XFEL pulse with a bandwidth $\hbar\Delta\omega = 1 \text{ eV}$ and a pulse energy $E_{\text{pulse}} = \hbar\omega_0 N_{\gamma_0} \Delta\omega/\gamma_0 \approx 460 \mu\text{J}$. Note that

this value does not include the additional energy of a broader SASE fraction. Unlike XFEL radiation and SPXR, the SPMR source can be monochromatized using time gating to the nuclear-resonant contribution, without the need for crystal-based monochromators. This way, detrimental effects of the intense off-resonant background on sample or instrumentation can be avoided. Interestingly, our results cannot only be attributed to the source advancement from synchrotron to XFEL. Rather, the grazing EAD geometry—previously regarded as optimal for PXR and PMR—is not optimum for SPMR. Instead, the grazing SD geometry maximizes the performance at higher crystal cutting angles by global optimization of SPMR geometry. This geometry is also favorable, since it involves electron beams passing above the crystal surface, rather than through the crystal, which likely reduces heat load effects and crystal damage.

Since the crystal is only of centimeter scale, we envision a compact laboratory for applications harnessing the microbunched electron beam without the need for new accelerator or undulator tunnels. This facility could be operated in parallel to the regular XFEL operation, and could also be used for other applications. Note that microbunch-preserving electron beam deflections have been studied before [65–67].

The higher-order SPMR harmonics could be used to excite Mössbauer isotopes with higher transition energies which cannot be reached directly by the current XFELs. For the future, next to a parasitic operation exploiting the otherwise dumped XFEL electron beam, we can also envision a parallel delivery of the regular XFEL and the SPMR beams into the experimental hutch. This could enable novel coherent pump-probe spectroscopy techniques [13] with variable delays on the nanosecond time scale of relevance for Mössbauer science, which are otherwise challenging to implement at XFELs.

Data availability—The data that support the findings of this article are openly available at [68].

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End Matter

Reverse picture—The reverse picture for determining the optimal geometry of the SPMR source is sketched in Fig. 4(a). The relevant wave vectors constructing the Bragg vector triangles in the real picture $\mathbf{k}_v + \mathbf{g} = \mathbf{k}_0$ and in the reverse picture $\mathbf{k}'_0 + \mathbf{g} = \mathbf{k}_g$ are depicted by the solid arrows and dashed arrows, respectively. The specific form of $\mathbf{k}_g$ in the reverse picture depends on the transverse position of electrons, i.e., $\mathbf{k}_g = \mathbf{k}_g^+$ for $z > 0$ and $\mathbf{k}_g = \mathbf{k}_g^-$ for $z < 0$.

SPMR spectra—For the SPMR spectra in Fig. 3, the geometric parameters in the grazing SD geometry are optimized as $(\Theta_0, a_0) = (89.55^\circ, 0.45 \mu\text{m})$ for a smaller electron beam rms diameter $\sigma_t = 2 \mu\text{m}$ and $(\Theta_0, a_0) = (89.92^\circ, 0.46 \mu\text{m})$ for $\sigma_t = 20 \mu\text{m}$. In the two considered magnetic field configurations, the hyperfine transitions of only circular ($\sigma^\pm$) polarization and only linear (π^0) polarization couple to the x-rays respectively. This can be seen from Fig. 3, which shows that the SPMR spectral profile results from the ratio $|\chi_g(\omega)/\chi_0(\omega)|$ which is nonzero at the involved nuclear resonances. We note that the spectral profiles are modified by the interference between the electronic and the nuclear diffractions. This Fano-like interference [69] leads to asymmetric line profiles depending on the cutting angle and thus the diffraction geometry, and was also found for nuclei in x-ray waveguides [70]. Moreover, the spectral profiles exhibit substantial line broadenings and resonance shifts, which arise from nuclear dynamical diffraction effects. These are stronger in the grazing SD geometry than in the previously considered grazing EAD geometry (see the companion paper [47]).

Dependence of the SPMR photon generation on the transverse size of electron beam—As discussed in the main text, the SPMR photon generation depends sensitively on the transverse size of electron beam, since the range of distances from the crystal surface with optimum photon yield is limited. To illustrate this, Fig. 4(b) shows the number of photons N_{SPMR} as a function of the beam size σ_t . It can be seen that the total field is dominated by the outside specularly reflected diffracted field over the entire range of σ_t in the grazing SD geometry.

Pulse energy of total SPMR photons—The standard figures of merit for x-ray sources also include the total number of emitted photons per pulse and the corresponding pulse energy. For the proposed SPMR source, the total number of photons N_{total} (N_{SPMR}) contains the contributions from both the nonresonant SPXR part and the near-resonant SPMR nuclear part. With the number of electron microbunches $K = L_b/d_0 \approx 2.67 \times 10^5$, the SPXR linewidth is $\hbar\Delta\omega = \hbar\omega_0/K \approx 53.9 \text{ meV}$, corresponding to a total pulse energy of $E_{\text{pulse}} \approx 6.9 \mu\text{J}$ for $N_{\text{total}} \approx 3 \times 10^9$ photons per pulse. This SPXR background linewidth, being below 100 meV, is narrower than the state-of-the-art undulator x-ray self-seeding spectral linewidths (typically on the order of 1 eV for example in Ref. [24]) and yields an average photon spectral flux of $\Phi \approx 5.6 \times 10^{10}$ photons/(eV · pulse). For comparison, the XFELs in Refs. [25,26] achieve pulse energies of 2 – 20 μJ , however, in an even more narrow spectral bandwidth of about 2 – 3 meV.

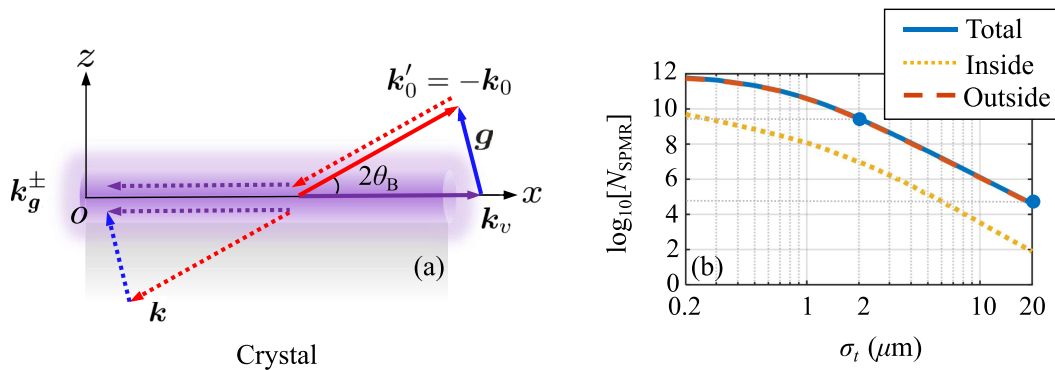


FIG. 4. (a) Real and reverse pictures, sketched using the relevant wave vectors indicated by the solid and dashed arrows, respectively. In the reverse picture, a plane x-ray with the reversed Mössbauer wave vector $\mathbf{k}'_0 = -\mathbf{k}_0$ is incident on the source-free crystal. The wave vectors of the crystal-diffracted wave $\mathbf{k}_g^-$ and the specularly reflected diffracted wave $\mathbf{k}_g^+$ are antiparallel to the velocity of the electron beam (cylinder) in the grazing-incidence geometry. (b) Total number of SPMR photons generated per bunch (blue solid line, logarithmic scale) as a function of the transverse size of the electron beam σ_t . The individual contributions from the crystal-diffracted wave $\mathbf{E}_{gs}^-$ (yellow dotted line) and the specularly reflected diffracted wave $\mathbf{E}_{gs}^+$ (red dashed line) are indicated separately, with the cases $\sigma_t = 20 \mu\text{m}$ and $\sigma_t = 2 \mu\text{m}$ highlighted. Results correspond to the (011) reflection of the ^{57}Fe α -iron crystal at $\Theta_0 = 89.55^\circ$ and $a_0 = 0$.