

Parametric Mössbauer radiation in the grazing-diffraction geometry: Specular diffraction and superradiant amplification

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Parametric x-ray radiation (PXR) is a tunable and versatile x-ray production mechanism that relies on relativistic electrons traversing through a crystal. When the crystal contains Mössbauer nuclei, they may also contribute to the x-ray generation, giving rise to spectrally narrow parametric Mössbauer radiation (PMR) as a promising alternative to undulator-based x-ray sources. It is well known that the x-ray generation strongly depends on the diffraction geometry. Motivated by this, here, we develop a framework for parametric x-ray generation in general diffraction geometries, encompassing the entire x-ray emission range from the conventional grazing extremely asymmetric diffraction (EAD) geometry with a large emission direction to the crystal surface up to the grazing specular diffraction (SD) geometry with emission near parallel to the surface. The framework further incorporates a range of physical effects arising from more realistic modeling of both the Mössbauer crystal and the electron beam. We show that the PMR generation in the grazing SD geometry qualitatively differs from the previously considered grazing EAD case, and explain the physical origins of these differences. We further apply our framework to analyze the geometry dependence of superradiant parametric Mössbauer radiation (SPMR) generated from microbunched electron beams provided by x-ray free-electron lasers. Our results set the stage for the exploration of PXR, PMR, superradiant amplification of parametric x-ray radiation, and SPMR beyond the established diffraction geometries.

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I. INTRODUCTION

So far, Mössbauer science is largely confined to the low-excitation regime, as third-generation synchrotron sources typically deliver far less than one near-resonant x-ray photon per pulse within the extremely narrow nuclear linewidths of Mössbauer isotopes [1–3]. This limitation severely constrains potential applications of x-ray optics with Mössbauer nuclei, such as correlation measurements, multi-excitation schemes, and nonlinear optical effects [4–7]. The situation has changed with the advent of x-ray free-electron lasers (XFELs) [8–10], which are capable of delivering significantly more than one resonant photon per pulse within the linewidth of the archetype ^{57}Fe Mössbauer transition, thereby opening new possibilities for Mössbauer spectroscopy. Enabled by this, at the Spring-8 Angstrom Compact Free Electron Laser (SACLA), resonant diffraction of x-rays from ^{57}Fe nuclei was observed for the first time with up to 68 signal photons per x-ray excitation, allowing for the study of superradiance from multiply-excited nuclear ensembles [11]. A substantial step forward was the commissioning of self-seeding [12–15].

At the European XFEL (EuXFEL), coherent nuclear forward scattering of self-seeded x-rays was recently observed with up to 900 signal photons per x-ray pulse scattered from ^{57}Fe and used to analyze Mössbauer spectra on the level of single x-ray excitations [16]. In parallel, the high average resonant flux at EuXFEL has enabled the resonant x-ray excitation of the ultranarrow nuclear clock transition in ^{45}Sc with a linewidth on the eV scale [17]. Nevertheless, further source progress is desirable, to render isotopes with higher nuclear transition energies accessible, or to allow for excitation of nuclei beyond the linear excitation regime [7,18,19]. A promising candidate is the x-ray free-electron laser oscillator (XFEL-O) or related concepts of cavity-based x-ray sources [20–22].

An alternative yet comparatively less explored approach for exciting Mössbauer isotopes is parametric x-ray radiation (PXR) [23], which is generated when ultrarelativistic charged particles traverse a crystal, and their virtual-photon fields undergo coherent diffraction from the crystallographic lattice planes. Over the past years, PXR has been demonstrated not only as a compact, tunable x-ray source [24]—a laboratory-scale alternative to synchrotron radiation—but also as a tool for electron-beam diagnostics in accelerator physics [25,26] and for potential applications in medicine [27]. In addition, PXR has stimulated extensive studies of related radiation mechanisms, including diffraction transition radiation from relativistic particles interacting with artificial superlattices [28] and quasi-Cherenkov parametric radiation from photonic crystals [29]. These developments are expected to further promote the integration of accelerator physics and materials science.

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In parallel, the potential of PXR for driving Mössbauer nuclear resonances—yielding parametric Mössbauer radiation (PMR)—has been explored [30–33]. For instance, PMR may render isotopes accessible that have a higher transition energy beyond the reach of state-of-the-art XFEL sources [30], highlighting its promising potential for applications in Mössbauer spectroscopy. Unfortunately, however, the previous studies concluded that the nuclear-resonant photon flux delivered by PMR is far below that of corresponding undulator-based radiation sources.

Enhancing the PXR intensity has traditionally relied on engineering optimized diffraction geometries, with the grazing extremely asymmetric diffraction (EAD) geometry being particularly effective due to the long coherence length along the electron trajectory [23,34,35]. In the grazing EAD geometry, electrons propagate parallel and close to the crystal surface, i.e., the grazing-incidence geometry, and the emitted PXR exits the crystal at a large angle nearly perpendicular to the crystal surface. This configuration minimizes the PXR absorption inside the crystal while simultaneously increasing the effective coherent interaction length between the electrons and the crystal, thereby substantially enhancing the overall PXR intensity.

In addition to optimizing diffraction geometry, superradiant amplification of parametric x-ray radiation (SPXR) was recently proposed [36] based on spatially structured electron bunches that exit from the XFEL undulator. The XFEL self-amplified spontaneous-emission (SASE) process periodically modulates the electron density to microbunches such that the virtual-photon fields generated by individual electrons in the crystal in each microbunch interfere constructively. This coherent amplification improves the PXR intensity by several orders of magnitude, reaching photon yields comparable to those of XFEL pulses generated by the undulator radiation. Building up on this mechanism, related proposals have explored the realization of SPXR resonators [37] and PXR-based XFEL ring cavities [38]. Notably, the schemes in Refs. [36,37] also exploited the advantages of the grazing EAD geometry [23,34,35].

However, the grazing EAD geometry imposes intrinsic constraints on the PMR generation at a specific Mössbauer resonance frequency. In particular, maintaining the advantage of an extended interaction length via the grazing-incidence geometry of the electron beam no longer guarantees that the emitted PMR still exits the Mössbauer crystal at a large angle with respect to the crystal surface, and it may even exit at a small angle. Consequently, the grazing EAD geometry generally becomes less effective for PMR generation. This raises the question of whether there are more favorable diffraction geometries for PMR.

Motivated by this, we develop herein a framework for both PXR and PMR in general diffraction geometries (see Fig. 1). We find that the established approach for modeling the grazing EAD geometry [Fig. 1(b)] remains valid over a large range of scattering geometries. However, if the emission direction of the parametric radiation approaches the crystal surface [Fig. 1(c)], a qualitatively different regime emerges, which requires a revision of the approach and an extension of the theory to four-wave interactions at the crystal boundary. In a related companion paper [39], we introduce a

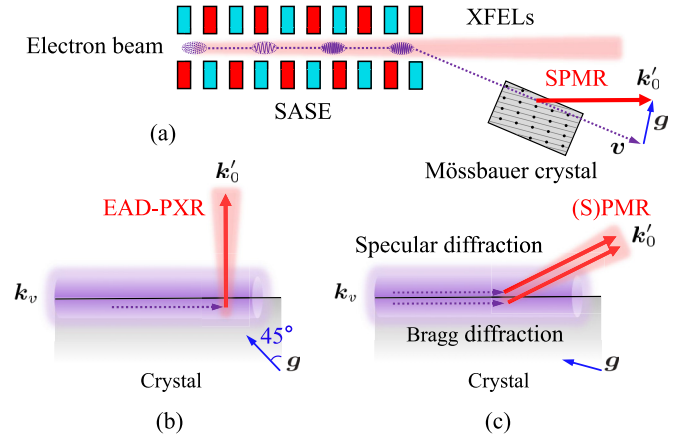


FIG. 1. (a) Schematic of SPMR generation. In the undulator, the self-amplified spontaneous emission (SASE) process modulates an electron pulse into a train of coherent microbunches. The resulting prebunched electron beam emerging from the undulator with the velocity v then propagates parallel to the Mössbauer crystal-vacuum interface in the grazing-incidence geometry and generates an SPMR pulse along the Bragg direction defined by the crystal reciprocal-lattice vector g . (b) Grazing EAD geometry used to enhance PXR intensity. The electron beam, with the virtual-photon wave vector k_v (purple dashed arrow), propagates in the grazing-incidence geometry, while the emitted PXR photons (red solid arrow) exit the crystal surface nearly normal to the crystal surface. (c) Grazing SD geometry considered in this work for (S)PMR. Virtual photons associated with the electrons distributed outside the crystal are reflected by the specular diffraction at the crystal surface, whereas those associated with the electrons inside the crystal undergo the regular Bragg diffraction, as in the conventional PXR.

superradiant parametric Mössbauer radiation (SPMR) source, as sketched in Fig. 1(a), and employ the framework developed here to optimize the scattering geometry in such a way that the nuclear-resonant SPMR yield is maximized. Interestingly, in the companion paper, we find that the SPMR photon number is maximized for emission close to the crystal surface, rather than in the established grazing EAD geometry. Therefore, here we provide an in-depth study of this grazing-specular diffraction (SD) geometry [see Fig. 1(c)], and detail the derivation of SPMR. In particular, we discuss the effects of the transverse electron-beam profile and the role of attenuation within and outside the Mössbauer crystal, study the competition between the radiation generated by electrons inside and outside the crystal, and elucidate the underlying physics that makes the grazing SD geometry more favorable for enhancing the SPMR intensity than the conventional grazing EAD geometry. Moreover, we show that the XFEL-specific parameters place PMR in a new parameter regime that differs substantially from the previously studied synchrotron-based PMR scenarios in Ref. [30]. Finally, we also include nuclear hyperfine splitting. As a whole, these extensions establish a comprehensive theoretical framework for (S)PXR and (S)PMR, and set the stage for an experimental exploration of the method at state-of-the-art accelerator-based electron sources.

We note that the grazing-diffraction geometry has been considered for PXR before [40], motivated by the

observation that it may reduce the likelihood of crystal damage and better preserve the quality of the electron beam. However, the comparison with other geometries, including the grazing EAD geometry that was employed for PXR, PMR, and SPXR in recent studies [30,34,36], in terms of the x-ray generation performance is an open question. In the following, we resolve this question by optimizing the PXR mechanisms over the geometry.

This paper is organized as follows: Section II introduces the mechanism of superradiant radiation generated by a microbunched electron beam moving in a medium and illustrates how it leads to the coherent amplification of PMR. In Sec. III, we describe the (S)PMR source, including the diffraction geometry and the Mössbauer crystal. In Sec. IV, we solve the exact four-beam electric fields of the (S)PMR problem within the theoretical framework of two-wave dynamical diffraction theory and discuss the conditions under which the solution reduces to the grazing EAD case. In Sec. V, we specialize the general (S)PMR description to the SPMR case and present the full derivation of the angular-spectral distribution of SPMR photons under the superradiant Mössbauer resonance condition. Section VI is devoted to analyzing in detail the properties of the diffracted waves involved in SPMR generation and reveals the physical mechanism responsible for SPMR enhancement in the grazing SD geometry, which is fundamentally different from those in the grazing EAD case. Section VII discusses the dependence of the generation of nuclear-resonant photons on the relevant system parameters. Also, the angular distribution and nuclear dynamical diffraction effects are discussed briefly. Finally, in Sec. VIII, we summarize our results.

II. (SUPERRADIANT) PARAMETRIC RADIATION

The starting point of our theoretical analysis is the angular-spectral distribution of the photons emitted by a charged particle moving inside a medium. The rigorous expression for this quantity can be derived using the electromagnetic Green's tensor in combination with the optical reciprocity theorem [23]. The elegance of this approach lies in the transformation of the problem of a radiating charged source in a medium into a source-free scattering problem of electromagnetic waves in the same medium under identical boundary conditions. The general expression for the angular spectral distribution of photons with s polarization emitted by an ensemble of electrons interacting with a medium can be written as [23,36]

$$\frac{\partial^2 N_{\mathbf{n},\omega s}}{\partial \omega \partial \Omega} = \frac{e_0^2 \omega}{16\pi^3 \hbar \epsilon_0 c^3} \sum_{a,b}^{N_e} \int E_{ks}^{(-)*}(\mathbf{r}_a(t), \omega) \cdot \mathbf{v}_a e^{i\omega t} dt \times \int E_{ks}^{(-)}(\mathbf{r}_b(t'), \omega) \cdot \mathbf{v}_b e^{-i\omega t'} dt', \quad (1)$$

where $\mathbf{k} = \omega \mathbf{n}/c$ is the wave vector of the emitted photons along the far-field observation direction $\mathbf{n} = \mathbf{r}/r$ within the solid angle $d\Omega$. Here, N_e denotes the number of electrons, and $\mathbf{r}_a(t) = \mathbf{r}_0^{(a)} + \mathbf{v}_a t$ is the trajectory of electron a with velocity $\mathbf{v}_a$. Note that the electric field $E_{ks}^{(-)}$ is the solution of the source-free scattering problem in terms of a plane wave

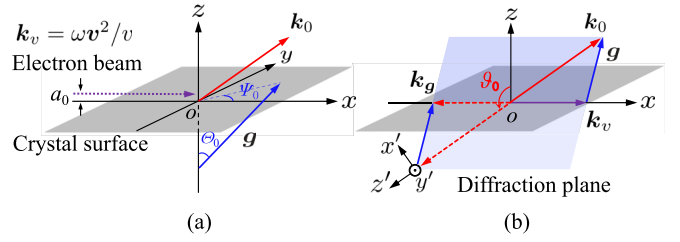


FIG. 2. Two pictures of (S)PMR. (a) Real picture, involving light emission along the direction $\mathbf{k}_0$ due to the diffraction of the electron virtual-photon fields on the crystal. The crystal cutting angle θ_0 is measured between the reciprocal-lattice vector $\mathbf{g}$ and the z axis, and the crystal rotation angle ψ_0 in the x - y plane is determined by the (S)PMR Bragg condition. (b) Reverse picture, obtained from (a) via the optical reciprocity theorem. In this picture, a plane wave in the reverse direction $-\mathbf{k}_0$ is incident on the source-free crystal and diffracted into a direction nearly antiparallel to the electron velocity in the real picture. The diffraction plane is determined by the vectors $(\mathbf{v}, \mathbf{g})$. In the coordinate system (x', y', z') for the reverse picture, the detection plane (x', y') is perpendicular to $\mathbf{k}_0$, and the y' axis is perpendicular to the diffraction plane.

and an asymptotically ingoing spherical wave. By the optical reciprocity theorem, it is related to the solution $E_{-ks}^{(+)}$ of the regular source-free scattering problem on the same crystal, however, with an incident plane wave with wave vector $-\mathbf{k}$ [41]. This alternative view, referred to as the reverse picture (see Fig. 2), allows for the well-studied corresponding source-free electromagnetic scattering problem to serve as a convenient starting point for evaluating the radiation fields in the original real picture. We note that if the radiation involves nuclear transitions, for instance, when the medium contains Mössbauer nuclei, the optical reciprocity theorem needs to be generalized to account for nuclear electromagnetic multipoles. The corresponding generalization is given in Appendix C for the case of a Mössbauer crystal (also see Refs. [42,43]).

From the sums in Eq. (1) on the level of field amplitudes, it is immediately apparent that the possibility of interference between the x-rays emitted by different electrons a, b is considered. For an electron pulse structured into different microbunches, a continuous distribution of electrons can be assumed within each microbunch. Therefore, the summation over the electrons in Eq. (1) may be converted into an integral, i.e.,

$$\sum_a^{N_e} \rightarrow \sum_{l=0}^K n_e \iiint \rho(\theta_e) \mathcal{D}_l(\mathbf{r}_l) \mathcal{D}_x(x_0^{(l)}) d^2\theta_e d^2\mathbf{r}_l dx_0^{(l)}. \quad (2)$$

Here, K is the number of microbunches in a single electron bunch and thus $n_e = N_e/K$ is the number of electrons distributed within each microbunch. Furthermore, $\mathbf{r}_l$ is the transverse position of the electrons and $x_0^{(l)} = x_0 - ld_0$ is the longitudinal coordinate of the electrons in the l th microbunch along the x axis with d_0 being the distance between two adjacent microbunches. We assume that the electrons in all microbunches follow the same distributions of angular spread and transverse position, i.e., $\rho(\theta_e^{(l)}) = \rho(\theta_e)$ and $\mathcal{D}_l(\mathbf{r}_l^{(l)}) = \mathcal{D}_l(\mathbf{r}_l)$, and the initial longitudinal relative positions of the

electrons within the corresponding microbunches are also described by the same distribution. These XFEL electron-beam distributions can be modeled using Gaussian functions [36–38,44],

$$\begin{aligned}\rho(\theta_e) &= \frac{1}{\pi\sigma_a^2} e^{-\theta_e^2/\sigma_a^2}, \\ \mathcal{D}_t(\mathbf{r}_t) &= \frac{1}{\pi\sigma_t^2} e^{-r_t^2/\sigma_t^2}, \\ \mathcal{D}_x(x_0^{(l)}) &= \frac{1}{\sqrt{\pi}\sigma_x} e^{-(x_0^{(l)} - l d_0)^2/\sigma_x^2},\end{aligned}\quad (3)$$

where σ_a is the rms angular spread, and σ_t and σ_x are the rms transverse size of the electron bunch and the fluctuation of the modulation period, respectively.

We note that superradiance appears in the x-ray generation if an electron bunch is small as compared with the radiation wavelength, similar to the condition of Dicke superradiance [45]. If instead the electrons are structured in multiple independent microbunches in such a way that each individual microbunch is small compared with the radiation wavelength λ_0 , i.e., $|\mathbf{r}_t|$ and $|x_0^{(l)} - l d_0| \ll \lambda_0$, and the mutual distance d_0 between these microbunches corresponds to the radiation wavelength, then all electrons are still in equivalent positions with respect to a single emission mode. As a result, the generated radiation scales with the number of electrons squared, since the radiation emitted from each individual microbunch is still enhanced by superradiance [44]. This mechanism of superradiance also applies to the virtual-photon fields of microbunched electron beams and hence to PXR. If the condition $k_v d_0 = 2\pi n$ ($n \in \{1, 2, \dots\}$) with the virtual-photon wave number $k_v = \omega/v$ of the electrons is fulfilled, the virtual-photon fields generated from K microbunches ($K \gg 1$) exhibit constructive interference. Then the summation for the field amplitudes in Eq. (1) leads to x-ray radiation proportional to the total number of electrons within a single bunch $(n_e K)^2 = N_e^2$.

In contrast with this superradiant case, for an electron bunch extended along the longitudinal direction without suitable microbunching, the cross-terms between different electrons in Eq. (1) destructively interfere, and a linear scaling of the emitted radiation with N_e is obtained. This is the previously considered regime of regular PMR [30–32].

In the following, we continue with the derivation of a unified theoretical framework for PMR and SPMR, which we indicate by the label (S)PMR. In particular, it includes a hyperfine splitting of the nuclei, and extends the PMR dynamical diffraction approach to a four-beam scenario valid for generalized scattering geometries. Afterward, we focus the discussion on the SPMR case and derive the qualitative properties of the generated SPMR x-rays in general scattering geometries. In a related companion paper [39], we use this theoretical framework to optimize the SPMR generation and show that the resulting SPMR source is superior to previous PMR sources by many orders of magnitude in different figures of merit, up to the point where SPMR becomes competitive with state-of-the-art self-seeded XFEL sources.

III. (S)PMR SOURCE

A. Diffraction geometry

In Refs. [23,34,35], the generation of PXR in the grazing EAD geometry was discussed. It was shown that strong absorption of PXR within a crystal can effectively be mitigated if the incident electron beam propagates parallel to and close to the crystal surface, i.e., the grazing-incidence geometry of the electron beam, and the orientation of the crystallographic planes is chosen properly such that the emitted PXR beam exits the crystal nearly perpendicular to the crystal surface. However, this geometry is applicable only when the emission frequency is unrestricted. In contrast, for (S)PMR, the stringent requirement that the emitted photon frequency must coincide with the nuclear resonance frequency makes it difficult to realize such a diffraction geometry where the radiation emerges normal to the crystal surface under the grazing-incidence condition for the electron beam [30]. Nevertheless, in the following, we still adopt the same grazing-incidence configuration for the electron beam as in the grazing EAD geometry, but investigate how (S)PMR varies with the scattering geometry, considering arbitrary emission directions.

To describe the geometric configuration of (S)PMR, we first consider the ideal case without any angular spreads of either the electrons or the parametric x-ray beam. As sketched in Fig. 2(a), the Mössbauer crystal has a surface in the x - y plane and occupies the region of negative z . In the real picture, the incident electron beam travels parallel to and close to the crystal surface with a central velocity $\mathbf{v} = v\mathbf{e}_x$ and at a transverse distance a_0 from the crystal surface. Parametric photons are emitted along the direction $\mathbf{k}_0$ at the Mössbauer resonance frequency $\omega_0 = ck_0$. Since the radiation problem can be reformulated as a source-free x-ray-scattering problem in the reverse picture by introducing the corresponding incident wave, we parametrize the incident Mössbauer wave vector in terms of the spherical angles as $\mathbf{k}'_0 = -\mathbf{k}_0 = k_0(\sin\vartheta_0 \cos\varphi_0, \sin\vartheta_0 \sin\varphi_0, \cos\vartheta_0) = k_0\mathbf{n}_0$, which in general is not perpendicular to the crystal surface. For clarity in the reverse picture, we redefine the notations of wave vectors such that $\mathbf{k}$ ($\mathbf{k}_0$) denotes the incident (Mössbauer) wave vector in the reverse picture, and $\mathbf{k}_g = \mathbf{k} + \mathbf{g}$ denotes the diffracted waves. Therefore $\vartheta_0 - \pi/2$ represents the actual emission angle of the generated (S)PMR with respect to the crystal surface. According to the virtual-photon concept, PXR is generated when the virtual-photon fields of a relativistic electron are diffracted by the crystallographic lattice planes (periodically arranged atoms or nuclei) of the crystal [23]. The generation of PXR occurs only for relativistic charged particles, since only in this regime is it possible for the wave vectors of the virtual-photon beam $\mathbf{k}_v = (\omega/v^2)\mathbf{v}$ and the emitted parametric beam $\mathbf{k} = (\omega/c)\mathbf{n}$ to satisfy the Bragg resonance condition $\mathbf{k}_v + \mathbf{g} = \mathbf{k}$, where $\mathbf{g}$ is the crystallographic lattice vector. After transforming to the reverse picture for the source-free-scattering problem, the PXR Bragg equation at the Mössbauer frequency ω_0 in the grazing-incidence geometry of the electron beam becomes

$$\frac{\omega_0}{c}\mathbf{n}_0 + \mathbf{g} = -\frac{\omega_0}{v^2}\mathbf{v}, \quad (4)$$

which means that the roles of the incident virtual-photon wave vectors and the diffracted parametric wave vectors are interchanged as a consequence of the optical reciprocity theorem. If we introduce the spherical angles Θ_0 and Ψ_0 for the crystallographic lattice vector $\mathbf{g}$, i.e., $\mathbf{g} = |\mathbf{g}|(\sin \Theta_0 \cos \Psi_0, \sin \Theta_0 \sin \Psi_0, \cos \Theta_0)$, as sketched in Fig. 2(a), the angles ϑ_0 and φ_0 corresponding to the central propagation direction of the parametric beam are determined by

$$\begin{aligned} \cos \vartheta_0 &= -\frac{g}{k_0} \cos \Theta_0, \\ \cos \varphi_0 &= -\frac{2k_0^2 - g^2}{2k_0 \sqrt{k_0^2 - g^2 \cos^2 \Theta_0}}, \end{aligned} \quad (5)$$

with the constraint condition for the orientation of $\mathbf{g}$

$$\sin \Theta_0 \cos \Psi_0 = -\frac{g}{2k_0}. \quad (6)$$

Once the crystal cutting angle Θ_0 is chosen, the angle Ψ_0 can be adjusted by rotating the crystal around the z axis until the (S)PMR Bragg condition is satisfied for the Mössbauer frequency. The two angles Θ_0 and Ψ_0 are relevant to the optimization of the geometric configuration of (S)PMR.

Next, we consider the nonideal single-electron case, for which a deviation of the electron velocity from the central velocity has to be considered. Due to the cone structure of the virtual-photon beam characterized by an angular width of the order of γ^{-1} [23], the parametric beam is also concentrated around the ideal propagation direction with a fine angular structure. Therefore, a deviation of the parametric photons also has to be considered. We parametrize the two deviations via

$$\begin{aligned} \mathbf{v} &\approx v \left[\left(1 - \frac{\theta_e^2}{2} \right) \mathbf{e}_x + \boldsymbol{\theta}_e \right], \quad \mathbf{e}_x \cdot \boldsymbol{\theta}_e = 0, \\ \mathbf{k} &\approx k \left[\left(1 - \frac{\theta^2}{2} \right) \mathbf{e}_z' + \boldsymbol{\theta} \right], \quad \mathbf{e}_z' \cdot \boldsymbol{\theta} = 0, \end{aligned} \quad (7)$$

where the electron angular spread $\boldsymbol{\theta}_e = \theta_{ey} \mathbf{e}_y + \theta_{ez} \mathbf{e}_z$ and photon angular spread $\boldsymbol{\theta} = \theta_x \mathbf{e}_x' + \theta_y \mathbf{e}_y'$ with $\theta_e = |\boldsymbol{\theta}_e|$ and $\theta = |\boldsymbol{\theta}|$. Here, a rotated coordinate system for the detection in the reverse picture is introduced as

$$\begin{aligned} \mathbf{e}_z' &= \frac{\mathbf{k}_0}{k_0}, \\ \mathbf{e}_y' &= \frac{\mathbf{k}_0 \times \mathbf{g}}{|\mathbf{k}_0 \times \mathbf{g}|}, \\ \mathbf{e}_x' &= \mathbf{e}_z' \times \mathbf{e}_y'. \end{aligned} \quad (8)$$

As sketched in Fig. 2(b), the y' axis is chosen perpendicular to the diffraction plane defined by the vectors $\mathbf{v}_0$ and $\mathbf{g}$.

B. Crystal susceptibility

1. Hyperfine splitting

For the (S)PMR source considered here, we focus on the most commonly used archetype Mössbauer isotope ^{57}Fe in the α -iron crystal. This isotope features a two-level transition from the ground state of the nuclear spin $I_g = 1/2$ to the first-excited state of $I_e = 3/2$, separated by the resonance

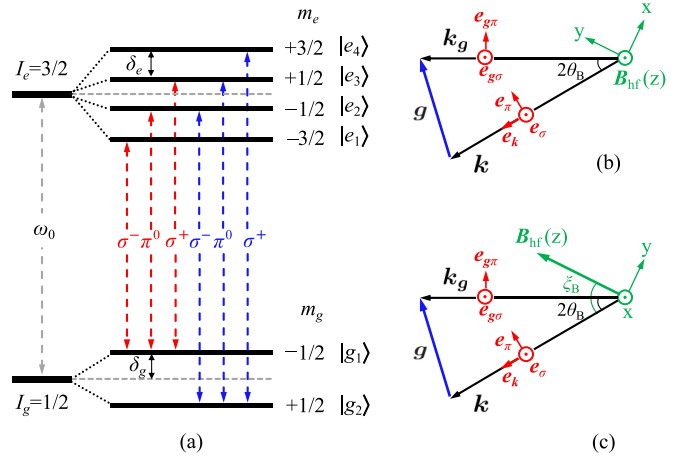


FIG. 3. (a) Energy-level scheme of ^{57}Fe with a magnetic hyperfine splitting. Due to the presence of the hyperfine magnetic field $\mathbf{B}_{\text{hf}}$, the two nuclear energy levels with spins $I_e = 3/2$ and $I_g = 1/2$ split up, and six $M1$ -transitions between the Zeeman magnetic sublevels are possible according to their nuclear-spin projections m_e and m_g on the quantization axis. Linear polarization is denoted by π^0 , right- and left-circular polarizations are denoted by σ^+ and σ^- , respectively. (b), (c) Two different orientations of the hyperfine magnetic field with respect to the diffraction plane. (b) The magnetic field is perpendicular to the diffraction plane, i.e., $\mathbf{B}_{\text{hf}} \parallel (\mathbf{k} \times \mathbf{g})$. (c) The magnetic field is parallel to the diffraction plane, i.e., $\mathbf{B}_{\text{hf}} \perp (\mathbf{k} \times \mathbf{g})$. The vectors $(\mathbf{e}_\sigma, \mathbf{e}_\pi)$ and $(\mathbf{e}_{g\sigma}, \mathbf{e}_{g\pi})$ are the unit polarization vectors of the incident plane wave $\mathbf{k}$ and the diffracted wave $\mathbf{k}_g$, respectively. The x , y , and z axes in panels (b) and (c) construct the quantization-axis Cartesian coordinate system. The quantization axis [the z axis in panels (b) and (c)] is along the direction of $\mathbf{B}_{\text{hf}}$. In panel (c), the orientation of $\mathbf{B}_{\text{hf}}$ is parametrized by the angle ξ_B between $\mathbf{B}_{\text{hf}}$ and $\mathbf{k}$.

frequency $\hbar\omega_0 = 14.4$ keV with the total natural linewidth $\hbar\gamma_0 = 4.66$ neV. However, in order to accurately describe the α -iron crystal, the PMR theory has to be generalized to include nuclear hyperfine splitting. This is due to the fact that the α -iron is a ferromagnetic crystal in a very broad temperature range. A relatively weak external hyperfine magnetic field can align a strong internal magnetization of $B_{\text{hf}} \approx 33$ T in an α -iron crystal. This results in six well-resolved nuclear hyperfine $M1$ (magnetic dipole) transitions between the ground and excited states. The transition-energy differences between two adjacent ground and excited sublevels are $\delta_g = 39.7\gamma_0$ and $\delta_e = 22.4\gamma_0$, respectively, as illustrated in Fig. 3(a). As shown below, this leads to a pronounced and versatile polarization dependence of the (S)PMR signal. To demonstrate this in more detail, let us consider the spatial Fourier components of the crystal susceptibility tensors via the expansion

$$\begin{aligned} \chi(\mathbf{r}, \mathbf{r}', \omega) &= \sum_{\mathbf{g}} \int \chi(\mathbf{k}, \mathbf{k}_g, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} e^{-i\mathbf{g} \cdot \mathbf{r}'} d^3\mathbf{k}, \\ \chi(\mathbf{k}, \mathbf{k}_g, \omega) &= \sum_{i,j} \chi^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) \mathbf{e}_i \mathbf{e}_j, \end{aligned} \quad (9)$$

where $\mathbf{k}_g = \mathbf{k} + \mathbf{g}$. The second expansion in Eq. (9) is expressed in a space-rotated Cartesian coordinate system $i, j \in (x, y, z)$ defined with respect to the quantization axis, i.e., the z axis, which is typically chosen parallel to the external

TABLE I. Coordinate systems involved in the evaluation of the geometry-dependence of (S)PMR in the presence of hyperfine splitting.

Symbols or basic vectors	
Crystal (original) coordinate system	(x, y, z)
Detection-plane coordinate system	(x', y', z')
Quantization-axis coordinate system	(x, y, z)
Spherical coordinate system	$(\epsilon_{+1}, \epsilon_0, \epsilon_{-1})$
Polarization coordinate system	$(\mathbf{e}_\sigma, \mathbf{e}_\pi, \mathbf{e}_k) = (\mathbf{e}'_y, \mathbf{e}'_x, \mathbf{e}'_z)$

magnetic field. We note that the space-rotated quantization-axis coordinate system (x, y, z) here should be distinguished from the original space-fixed Cartesian coordinate system (x, y, z) based on the crystal surface. In fact, as listed in Table I, there is another space-rotated coordinate system in our scheme for the polarizations of the parametric beam, which is spanned by $(\mathbf{e}_k, \mathbf{e}_\sigma, \mathbf{e}_\pi)$, as shown in Figs. 3(b) and 3(c). Here $\mathbf{e}_\sigma$ and $\mathbf{e}_\pi$ are the unit polarization vectors of the incident plane wave in the reverse picture, which are perpendicular and parallel, respectively, to the diffraction plane, i.e., $\mathbf{e}_\sigma \parallel (\mathbf{k} \times \mathbf{g})$ and $\mathbf{e}_\pi \parallel (\mathbf{k} \times \mathbf{e}_\sigma)$, and $\mathbf{e}_k = \mathbf{k}/k$ is the unit Mössbauer wave vector.

In the case of (S)PMR with the detection frequency close to the Mössbauer resonance frequency ω_0 , both atomic electrons and nuclei contribute to the parametric signal beam. Thus, the spatial Fourier components of the crystal susceptibility tensor $\chi(\mathbf{r}, \omega)$ should be expressed in the general case as

$$\chi(\mathbf{k}, \mathbf{k}', \omega) = \chi_e(\mathbf{k}, \mathbf{k}', \omega) + \chi_n(\mathbf{k}, \mathbf{k}', \omega). \quad (10)$$

Here, $\chi_e(\mathbf{k}, \mathbf{k}', \omega)$ and $\chi_n(\mathbf{k}, \mathbf{k}', \omega)$ are the electronic and nuclear parts, respectively. Under the two-wave diffraction dynamics, $\mathbf{k}'$ is equal to $\mathbf{k}$ or $\mathbf{k}_g$, such that the Fourier components of susceptibilities in Eq. (10) can be denoted as $\chi(\mathbf{k}, \mathbf{k}, \omega) = \chi_0(\omega)$ and $\chi(\mathbf{k}, \mathbf{k}_g, \omega) = \chi_g(\omega)$.

2. Electronic susceptibility

The electronic part of crystal susceptibility in Eq. (10) dominantly arises from the $E1$ (electric dipole)-atomic transitions, and its components take the explicit form [23,46]

$$\chi_e^{ij}(\mathbf{k}, \mathbf{k}', \omega) = -\delta_{ij} \frac{4\pi c^2 r_e}{\omega^2 \Omega_0} \sum_a f_{ij,a}(\mathbf{g}, \omega) e^{ig\mathbf{r}_a}, \quad (11)$$

where Ω_0 is the volume of the crystallographic unit cell, $\mathbf{r}_a$ is the position of the a th atom in the crystallographic unit cell, $r_e = e^2/4\pi\epsilon_0 m_e c^2$ is the classical electron radius, and $f_{ij,a}(\mathbf{g}, \omega)$ is the coherent-scattering amplitude of the a th atom with the anomalous dispersion corrections of the atomic scattering factor being included. Since the electronic susceptibility does not depend on the presence of the hyperfine magnetic field $\mathbf{B}_{\text{hf}}$, one has $\chi_{(0,g)e}^{xx} = \chi_{(0,g)e}^{yy} = \chi_{(0,g)e}^{zz} = \chi_{(0,g)e}$. For the (011) reflection of the α -iron crystal, which is widely used for Mössbauer spectroscopy based on x-ray nuclear-resonance diffraction [47–49], $\chi_{0e} = -0.15 \times 10^{-4} + 0.69 \times 10^{-6} i$ and $\chi_{ge} = -0.10 \times 10^{-4} + 0.67 \times 10^{-6} i$ at the relevant Mössbauer frequency $\hbar\omega_0 = 14.41$ keV [50].

3. Nuclear susceptibility with hyperfine splitting

The nuclear contribution to the crystal susceptibility plays the key role in (S)PMR. Its components in the case of hyperfine splitting take the form [42,51]

$$\chi_n^{ij}(\mathbf{k}, \mathbf{k}', \omega) = -\frac{4\pi c^3}{\omega^3 \Omega_0} \sum_a f_{ij,a}(\mathbf{k}, \mathbf{k}', \omega) e^{ig\mathbf{r}_a}, \quad (12)$$

where the nuclear scattering amplitude is given by

$$\begin{aligned} f_{ij,a}(\mathbf{k}, \mathbf{k}', \omega) &= \frac{2I_e + 1}{2(2I_g + 1)} \eta_a \sum_{m_e, m_g} 3 \begin{pmatrix} I_g & 1 & I_e \\ -m_g & q & m_e \end{pmatrix}^2 \\ &\times \frac{\tilde{\gamma}/2}{\omega - \omega(m_e, m_g) + i\gamma_0/2} \mathcal{P}_{-q}^{ij}(\mathbf{k}, \mathbf{k}') f_{\text{LM},a}(\mathbf{k}, \mathbf{k}'). \end{aligned} \quad (13)$$

Here I_e and I_g are the nuclear spins of the excited state and ground state, respectively. The magnetic quantum numbers m_g and m_e of the ground and excited states determine the polarizations of the hyperfine Zeeman transitions via $q = -\Delta m = m_g - m_e$. Specifically, linear polarization, denoted by π^0 , and right- and left-circular polarizations, denoted by σ^+ and σ^- , are determined by $q = 0, -1$, and $+1$, respectively. The elastic linewidth $\tilde{\gamma}$ is related to the total natural linewidth γ_0 via $\tilde{\gamma} = \gamma_0/(1 + \alpha_c)$, with α_c being the internal-conversion coefficient, η_a is the abundance of the isotope, and $f_{\text{LM},a}(\mathbf{k}, \mathbf{k}')$ is the Lamb-Mössbauer factor. The polarization factor $\mathcal{P}_{-q}^{ij}(\mathbf{k}, \mathbf{k}')$ in Eq. (13) is determined by the nature of the transition:

$$\begin{aligned} E1 : \mathcal{P}_{-q}^{ij}(\mathbf{k}, \mathbf{k}') &= \epsilon_{-q}^i \epsilon_{-q}^{j*}, \\ M1 : \mathcal{P}_{-q}^{ij}(\mathbf{k}, \mathbf{k}') &= \frac{(\mathbf{k} \times \boldsymbol{\epsilon}_{-q})^i (\mathbf{k}' \times \boldsymbol{\epsilon}_{-q}^*)^j}{kk'}, \end{aligned} \quad (14)$$

where $\boldsymbol{\epsilon}_{\pm 1} = \mp(\mathbf{e}_x \pm i\mathbf{e}_y)/\sqrt{2}$ and $\boldsymbol{\epsilon}_0 = \mathbf{e}_z$ are the spherical bases expressed in terms of the quantization-axis Cartesian coordinate system $(\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z)$, and $\mathbf{e}_z \parallel \mathbf{B}_{\text{hf}}$ is defined as the quantization axis. If the hyperfine splitting is absent and one assumes that the Mössbauer crystal only contains one isotope, by using the sum rules of the $3j$ symbol, Eq. (12) can be simplified to [23,30,43,46]

$$\begin{aligned} \chi_n^{ij}(\mathbf{k}, \mathbf{k}', \omega) &= -\frac{4\pi c^3}{\omega^3 \Omega_0} \frac{2I_e + 1}{2(2I_g + 1)} \frac{\eta e^{-W_{\text{LM}}}}{1 + \alpha_c} \mathcal{P}^{ij}(\mathbf{k}, \mathbf{k}') \\ &\times S(\mathbf{k}, \mathbf{k}') \frac{\gamma_0/2}{\omega - \omega_0 + i\gamma_0/2}, \end{aligned} \quad (15)$$

where $S(\mathbf{k}, \mathbf{k}') = \sum_a e^{-i(\mathbf{k}' - \mathbf{k}) \cdot \mathbf{r}_a}$ is the structure factor, and the Lamb-Mössbauer factor $f_{\text{LM}} = e^{-W_{\text{LM}}}$. The polarization factor now is the sum of all possible polarization factors of different q [23,46]:

$$\begin{aligned} E1 : \mathcal{P}^{ij}(\mathbf{k}, \mathbf{k}') &= \sum_q \mathcal{P}_{-q}^{ij} = \delta_{ij}, \\ M1 : \mathcal{P}^{ij}(\mathbf{k}, \mathbf{k}') &= \sum_q \mathcal{P}_{-q}^{ij} = \frac{(\mathbf{k} \cdot \mathbf{k}') \delta_{ij} - k_i k_j}{kk'}, \end{aligned}$$

$$E2: \mathcal{P}^{ij}(\mathbf{k}, \mathbf{k}') = \sum_q \mathcal{P}_{-q}^{ij} = \frac{(\mathbf{k} \cdot \mathbf{k}')\delta_{ij} + k'_i k_j - 2k_i k'_j}{kk'}, \quad (16)$$

where k_i (k'_j) is the projection of the wave vector $\mathbf{k}$ ($\mathbf{k}'$) onto the i (j) axis of the quantization-axis Cartesian coordinate system.

Due to the presence of the $M1$ -polarization factors $\mathcal{P}_{0,\pm 1}^{ij}$ in the nuclear susceptibilities, the $M1$ -interactions between the generated parametric radiation of a specific polarization (σ or π polarization) and the Mössbauer crystal depend strongly on the orientation of $\mathbf{B}_{\text{hf}}$ (the quantization axis). In principle, the source-free x-ray diffraction problem can be solved by projecting the electric fields and susceptibility tensors onto an arbitrary coordinate system. However, only the electric-field components of the incident and diffracted waves with the same polarizations (σ or π polarizations) are coupled. Therefore, it is more convenient to employ the polarization Cartesian coordinate system spanned by $(\mathbf{e}_k, \mathbf{e}_\sigma, \mathbf{e}_\pi)$, as shown in Figs. 3(b) and 3(c). Then, only the susceptibility components $\chi_{0,g}^{(\sigma)} = \mathbf{e}_\sigma \cdot \chi_{0,g} \cdot \mathbf{e}_\sigma$ and $\chi_{0,g}^{(\pi)} = \mathbf{e}_\pi \cdot \chi_{0,g} \cdot \mathbf{e}_\pi$ are relevant for diffraction. Obviously, both $\chi_{0,g}^{(\sigma)}$ and $\chi_{0,g}^{(\pi)}$ rely on the orientation of the quantization axis with respect to the polarization vectors $\mathbf{e}_\sigma$ and $\mathbf{e}_\pi$. We thus specialize to the following two cases for our further discussion: (i) The magnetic field is perpendicular to the diffraction plane, i.e., $\mathbf{B}_{\text{hf}} \parallel (\mathbf{k} \times \mathbf{k}_g)$, as shown in Fig. 3(b). (ii) The magnetic field is parallel to the diffraction plane, i.e., $\mathbf{B}_{\text{hf}} \perp (\mathbf{k} \times \mathbf{k}_g)$, as shown in Fig. 3(c).

4. Nuclear susceptibility of ^{57}Fe crystal

We now focus the discussion of the susceptibilities to the six-resonance structure of α -iron. For the (011) reflection of α -iron, the relevant parameters for the nuclear susceptibility can be taken from the x-ray database as follows [50]: The crystal interplanar spacing $d_{(011)} = 2.03 \text{ \AA}$ and the Mössbauer wave number of the ^{57}Fe isotope $k_0 = 7.29 \text{ \AA}^{-1}$ give the Bragg angle $\theta_B \approx 12.26^\circ$. The volume of the crystallographic unit cell is $\Omega_0 = d^3$ with the cell constant $d = 2.87 \text{ \AA}$, the internal conversion coefficient $\alpha_c = 8.56$, the structure factor $S(\mathbf{k}, \mathbf{k}') = 2$, and the total natural linewidth $\hbar\gamma_0 = 4.66 \text{ neV}$. The Lamb-Mössbauer factor of the α -iron crystal at room temperature 300 K is $f_{\text{LM}} \approx 0.76$, corresponding to the Debye temperature $\Theta_D^{\text{Fe}} = 470 \text{ K}$. The specific expressions for the nuclear susceptibilities $\chi_{0,g}^{(\sigma)}$ and $\chi_{0,g}^{(\pi)}$ of ^{57}Fe crystal are presented in Appendix B.

In Fig. 4, we investigate the ratios $|\chi_g(\omega)/\chi_0(\omega)|$ and $|\chi_{\text{gn}}(\omega)/\chi_{\text{ge}}|$ as a function of the dimensionless frequency $\omega_x = (\omega - \omega_0)/(\gamma_0/2)$ around the Mössbauer resonance frequency ω_0 for the (011) reflection of the α -iron crystal. This ratio $|\chi_g(\omega)/\chi_0(\omega)|$ determines the amplitude of the diffracted field via the dynamical coupling between the diffracted and incident x-ray waves (see Sec. IV). The second ratio, $|\chi_{\text{gn}}(\omega)/\chi_{\text{ge}}(\omega_0)|$, approximately characterizes the excess nuclear scattering over the electronic counterpart and plays a crucial role in (S)PMR, in contrast with PXR. Figures 4(a)–4(d) show the σ -polarization components of the relevant susceptibilities, with the hyperfine splitting of the isotope ^{57}Fe

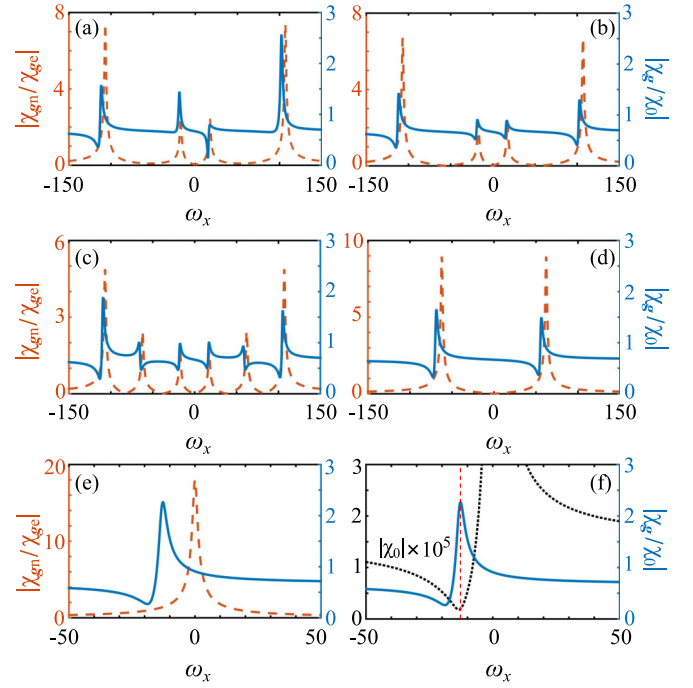


FIG. 4. (a)–(e) The ratios $|\chi_g(\omega)/\chi_0(\omega)|$ (blue solid lines) and $|\chi_{\text{gn}}(\omega)/\chi_{\text{ge}}|$ (red dashed lines) as a function of the dimensionless frequency $\omega_x = (\omega - \omega_0)/(\gamma_0/2)$ around the Mössbauer resonance frequency ω_0 for the (011) reflection of the α -iron crystal. Panels (a)–(d) show the cases with hyperfine splitting, while panel (e) is without hyperfine splitting. In panel (a), the hyperfine magnetic field is perpendicular to the diffraction plane, i.e., $\mathbf{B}_{\text{hf}} \parallel (\mathbf{k} \times \mathbf{k}_g)$. In panels (b)–(d), the hyperfine magnetic field is in the diffraction plane, i.e., $\mathbf{B}_{\text{hf}} \perp (\mathbf{k} \times \mathbf{k}_g)$, but for different orientations: (b) $\xi_B = 0^\circ$, (c) $\xi_B = 45^\circ$, and (d) $\xi_B = 90^\circ$. (f) Comparison between $|\chi_g(\omega)/\chi_0(\omega)|$ (blue solid line) in panel (e) and the corresponding $|\chi_0(\omega)|$ (black dotted line, scaled by 10^5). The frequencies at which $|\chi_0(\omega)|$ is minimal and $|\chi_g(\omega)/\chi_0(\omega)|$ is maximal approximately coincide. The results correspond to the σ polarization, perpendicular to the diffraction plane.

being taken into account for two orientations of $\mathbf{B}_{\text{hf}}$. Figure 4(a) corresponds to $\mathbf{B}_{\text{hf}} \parallel (\mathbf{k} \times \mathbf{k}_g)$, while Figs. 4(b)–4(d) correspond to $\mathbf{B}_{\text{hf}} \perp (\mathbf{k} \times \mathbf{k}_g)$ with different angles ξ_B between $\mathbf{B}_{\text{hf}}$ and the incident wave vector $\mathbf{k}$: $\xi_B = 0^\circ$ in Fig. 4(b), $\xi_B = 45^\circ$ in Fig. 4(c), and $\xi_B = 90^\circ$ in Fig. 4(d). In Fig. 4(e), we also present the case without hyperfine splitting for comparison. In the case where $\mathbf{B}_{\text{hf}}$ is perpendicular to the diffraction plane, the x-ray scattering predominantly arises from the nuclear resonance transitions of $\sigma^\pm$ polarizations due to the relatively large values of $|\chi_{\text{gn}}(\omega)/\chi_{\text{ge}}(\omega_0)| \approx 7.38$ at the highest- and the lowest-frequency Zeeman transitions $\omega_x \approx \pm 106.9$, as shown in Fig. 4(a). However, in the case where $\mathbf{B}_{\text{hf}}$ is parallel to the diffraction plane, as shown in Figs. 4(b)–4(d), the spectral distributions of both the ratios $|\chi_g(\omega)/\chi_0(\omega)|$ and $|\chi_{\text{gn}}(\omega)/\chi_{\text{ge}}(\omega_0)|$ undergo a geometric transition from four-peak and six-peak to double-peak structures as the angle ξ_B increases from 0° to 90° , indicating a strong polarization dependence of the scattering. It is worth noting that the maximal value of the ratio $|\chi_{\text{gn}}(\omega)/\chi_{\text{ge}}(\omega_0)|$ increases as the number of hyperfine spectral peaks decreases. This is most obvious in the degenerate case where the isotope ^{57}Fe is modeled by

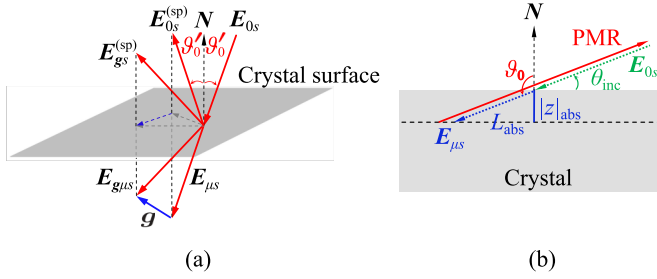


FIG. 5. (a) Four-beam interactions in the x-ray dynamical diffraction problem at the crystal surface, including the specularly reflected diffracted wave $E_{0s}^{(sp)}$, the forward-transmitted wave $E_{\mu s}$, the Bragg-diffracted wave $E_{g\mu s}$ inside the crystal, and the specularly diffracted reflected wave $E_{gs}^{(sp)}$ outside the crystal. The reflection angle of the specularly reflected wave $E_{0s}^{(sp)}$ and the incident angle of the incident wave E_{0s} with respect to the outside normal vector of the crystal surface $N = e_z$ are equal ($\vartheta_0' = \pi - \vartheta_0$) due to the same parallel component of their wave vectors $k_{||}$. (b) Sketch of the absorption length L_{abs} and the absorption depth $|z|_{abs}$. The propagation distance of the forward-transmitted wave $E_{\mu s}$ (blue dashed arrow) within the crystal is limited by the absorption length $L_{abs} = |z|_{abs} / \sin \theta_{inc} = |z|_{abs} / |v_0|$. Here, $|z|_{abs} = (k_0 \text{Im}[\varepsilon_{\mu s}])^{-1}$ is the absorption depth (blue solid line) and $\theta_{inc} = \vartheta_0 - \pi/2$ is the incident angle of the incident wave in the reverse picture, corresponding to the actual emission direction of the generated (S)PMR (red solid arrow) in the real picture.

a two-level system. As shown in Fig. 4(e), in this degenerate case, the maximal value of $|\chi_{gn}(\omega)/\chi_{ge}(\omega_0)| \approx 17.8$ is achieved at $\omega = \omega_0$ resulting from all six Zeeman hyperfine transitions together. Although the nuclear resonance can be characterized by the ratio $|\chi_{gn}(\omega)/\chi_{ge}(\omega_0)|$ or $|\chi_{gn}(\omega)|$, the emission peak could be shifted from the resonance frequency, as illustrated in Fig. 4(f) for the degenerate case as an example. This is due to the interference between electronic and nuclear scatterings, i.e., $|\chi_g(\omega)| = |\chi_{ge}(\omega_0) + \chi_{gn}(\omega)|$, rather than by the nuclear term $|\chi_{gn}(\omega)|$ alone. This situation is well described by the Fano-type interference, which is known to lead to non-Lorentzian line shapes and frequency shifts in the spectrum [52].

IV. X-RAY DYNAMICAL-DIFFRACTION APPROACH

A. Two-wave coupling equations

When Eq. (1) is applied to calculate the radiation generated by the interactions between relativistic charged particles and a crystal, the electric field $E_{ks}^{(+)}(\mathbf{r}, \omega)$ can be solved for within the framework of two-wave dynamical x-ray diffraction theory [23]. The latter assumes the existence of only two relevant waves inside the crystal: a forward-transmitted wave $E_{\mu s}$ and a Bragg-diffracted wave $E_{g\mu s}$, which arises from the diffraction of the crystallographic lattice planes characterized by the reciprocal-lattice vector g . For the grazing-incidence geometry of the electron beam, as considered in the grazing EAD geometry [34,35], the wave vector of the forward-transmitted wave inside the crystal is nearly parallel to the incident wave vector k in vacuum and can be written as $k_{\mu s} = k - k\varepsilon_{\mu s}N$. Here $N = e_z$ denotes the unit outward normal vector of the crystal surface. The wave vector of the Bragg-diffracted wave

$E_{g\mu s}$ is then given by $k_{g\mu s} = k_g - k\varepsilon_{\mu s}N$ with $k_g = k + g$. Thus, the parameter $\varepsilon_{\mu s}$ characterizes the deviation of the transmitted wave vector inside the crystal from the incident wave vector in vacuum. The amplitudes of the transmitted and the diffracted waves inside the crystal are dominated by the two-wave dispersion relations [23,53],

$$\begin{aligned} \left[\frac{k_{\mu s}^2}{k^2} - (1 + \chi_0^{(s)}) \right] E_{\mu s} - c_s \chi_{-g}^{(s)} E_{g\mu s} &= 0, \\ \left[\frac{k_{g\mu s}^2}{k^2} - (1 + \chi_0^{(s)}) \right] E_{g\mu s} - c_s \chi_g^{(s)} E_{\mu s} &= 0, \end{aligned} \quad (17)$$

with the spatial Fourier components of the polarization-dependent crystal susceptibilities $\chi_0^{(s)} = \chi^{(s)}(\mathbf{k}, \mathbf{k}, \omega)$ and $\chi_g^{(s)} = (\chi_{-g}^{(s)})^* = \chi^{(s)}(\mathbf{k}, \mathbf{k}_g, \omega)$. The polarization factors $c_\sigma = 1$ and $c_\pi = \cos(\mathbf{k}, \mathbf{k}_g) = \cos(2\theta_B)$ correspond to the polarization vectors e_σ and e_π , respectively, with θ_B being the Bragg angle. In the following, unless otherwise specified, we temporarily omit the polarization index of the susceptibilities.

B. Exact four-beam solutions

The nontrivial solution of Eq. (17) can be obtained by the condition of a vanishing determinant, i.e.,

$$\begin{aligned} & \left[\frac{k_{\mu s}^2}{k^2} - (1 + \chi_0) \right] \left[\frac{k_{g\mu s}^2}{k^2} - (1 + \chi_0) \right] - c_s^2 \chi_{-g} \chi_g \\ &= \varepsilon_{\mu s}^4 - 2(v_0 + v_g)\varepsilon_{\mu s}^3 + (4v_0v_g - \alpha_B - 2\chi_0)\varepsilon_{\mu s}^2 \\ &+ [2v_0(\alpha_B + \chi_0) + 2\chi_0v_g]\varepsilon_{\mu s} \\ &+ \chi_0(\alpha_B + \chi_0) - c_s^2 \chi_g \chi_{-g} = 0, \end{aligned} \quad (18)$$

which also determines the wave vectors of the relevant electromagnetic waves. Here, $\alpha_B = -(\mathbf{k}_g^2 - \mathbf{k}^2)/k^2$ is the Bragg deviation parameter, $v_0 = \mathbf{k} \cdot \mathbf{N}/k = \cos \vartheta_0$, and $v_g = \mathbf{k}_g \cdot \mathbf{N}/k$. The two-wave dispersion equation (18) is a fourth-order algebraic equation and thus has four solutions ε_{1s} , ε_{2s} , ε_{3s} , and ε_{4s} , which can be solved numerically. Therefore, in general, there exist four transmitted waves and thus four corresponding diffracted waves inside the crystal. However, two of these solutions are unphysical, since their imaginary parts $\text{Im}[\varepsilon_{3s}]$, $\text{Im}[\varepsilon_{4s}] < 0$, which would correspond to exponentially amplified amplitudes of the respective electric fields inside the crystal. For this reason, only two transmitted waves, E_{1s} , E_{2s} , and the corresponding two diffracted waves, E_{g1s} , E_{g2s} , inside the crystal are relevant to the boundary problem.

As shown in Fig. 5(a), a plane x-ray wave E_{0s} with the amplitude $|E_{0s}| = 1$ and the wave vector k is incident on the crystal surface. Due to the conservation of the parallel components of the wave vectors at the crystal surface, two reflected waves, i.e., the specularly reflected wave $E_{0s}^{(sp)}$ and the reflected diffracted wave $E_{gs}^{(sp)}$ with the parallel components of the wave vectors $(\mathbf{k}_0^{(sp)})_{||} = \mathbf{k}_{||}$ and $(\mathbf{k}_g^{(sp)})_{||} = \mathbf{k}_{g||} = \mathbf{k}_{||} + \mathbf{g}_{||}$, respectively, need to be considered in general. Thus, the boundary problem is treated as a four-beam $[E_{\mu s}, E_{g\mu s}, E_{0s}^{(sp)}, E_{gs}^{(sp)}]$ reflection-diffraction case [40,54–56], which yields the following boundary

equations:

$$\begin{aligned} 1 + E_{0s}^{(sp)} &= E_{1s} + E_{2s}, \\ E_{gs}^{(sp)} &= E_{g1s} + E_{g2s}, \\ \nu_0 + \nu_0^{(sp)} E_{0s}^{(sp)} &= (\nu_0 - \varepsilon_{1s})E_{1s} + (\nu_0 - \varepsilon_{2s})E_{2s}, \\ \nu_g^{(sp)} E_{gs}^{(sp)} &= (\nu_g - \varepsilon_{1s})E_{g1s} + (\nu_g - \varepsilon_{2s})E_{g2s}, \end{aligned} \quad (19)$$

with the two-wave dynamical coupling relations obtained from Eq. (17) for the forward-transmitted and the Bragg-diffracted fields inside the crystal

$$\begin{aligned} E_{g1s} &= -\frac{c_s \chi_g}{\alpha_B + \chi_0 + (2\nu_g - \varepsilon_{1s})\varepsilon_{1s}} E_{1s}, \\ E_{g2s} &= -\frac{c_s \chi_g}{\alpha_B + \chi_0 + (2\nu_g - \varepsilon_{2s})\varepsilon_{2s}} E_{2s}, \end{aligned} \quad (20)$$

where $\nu_0^{(sp)} = -\nu_0 = \mathbf{k}_0^{(sp)} \cdot \mathbf{N}/k$ and $\nu_g^{(sp)} = \mathbf{k}_g^{(sp)} \cdot \mathbf{N}/k$. Then the explicit forms of all relevant electric fields which are valid for arbitrary incident angle are obtained as

$$\begin{aligned} E_{1s} &= \frac{(\nu_0 - \nu_0^{(sp)})D_{2s}}{(\nu_0 - \nu_0^{(sp)} - \varepsilon_{1s})D_{2s} - (\nu_0 - \nu_0^{(sp)} - \varepsilon_{2s})D_{1s}}, \\ E_{2s} &= \frac{(\nu_0 - \nu_0^{(sp)})D_{1s}}{(\nu_0 - \nu_0^{(sp)} - \varepsilon_{2s})D_{1s} - (\nu_0 - \nu_0^{(sp)} - \varepsilon_{1s})D_{2s}}, \\ E_{0s}^{(sp)} &= E_{1s} + E_{2s} - 1, \\ E_{gs}^{(sp)} &= E_{g1s} + E_{g2s}, \end{aligned} \quad (21)$$

with Eq. (20) and

$$\begin{aligned} D_{1s} &= (\nu_0 - \nu_g^{(sp)} - \varepsilon_{1s})[(2\nu_0 - \varepsilon_{1s})\varepsilon_{1s} + \chi_0], \\ D_{2s} &= (\nu_0 - \nu_g^{(sp)} - \varepsilon_{2s})[(2\nu_0 - \varepsilon_{2s})\varepsilon_{2s} + \chi_0]. \end{aligned} \quad (22)$$

C. Parameter cases and approximate solutions

For the situation in which the outgoing PXR wave vector is *not* close to the crystal surface, the above solutions can be approximated by estimating the orders of magnitude for all the relevant parameters as follows:

(1) The parameter ν_g characterizes the angular spread of the Bragg-diffracted wave vector $\mathbf{k}_g$, which is nearly parallel to the crystal surface such that it aligns with the electron velocity. The upper limit of $|\nu_g|$ can therefore be estimated from the condition of total reflection at x-ray energies, i.e., $\cos \theta_g = (1 + \chi_g')^{1/2} \approx 1 - |\chi_g'|/2$, where θ_g is the angle between $\mathbf{k}_g$ and the x axis and $\chi_g' = \text{Re}[\chi_g]$ is the real part of χ_g . Thus we have $|\nu_g| \sim |\theta_g| \sim \sqrt{|\chi_{0,g}|} \approx 10^{-2}$ to 10^{-3} .

(2) For the Bragg deviation parameter α_B , we consider the relevant case in which the emission direction of parametric beam is close to the Bragg angle. Then, $|\alpha_B|$ is of the order of γ^{-2} . For electrons with energy $E \approx 100$ MeV as in the Mainz Microtron (MAMI) synchrotron, $\gamma^{-1} \approx 10^{-3}$. Thus $|\alpha_B| \sim \gamma^{-2} \approx 10^{-5}$ to 10^{-6} , which is of the order of $|\chi_{0,g}|$. However, for XFEL electrons with several GeV energy, a different condition is realized: $|\alpha_B| \sim \gamma^{-2} \approx 10^{-8}$ to 10^{-9} , such that $|\alpha_B| \ll |\chi_{0,g}|$.

(3) The parameter ν_0 is independent of the electron energy, implying that the detection direction can, in principle, be

arbitrary. In the grazing EAD geometry, the wave vector of the parametric beam is perpendicular to the crystal surface, so that the parametric beam exits the crystal along the shortest possible path with minimal absorption [23,34,35]. In this case, $|\nu_0| = |\cos \vartheta_0| = 1$, which is much larger than $|\nu_g|$, $|\chi_{0,g}|$, and $|\alpha_B|$. For (S)PMR, however, the emission direction can no longer be expected to be perpendicular to the crystal surface due to the Mössbauer resonance condition. Nevertheless, as long as the emission direction is not extremely close to the crystal surface, it can still be assumed that $|\nu_0|$ remains the largest among the above parameters. This assumption will be quantified and discussed in more detail in Sec. VI A.

Based on the above considerations, the relevant parameter relations for XFEL electrons can be summarized as

$$|\nu_0| \gg |\nu_g| (\sim \sqrt{|\chi_{0,g}|}) \gg |\chi_{0,g}| \gg |\alpha_B|. \quad (23)$$

With this hierarchy, the exact solutions of the electric fields in Eqs. (20) and (21) are approximated as

$$\begin{aligned} E_{1s} &\approx E_{0s} = 1, \quad E_{2s} \approx 0, \\ E_{g1s} &\approx -\frac{c_s \chi_g}{\alpha_B + \chi_0}, \quad E_{g2s} \approx 0, \\ E_{0s}^{(sp)} &\approx 0, \quad E_{gs}^{(sp)} \approx E_{g1s}. \end{aligned} \quad (24)$$

Also, Eq. (18) can be simplified to

$$\begin{aligned} \varepsilon_{\mu s}^4 - 2\nu_0 \varepsilon_{\mu s}^3 + 4\nu_0 \nu_g \varepsilon_{\mu s}^2 + [2\nu_0(\alpha_B + \chi_0)]\varepsilon_{\mu s} \\ + \chi_0(\alpha_B + \chi_0) = c_s^2 \chi_{-g} \chi_g. \end{aligned} \quad (25)$$

After employing the perturbative approach, it is found that two solutions, ε_{3s} and ε_{4s} , are still unphysical, and one of the remaining two solutions ε_{2s} is comparably large and thus not relevant. Therefore, only the solution ε_{1s} fixes the amplitudes of the transmitted and the diffracted waves. In Sec. VI, we further substantiate this result by considering the asymptotic regimes of the diffraction geometry, where the outgoing wave vectors are nearly parallel to the crystal surface.

For the regular PXR generated by the electrons moving inside the crystal, in the grazing EAD geometry, the parametric beam is dominated by the crystal diffracted wave [23,34]

$$\begin{aligned} \mathbf{E}_{g1s}(\mathbf{r}, \omega) &= \mathbf{e}_{gs} E_{g1s} e^{i\mathbf{k}_{g1s} \cdot \mathbf{r}}, \\ E_{g1s} &\approx -\frac{c_s \chi_g}{\alpha_B + \chi_0}. \end{aligned} \quad (26)$$

If the electron beam is moving in vacuum but slightly above the crystal surface, which is the so-called ‘‘Smith-Purcell geometry’’ [35], a parametric beam can also be generated. It is determined by the specularly reflected diffracted wave outside the crystal in the reverse picture

$$\begin{aligned} \mathbf{E}_s^{(sp)}(\mathbf{r}, \omega) &= \mathbf{e}_{gs} E_s^{(sp)} e^{i\mathbf{k}_{sp} \cdot \mathbf{r}}, \\ E_s^{(sp)} &\approx E_{g1s}, \end{aligned} \quad (27)$$

where $\mathbf{k}_{sp} = \mathbf{k}_{g||} + k_{sp,z} \mathbf{N}$ and the evanescent wave vector component $k_{sp,z}$ is fixed as $k_{sp,z} = [k^2 - (\mathbf{k}_{||} + \mathbf{g}_{||})^2]^{1/2}$.

We note that, unlike in the conventional EAD-PXR geometry, where $|\nu_0| = 1$ ($\vartheta_0 = 45^\circ$ and $g = \sqrt{2}k_0$) [23,34,35], the emission direction of (S)PMR generated by the grazing-incidence electron beam is generally not perpendicular to the crystal surface. In this diffraction geometry, the parametric

beam propagates at a shallower exit angle from the crystal, resulting in a longer absorption length $L_{\text{abs}} = (k_0 \text{Im}[\varepsilon_{\mu s}][v_0])^{-1}$ (L_{abs} is minimal at $|v_0| = 1$), as illustrated in Fig. 5(b). This limitation motivates us to develop (S)PMR theory here in more general diffraction geometries with arbitrary (S)PMR emission angle, enabling the search for optimal (S)PMR diffraction configurations, as they are utilized in the companion paper [39] for developing a powerful (S)PMR source.

V. ANGULAR-SPECTRAL DISTRIBUTION OF SPMR PHOTONS

A. Coherent amplitude

We now focus on the SPMR case to calculate the properties of the emitted radiation, i.e., the angular-spectral distribution and total number of the generated photons. To this end, we insert the diffracted fields derived above in Eq. (1). This generalizes the approach in Ref. [36] by explicitly incorporating the Mössbauer resonance condition as well as the transverse distance of XFEL electron beam from the crystal surface. Thereby, our analytical formalism can simultaneously account for the diffracted fields both inside and outside the crystal. Furthermore, we extend the theoretical analysis beyond the grazing EAD case to general diffraction geometries.

The time integral in Eq. (1) leads to a generalized coherent amplitude of the emitted radiation, denoted as $\mathcal{A}_s(\mathbf{k})$ in the following. For PXR generated by an electron beam, this coherent amplitude can be naturally decomposed into two contributions corresponding to the regions where the electrons are distributed inside ($z_0 \leq 0$) and outside ($z_0 \geq 0$) the crystal, respectively. Accordingly, the time integral in Eq. (1) can be expressed as $\mathcal{A}_s(\mathbf{k}) = \mathcal{A}_s^+(\mathbf{k}) + \mathcal{A}_s^-(\mathbf{k})$, where

$$\mathcal{A}_s^\pm(\mathbf{k}) = \int d^2\theta_e \int d^3r_0 \mathcal{D}(\mathbf{r}_0) \rho(\theta_e) \theta(\pm z_0) \times \int_0^{\frac{L}{v}} \mathbf{E}_{gs}^\pm \cdot \mathbf{v} e^{ik_g^\pm r(t)} e^{i\omega t} dt, \quad (28)$$

with the total spatial distribution function of the initial electron positions

$$\mathcal{D}(\mathbf{r}_0) = \sum_{l=0}^K \mathcal{D}_l(\mathbf{r}_l) \mathcal{D}_x(x_0^{(l)}). \quad (29)$$

The superscripts “ $\pm$ ” in Eq. (28) correspond to the regions outside and inside the crystal, respectively, and the Heaviside step function $\theta(\pm z_0)$ is introduced to formally separate the integrations over the initial electron positions in these two regions. The general coherent amplitude $\mathcal{A}_s^\pm(\mathbf{k})$ with $\mathbf{E}_{gs}^- = E_{g1s} \mathbf{e}_{gs}$ and $\mathbf{k}_{gs}^- = \mathbf{k}_{g1s}$ is contributed to by the dominant Bragg-diffracted wave inside the crystal, and the amplitude $\mathcal{A}_s^+(\mathbf{k})$ with $\mathbf{E}_{gs}^+ = E_{gs}^{(\text{sp})} \mathbf{e}_{gs}$ and $\mathbf{k}_{gs}^+ = \mathbf{k}_{\text{sp}}$ corresponds to the specularly reflected diffracted wave outside the crystal.

In the following, we drop the polarization subscript “ s ” for notational convenience. Furthermore, instead of considering the explicit expressions of $\mathbf{k}_{g1s}$ and $\mathbf{k}_{\text{sp}}$, we proceed with our calculation in a more general way, which is applicable to both the inside and the outside of the crystal. Thus, we can also drop the superscripts $\pm$ in $\mathbf{k}_{gs}^\pm$ so that $k_{g,j}$ ($j \in \{x, y, z\}$) can be the j component of either $\mathbf{k}_{g1s}$ or $\mathbf{k}_{\text{sp}}$. In fact, $\mathbf{k}_{g1s}$ and $\mathbf{k}_{\text{sp}}$ differ

only in their z components, such that $k_{g1s,x} = k_{\text{sp},x} = k_{g,x}$ and $k_{g1s,y} = k_{\text{sp},y} = k_{g,y}$, as shown in Eq. (D5). The derivation of the expressions for the diffraction wave vectors ($\mathbf{k}_g, \mathbf{k}_g^{(\text{sp})}$) and other related quantities in the two-wave dynamical diffraction formalism for SPMR is presented in detail in Appendix D.

After performing the integrations in Eq. (28) with the Gaussian bunch profile in Eq. (3), we split the result in different factors in order to gain physical insight into the contributions to the coherent amplitudes,

$$\mathcal{A}_s^\pm(\mathbf{k}) = \mathbf{E}_{gs}^\pm \cdot \mathbf{v} \mathcal{A}_x \mathcal{A}_z^\pm \mathcal{K}^\pm, \quad (30)$$

where

$$\mathcal{K}^\pm = e^{-\sigma^2/4}, \quad (31a)$$

$$\mathcal{A}_z^\pm = \frac{1}{2} [1 \mp \text{Erf}(\Phi_3)] e^{ik_{g,z} a_0}, \quad (31b)$$

$$\mathcal{A}_x = \frac{1 - e^{-ik_{g,x} L_b}}{K(1 - e^{-ik_{g,x} d_0})} \frac{\sqrt{\pi}}{\delta} e^{-q_c^2/\delta^2} [\text{Erf}(\Phi_1) - \text{Erf}(\Phi_2)]. \quad (31c)$$

Here $\text{Erf}(x)$ is the Gaussian error function, and the definitions of the parameters $\delta, \sigma, \Phi_1, \Phi_2, \Phi_3$, and q_c are

$$q_c = k_{g,x} v + \omega, \quad (32a)$$

$$\delta^2 = (k_{g,y}^2 + k_{g,z}^2) v^2 \sigma_a^2, \quad (32b)$$

$$\sigma^2 = k_{g,x}^2 \sigma_x^2 + (k_{g,y}^2 + k_{g,z}^2) \sigma_t^2, \quad (32c)$$

$$\Phi_1 = \frac{L\delta}{2v} - i \frac{q_c}{\delta}, \quad (32d)$$

$$\Phi_2 = -i \frac{q_c}{\delta}, \quad (32e)$$

$$\Phi_3 = -\frac{a_0}{\sigma_t} - \frac{i}{2} k_{g,z} \sigma_t. \quad (32f)$$

From the factorization in Eq. (30), one can see that the amplitudes $E_{gs}^\pm$ of the diffracted fields for the single-electron case are now governed by the function $\mathcal{A}_x$ with a longitudinal periodic modulation. The functions $\mathcal{A}_z^\pm$ with the Gaussian error function arise from the dependence on the transverse distance a_0 of the electron beam to the crystal surface, which reflect the amplitude and phase modulations along the transverse direction. Additionally, the coherent amplitudes decay with the photon angular spread θ , as described by $\mathcal{K}^\pm$. According to Eq. (D5), the factors $\exp(ik_{g,z}^\pm a_0)$ give rise to the crystal-absorption term $\exp(-k \text{Im}[\varepsilon_{1s}] |a_0|)$ ($a_0 \leq 0$) inside the crystal and the evanescent-decay term $\exp(-k \eta a_0)$ ($a_0 \geq 0$) with the real-valued evanescence rate $\eta = i v_g^{(\text{sp})}$ in vacuum, respectively. We also note that multiple-electron scattering can be considered inside the crystal by replacing the variance of the electron angular spread, σ_a , by $\tilde{\sigma}_a = (\sigma_a^2 + \theta_s^2)^{1/2}$, where θ_s quantifies the multiple scattering [30,36,37].

It is important to note that the condition of superradiant amplification is included in $\mathcal{A}_x$. Explicitly, the superradiant amplification of the coherent amplitude is determined by the condition of constructive interference $1 - \exp(-ik_{g,x} d_0) = 0$ with the virtual-photon wave number $k_{g,x} \approx \omega/v$, which can

also be expressed by the “superradiance frequency” as

$$\omega_s^{(n)} = n \frac{2\pi v}{d_0}, \quad n \in \{1, 2, \dots\}. \quad (33)$$

Before we continue with the derivation, we estimate the parameters appearing in Eq. (32) to justify subsequent approximations. The XFEL electron energies can reach several GeV [57]—we adopt $E_e = 17.5$ GeV for the EuXFELs [58,59], which corresponds to $\gamma \approx 34143$. The typical angular divergence of XFEL electron beams is of the order of μrad . For a bunch charge $Q = 1$ nC, the emittance is $\epsilon \approx 2 \times 10^{-11}$ m rad. Accordingly, we consider two representative beam cases: (i) without beam focusing, $\sigma_a = 1$ μrad and $\sigma_t = 20$ μm , and (ii) with beam focusing, $\sigma_a = 10$ μrad and $\sigma_t = 2$ μm . The crystal length available for PXR generation is generally limited by the absorption length $L_{\text{abs}} = (k\text{Im}[\chi_g])^{-1} \approx 10^{-4}$ cm [23,31]. However, in the grazing EAD geometry, where the electron beam propagates along the crystal surface, the effective interaction length can scale with the full crystal length L even when $L \gg L_{\text{abs}}$ [23,34]. In that case, the crystal length is instead constrained by the coherence length L_{coh} , with $L_{\text{coh}}^{\text{max}} \approx 2\gamma^2/k$ [23,60]. This length can be of the order of several centimeters; however, to suppress multiple-scattering effects, we choose a crystal length $L = 1$ cm. With the above parameters, one finds that $|q_c/\delta| \gg 1$ and $|q_c v/(L\delta^2)| \gg 1$, which justifies the approximation

$$\frac{\sqrt{\pi}}{\delta} e^{-\frac{q_c^2}{\delta^2}} [\text{Erf}(\Phi_1) - \text{Erf}(\Phi_2)] \approx \frac{1 - e^{iq_c \frac{L}{v}}}{-iq_c} \quad (34)$$

in the amplitude $\mathcal{A}_x$.

B. Angular-spectral distribution and total photon number

Now we turn back to Eq. (1) for the angular-spectral distribution of the number of SPMR photons. With Eqs. (30)–(34), and in the limit of $L_b/d_0 = K \gg 1$, Eq. (1) becomes

$$\begin{aligned} \frac{\partial^2 N_{n,\omega s}}{\partial \omega \partial \Omega} &= N_e^2 \frac{e_0^2 \omega}{8\pi^2 \epsilon_0 \hbar c^2 L_b} |f_s^+(\mathbf{k}) + f_s^-(\mathbf{k})|^2 \\ &\times \sum_{n=1}^K \delta(\omega - \omega_s^{(n)}) \left| \frac{1 - e^{iq_c \frac{L}{v}}}{q_c} \right|^2, \end{aligned} \quad (35)$$

where $\omega_s^{(n)} = 2\pi v n/d_0$ ($n \in \{1, 2, \dots\}$) is the superradiance frequency of the n th-order SPMR harmonic component, and $f_s^\pm(\mathbf{k}) = \mathbf{E}_{gs}^\pm \cdot \mathbf{v} \mathcal{A}_z^\pm \mathcal{K}^\pm$ are the reduced coherent amplitudes. To achieve superradiant enhancement of the PMR intensity, the Mössbauer resonance condition must be satisfied; that is, at least one of the δ peaks in Eq. (35) should coincide with the Mössbauer frequency ω_0 . However, the spectral separation between adjacent superradiant harmonics, $\Delta\omega_n = 2\pi v/d_0 \approx \omega_0$, is much larger than the Mössbauer linewidth γ_0 . Consequently, only a single δ peak contributes to the SPMR. The SPMR angular distribution can then be obtained by integrating Eq. (35) around the superradiance frequency $\omega_0 = 2\pi v n/d_0$ of the n th-order harmonic component, yielding

$$\frac{\partial^2 N_{n,s}}{\partial \theta_x \partial \theta_y} = N_e^2 \frac{e_0^2 \omega_0}{8\pi^2 \epsilon_0 \hbar c^2 L_b} |f_s^+(\boldsymbol{\theta}) + f_s^-(\boldsymbol{\theta})|^2 \left| \frac{1 - e^{iq_0 \frac{L}{v}}}{q_0} \right|^2, \quad (36)$$

where $q_0 = q_c(\omega_0) = \omega_0 \theta_x v_{x'x}$. Next, we apply the relation

$$\left| \frac{1 - e^{iq_0 \frac{L}{v}}}{q_0} \right|^2 \approx \frac{2\pi L}{v} \delta(q_0) = \frac{2\pi L}{v \omega_0 |v_{x'x}|} \delta(\theta_x), \quad (37)$$

in the limit of $k_0 L \gg 1$ and integrate over θ_x and θ_y . This yields the total number of SPMR photons generated within a single harmonic component

$$N_\sigma = \frac{N_e^2 e_0^2}{4\pi \epsilon_0 \hbar c^3} \frac{L}{L_b |v_{x'x}|} \int |f_\sigma^+(\theta_y) + f_\sigma^-(\theta_y)|^2 d\theta_y, \quad (38)$$

where $f_\sigma^\pm(\theta_y)$ are calculated at $\theta_x = 0$ and $\omega = \omega_0$. From this expression, it is evident that the polarization of SPMR remains unchanged in the transition from the conventional grazing EAD geometry to the generalized SPMR geometry. The emitted photons are predominantly polarized perpendicular to the diffraction plane (σ -polarization wave) [36].

VI. GEOMETRY DEPENDENCE OF DIFFRACTION DYNAMICS

In this section, we employ the general applicability of the analytical framework developed above to study the dependence of SPMR on the scattering geometry. This analysis likewise applies to PXR in the absence of a Mössbauer resonance condition. In the reverse picture of PXR generated by a multi-electron beam, the total coherent amplitude $\mathcal{A}_s^\pm$ in Eq. (30) arises from the modulation of individual diffraction modes by the transverse distribution of the electron beam. Accordingly, we first analyze the dependence of the electric field associated with a single diffraction mode on the geometry. In terms of physics, this corresponds to the idealized single-electron case, representing the most fundamental step toward identifying the optimal diffraction configuration reported in the companion paper [39]. Subsequently, we explore how the attenuation characteristics of the diffracted waves responsible for the SPMR (or SPXR) generation depend on the diffraction geometry. Finally, we include the envelope modulation due to the transverse electron distribution in the analysis.

A. Electric fields

In Fig. 6, we show the dependencies of the amplitudes of all relevant electric fields on the cutting angle Θ_0 of the crystal surface (the angle between the reciprocal lattice vector $\mathbf{g}$ and the surface normal direction $\mathbf{N}$) over its entire range from the grazing EAD geometry up to the grazing SD case ($45^\circ \leq \Theta_0 \leq 90^\circ$). The results are shown for the (011) reflection of the α -iron crystal described above. Upon increasing Θ_0 , the emission direction of the parametric beam gradually inclines toward the crystal surface. From Figs. 6(a) and 6(b), however, we find that the amplitudes of all relevant electric fields remain approximately independent of the diffraction geometry for cutting angles up to about $\Theta_0 \approx 89^\circ$. This confirms the validity of the approximate solutions of the x-ray dynamical diffraction problem in Eq. (24) over a wide range, $\theta_B(12.26^\circ) \leq \Theta_0 \lesssim 89^\circ$, corresponding to an incidence angle range $90.42^\circ \lesssim \vartheta_0 \leq 114.52^\circ$ and thus $7.4 \times 10^{-3} \lesssim |v_0| \leq 0.42$.

The case of higher Θ_0 approaching the limiting case of $\Theta_0 = 90^\circ$ requires particular attention. As highlighted by the

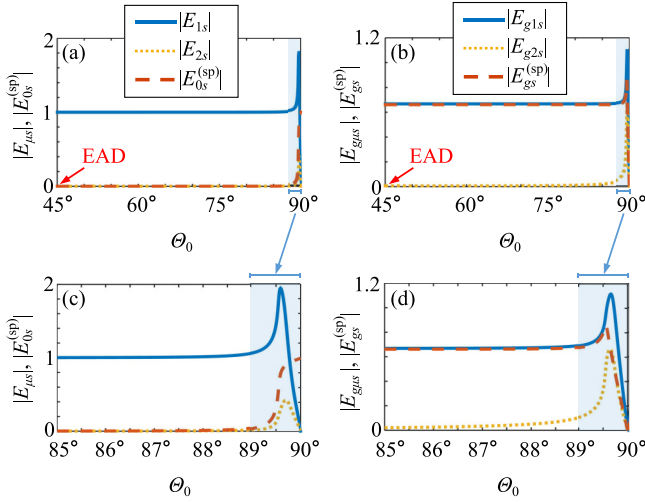


FIG. 6. (a), (b) Moduli of the amplitudes of all relevant electric fields as functions of the cutting angle θ_0 , ranging from the grazing EAD geometry ($\theta_0 = 45^\circ$) up to 90° . Panel (a) shows the forward-transmitted fields E_{1s} (blue solid line), E_{2s} (yellow dotted line), and the specularly reflected field $E_{0s}^{(sp)}$ (red dashed line). Panel (b) shows the Bragg-diffracted field E_{g1s} (blue solid line), E_{g2s} (yellow dotted line), and the specularly reflected diffracted field $E_{gs}^{(sp)}$ (red dashed line). The amplitudes are normalized to the incident plane wave in vacuum ($|E_{0s}| = 1$). Panels (c) and (d) show the magnifications of the region of higher θ_0 for panels (a) and (b), respectively. Results are presented for $s = \sigma$ polarization, using the (011) reflection of the α -iron crystal. The blue shaded areas denote the grazing SD regime, for which the approximate solutions of the two-wave dynamical diffraction problem are no longer valid.

blue-shaded regions in Figs. 6(c) and 6(d), which are the magnifications of Figs. 6(a) and 6(b), respectively, in this limit, the amplitudes of all electric fields decrease and vanish at $\theta_0 = 90^\circ$, except for the specularly reflected field $E_{0s}^{(sp)}$, whose amplitude returns to unity.

However, the most notable feature of the geometry dependence is that the forward-transmitted fields inside the crystal (E_{1s} and E_{2s}) and the corresponding Bragg-diffracted fields (E_{g1s} , E_{g2s} , and $E_{gs}^{(sp)}$) exhibit maxima at nearly the same large cutting angle in the grazing SD geometry. Moreover, the transmission coefficient $|E_{1s}/E_{0s}|$ exceeds unity, as shown in Fig. 6(c). This can be understood by noting that, on the one hand, the crystal acts as a continuous medium with an average refractive index slightly less than unity, so that x-rays incident at small grazing angles experience total external reflection. At the same time, the Bragg-resonance condition enables the coherent dynamical coupling between the forward-transmitted and the Bragg-diffracted waves. Consequently, the amplitudes of all diffracted fields (E_{g1s} , E_{g2s} , and $E_{gs}^{(sp)}$) exhibit pronounced peaks in the grazing SD geometry, as shown in Fig. 6(d). This feature offers a favorable condition for optimizing SPMR intensity, as we utilize in the companion paper [39]. However, the approximate field solutions in Eq. (24), which were employed in Refs. [30,34–36], break down in the grazing SD regime and must be replaced by the exact solutions given in Eqs. (19)–(21).

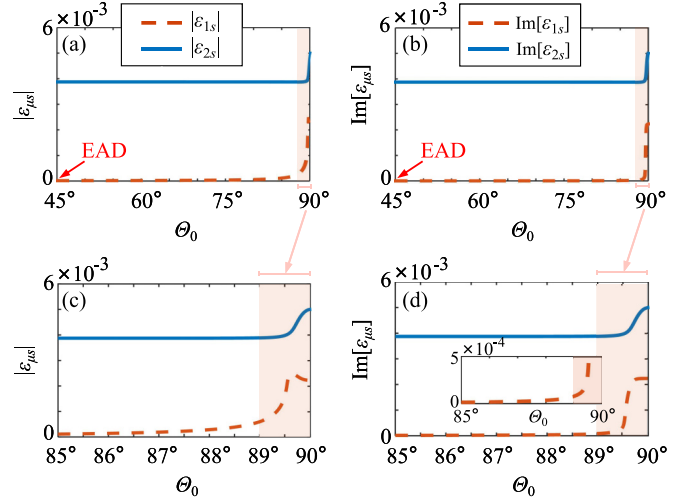


FIG. 7. (a) Moduli of the solutions ϵ_{1s} (red dashed line) and ϵ_{2s} (blue solid line), and (b) their imaginary parts $\text{Im}[\epsilon_{1s}]$ (red dashed line) and $\text{Im}[\epsilon_{2s}]$ (blue solid line) as functions of the cutting angle θ_0 , ranging from the grazing EAD geometry ($\theta_0 = 45^\circ$) up to 90° . Panels (c) and (d) show the magnifications of the region of higher θ_0 for panels (a) and (b), respectively. The inset in panel (b) shows the magnification of the region only for $\text{Im}[\epsilon_{1s}]$, in which the rapid increase of $\text{Im}[\epsilon_{1s}]$ is indicated by the red shaded area. Results are presented for $s = \sigma$ polarization, using the (011) reflection of the α -iron crystal. The red shaded areas denote the grazing SD regime, for which the approximate solutions of the two-wave dynamical diffraction problem are no longer valid.

B. Diffracted-wave attenuation

Further insight into the geometry-dependent propagation characteristics of the diffracted waves in the grazing SD regime is provided by Fig. 7, which shows the dependence of $|\epsilon_{1s}|$ and $|\epsilon_{2s}|$ and the imaginary parts $\text{Im}[\epsilon_{1s}]$ and $\text{Im}[\epsilon_{2s}]$ on θ_0 , as determined by Eq. (18).

It can be seen from Figs. 7(a) and 7(b) that over a wide range of diffraction geometries from the grazing EAD case up to $\theta_0 = 90^\circ$, the magnitude of the solution ϵ_{2s} remains on the order of $\sqrt{|\chi_{0,g}|} \approx 10^{-3}$. In contrast with the usual case of PXR, where the absorption is governed by $|\chi_0''| = |\text{Im}[\chi_0]| \approx 10^{-5}$ to 10^{-6} , the solution ϵ_{2s} implies a much stronger absorption of the parametric beam by the crystal. As a result, the contribution of the corresponding diffracted field E_{g2s} to the total intensity is negligible [23,34], despite its amplitude peak in Fig. 6(d). However, in the grazing SD regime, Fig. 7(d) shows that $\text{Im}[\epsilon_{1s}]$ increases rapidly as θ_0 approaches 90° , rising from a second-order-small quantity $\text{Im}[\epsilon_{1s}] \sim |\chi_{0,g}| \approx 10^{-5}$ to a first-order-small quantity $\text{Im}[\epsilon_{1s}] \sim \text{Im}[\epsilon_{2s}] \approx 10^{-3}$. This indicates that, in the grazing SD regime, the diffracted wave E_{g1s} also experiences a strong absorption within the crystal.

The transverse attenuation of the diffracted waves is manifested in the imaginary parts of their wave vectors. As shown in Eq. (D5), the difference between $\mathbf{k}_{g\mu s}$ and $\mathbf{k}_{sp}$ lies in their z components: The former determines the absorption of the Bragg-diffracted field $E_{g\mu s}$ inside the crystal, whereas the latter governs the evanescent decay of the specularly reflected diffracted field $E_{gs}^{(sp)}$ in vacuum.

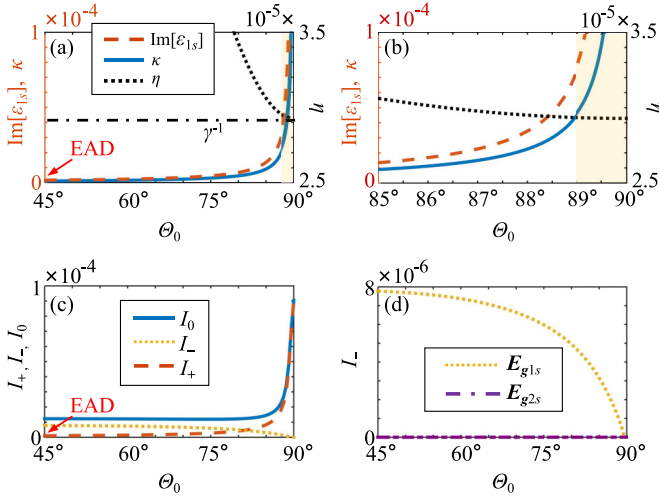


FIG. 8. (a) Transverse attenuation rates of the specularly reflected diffracted wave η (black dotted line, $\theta_x = \theta_y = 10^{-4}$ rad), the Bragg-diffracted wave $\text{Im}[\varepsilon_{1s}]$ (red dashed line, $\theta_x = \theta_y = 0$), with its approximate value $\kappa = \chi_g''/(2 \sin \theta_{\text{inc}})$ (blue solid line) as functions of the cutting angle Θ_0 , ranging from the grazing EAD geometry ($\Theta_0 = 45^\circ$) up to 90° . For $\theta_x = \theta_y = 0$, $\eta = \gamma^{-1} \approx 2.93 \times 10^{-5}$ is indicated by the black dash-dotted line. (b) Magnified view of the higher- Θ_0 region of panel (a). (c) Integrals I_+ , I_- , and I_0 of the transverse modulation function as functions of Θ_0 for $a_0 = 0$. Panel (d) shows a comparison of I_- evaluated by the dominant Bragg-diffracted wave E_{g1s} (yellow dotted line) and by the negligible Bragg-diffracted wave E_{g2s} (purple dash-dotted line, on the order of 10^{-9}). Results are presented for $s = \sigma$ polarization, using the (011) reflection of the α -iron crystal.

The amplitude of the Bragg-diffracted field E_{g1s} decays as $\exp(-k\text{Im}[\varepsilon_{1s}]|z|)$ and can be approximated at lower cutting angles as $\exp[-k\chi_g''|z|/(2 \sin \theta_{\text{inc}})]$ [34] with $\chi_g'' = \text{Im}[\chi_g]$ and the incident angle $\theta_{\text{inc}} = \vartheta_0 - \pi/2$ [see Fig. 5(b)]. As already discussed and shown in Figs. 8(a) and 8(b), its absorption coefficient, $\text{Im}[\varepsilon_{1s}]$, increases rapidly with Θ_0 . In contrast, the evanescence rate η of the specularly reflected diffracted field $E_{gs}^{(\text{sp})}$ can decrease with Θ_0 and remains essentially constant for the ideal case $\theta_x = \theta_y = 0$. For $\gamma = 34\,143$, one obtains $\eta = \gamma^{-1} \approx 2.93 \times 10^{-5}$, which is orders of magnitude lower than $\text{Im}[\varepsilon_{1s}]$ ($\approx 10^{-3}$) in the grazing SD regime. At lower cutting angles, the attenuation rates $\text{Im}[\varepsilon_{1s}]$ and η are comparable (both are on the order of 10^{-5}), such that the Bragg-diffracted wave E_{g1s} may dominate the (S)PMR intensity. However, once Θ_0 exceeds a certain critical value, particularly in the grazing SD regime at a higher Θ_0 , the contribution from the specularly reflected diffracted wave $E_{gs}^{(\text{sp})}$ may surpass that of E_{g1s} . This opposite dependence can effectively compensate for the absorption of E_{g1s} and therefore ultimately lead to the dominance of the specularly reflected diffracted field in the total (S)PMR intensity.

C. Transverse modulation

It is worth noting that the competition between the attenuation characteristics of the Bragg-diffracted wave E_{g1s} and the specularly reflected diffracted wave $E_{gs}^{(\text{sp})}$ is further modulated by the transverse distribution of the electron

beam, which determines how the individual electron contributions to the attenuation add coherently. Specifically, owing to the continuous transverse Gaussian distribution of the XFEL electron bunch, in the reverse picture, the specularly reflected diffracted wave experiences amplitude modulation through the evanescence factor $\exp(-k\eta z)$ ($z \geq 0$). In contrast, the Bragg-diffracted wave undergoes not only amplitude modulation via $\exp(-k\text{Im}[\varepsilon_{1s}]|z|)$ ($z \leq 0$), but also phase modulation arising from the real part of ε_{1s} . To quantitatively examine how these transverse modulations of the attenuation characteristics influence the geometry dependence of the SPMR intensity, we consider the following integrals based on Eq. (38):

$$I_+ = \int |\mathcal{A}_z^+(\theta_y) \mathcal{K}^+(\theta_y)|^2 d\theta_y, \quad (39a)$$

$$I_- = \int |\mathcal{A}_z^-(\theta_y) \mathcal{K}^-(\theta_y)|^2 d\theta_y, \quad (39b)$$

$$I_0 = \int |\mathcal{A}_z^+(\theta_y) \mathcal{K}^+(\theta_y) + \mathcal{A}_z^-(\theta_y) \mathcal{K}^-(\theta_y)|^2 d\theta_y, \quad (39c)$$

where $\mathcal{K}^\pm$ and $\mathcal{A}_z^\pm$ are given by Eq. (31) evaluated at $\theta_x = 0$. These integrals can be understood as the SPMR intensities corresponding to the diffracted waves in the reverse picture with unit diffracted-field amplitudes, where I_- and I_+ represent the individual contributions of E_{g1s} and $E_{gs}^{(\text{sp})}$, respectively, to I_0 . Although separating the modulation functions $\mathcal{A}_z^+ \mathcal{K}^+$ and $\mathcal{A}_z^- \mathcal{K}^-$ from the complete coherent amplitudes $\mathcal{A}_z^\pm$ in Eq. (31) does not correspond to a physically realizable situation, this quantitative treatment nevertheless provides useful insight into how the overall attenuation characteristics of the relevant diffracted waves determine the SPMR intensity.

In Fig. 8(c), we show $I_\pm$ and I_0 as functions of Θ_0 over a wide range starting from the grazing EAD case, which governs the envelope modulation of the SPMR intensity. The results show that the contribution from the specularly reflected diffracted wave, I_+ , becomes dominant in the grazing SD regime at very large Θ_0 , precisely the range in which the electric-field amplitudes reach their maximum. Figure 8(d) compares the values of I_- evaluated for the two Bragg-diffracted waves inside the crystal, E_{g1s} and E_{g2s} . It confirms the negligible contribution of E_{g2s} owing to its relatively large absorption coefficient, $\text{Im}[\varepsilon_{2s}]$, over the entire range of Θ_0 .

VII. SPMR PHOTONS

A. Total parametric photons

Building on the analysis of the diffracted waves and their attenuation characteristics presented above, we now discuss the total number of the superradiant parametric photons, N_{SPMR} , generated per electron bunch. The total photon yield is dominated by the spectrally broad nonresonant SPXR component, produced through the electron scattering χ_{0e} and χ_{ge} , with a comparably small spectrally narrow SPMR contribution from the near-resonant Mössbauer nuclear response $\chi_{0n}(\omega)$ and $\chi_{gn}(\omega)$. Nevertheless, because the PXR source under consideration contains a Mössbauer crystal, we still refer to the total superradiant parametric photons—including both the nonresonant electronic contribution (SPXR) and

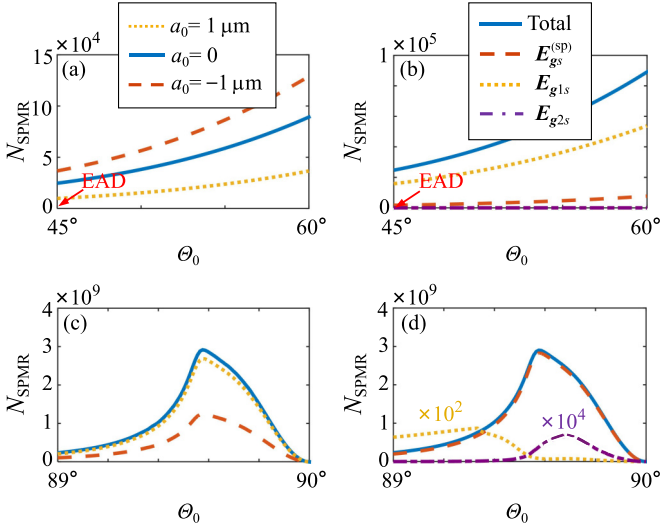


FIG. 9. Total number of SPMR photons per pulse, N_{SPMR} . (a) N_{SPMR} as a function of the cutting angle Θ_0 in the lower-angle range $45^\circ \leq \Theta_0 \leq 60^\circ$, starting from the grazing EAD geometry, for different transverse distances of the electron beam from the crystal surface: $a_0 = 1 \mu\text{m}$ (yellow dotted line), $a_0 = 0$ (blue solid line), and $a_0 = -1 \mu\text{m}$ (red dashed line). (b) Decomposition of N_{SPMR} for $a_0 = 0$ from panel (a) into the contributions from different individual diffracted waves: calculated using only $E_{gs}^{(\text{sp})}$ (red dashed line), only E_{g1s} (yellow dotted line), only E_{g2s} (purple dot-dashed line), and the total N_{SPMR} (blue solid line). Panels (c) and (d) show the corresponding results of panels (a) and (b), respectively, but in the higher-angle range $89^\circ \leq \Theta_0 \leq 90^\circ$, i.e., the grazing SD geometry. In panel (d), the contributions from E_{g1s} (yellow dotted line) and E_{g2s} (purple dot-dashed line) are scaled by factors of 10^2 and 10^4 , respectively, for comparison. Results are shown for the (011) reflection of the α -iron crystal with the electron-beam size $\sigma_b = 2 \mu\text{m}$. Polarization of SPMR is $s = \sigma$.

the near-resonant nuclear contribution—as SPMR photons, N_{SPMR} , throughout this discussion. For an SPMR source, although the contribution from the nuclear scattering constitutes the centerpiece of interest, it remains instructive to examine the dependence of N_{SPMR} on the diffraction geometry over the whole range. This examination not only corroborates the physical mechanisms analyzed above, including the asymptotic behavior of the electric fields, the attenuation properties of the diffracted waves, and the influence of the multi-electron transverse modulation, but also motivates the geometrical optimization scheme reported in the companion paper [39]. Moreover, the insights gained here are equally applicable to other SPXR sources even in the absence of nuclear isotopes.

We first examine in Fig. 9(a) the dependence of N_{SPMR} , calculated for the (011) reflection of the α -iron crystal, on the cutting angle Θ_0 in the lower range starting from the grazing EAD geometry $45^\circ \leq \Theta_0 \leq 60^\circ$. Results are presented for three representative transverse positions of the electron beam relative to the crystal surface: outside the crystal ($a_0 = 1 \mu\text{m}$), inside the crystal ($a_0 = -1 \mu\text{m}$), and exactly along the surface ($a_0 = 0$). Figure 9(b) further decomposes the total value of N_{SPMR} for the case of $a_0 = 0$ in Fig. 9(a) into the individual contributions from all relevant diffracted waves: E_{g1s} , E_{g2s} ,

and $E_{gs}^{(\text{sp})}$. The exact value of N_{SPMR} results from the coherent superposition of these diffraction components. Figure 9(a) reproduces the well-known grazing EAD result that electrons traveling inside the crystal generate substantially stronger parametric radiation than those propagating above the surface. Figure 9(b) further demonstrates that, in this regime, electrons traveling exactly along the crystal surface still yield negligible contributions from both the specularly reflected diffracted wave $E_{gs}^{(\text{sp})}$ and the Bragg-diffracted wave E_{g2s} . This justifies the common assumption made in Refs. [30,34,36] that only the diffracted wave E_{g1s} provides a significant channel for the EAD-PXR, while the contributions from all other diffracted modes are negligible.

For comparison, Figs. 9(c) and 9(d) show the counterparts of Figs. 9(a) and 9(b) but in the grazing SD regime, where the cutting angle spans $89^\circ \leq \Theta_0 \leq 90^\circ$. As seen in Fig. 9(c), the value of N_{SPMR} exhibits a pronounced peak, in striking contrast with the behavior in the grazing EAD case shown in Fig. 9(a). This enhancement arises from the combined effects of the relatively strong diffracted fields at a large Θ_0 and the weak attenuation of the specularly reflected diffracted wave. Notably, as shown in Fig. 9(c), opposite the behavior in Fig. 9(a), the electrons moving outside the crystal ($a_0 = 1 \mu\text{m}$) can generate a substantially higher SPMR intensity than those moving inside the crystal ($a_0 = -1 \mu\text{m}$). As already discussed in Sec. VIB, in this grazing SD geometry, the diffraction plane lies extremely close to the crystal surface, so that the parametric beam generated inside the crystal must traverse a significantly longer path before exiting, undergoing much stronger absorption. If the electrons move slightly above the crystal surface, the specular diffraction of electron virtual-photon fields on the crystal surface could effectively compensate for the absorption losses experienced by internally generated photons, thus dominating the SPMR generation and yielding a markedly enhanced SPMR intensity. The comparison in Fig. 9(d) confirms this mechanism: The dominant contribution arises from the specularly reflected diffracted wave $E_{gs}^{(\text{sp})}$ in the grazing SD geometry, while the contributions from the Bragg-diffracted waves E_{g1s} and E_{g2s} inside the crystal are at least two orders of magnitude lower.

B. Parametric Mössbauer photons

1. Emission spectra

After discussing the total number of photons emitted per pulse from the SPMR source, we turn to examine the parametric photons distributed around the Mössbauer resonance. The quantities of these near-resonant parametric photons represent the most physically relevant aspect of SPMR as a novel Mössbauer radiation source. To this end, we now focus on the extremely narrow frequency window within the overall broad SPMR spectral range discussed above.

Figure 10 shows the SPMR emission spectra $n_{\text{SPMR}}(\omega)$ for the (011) reflection of the α -iron crystal around the Mössbauer resonance, plotted as a function of the dimensionless frequency $\omega_x = (\omega - \omega_0)/(\gamma_0/2)$, for several different scenarios including different diffraction geometries, electron beam size, and hyperfine configurations. Figures 10(a)–10(d) display the spectra at a lower cutting angle $\Theta_0 = 45^\circ$ in the grazing EAD geometry considered in Ref. [30] with the transverse electron-

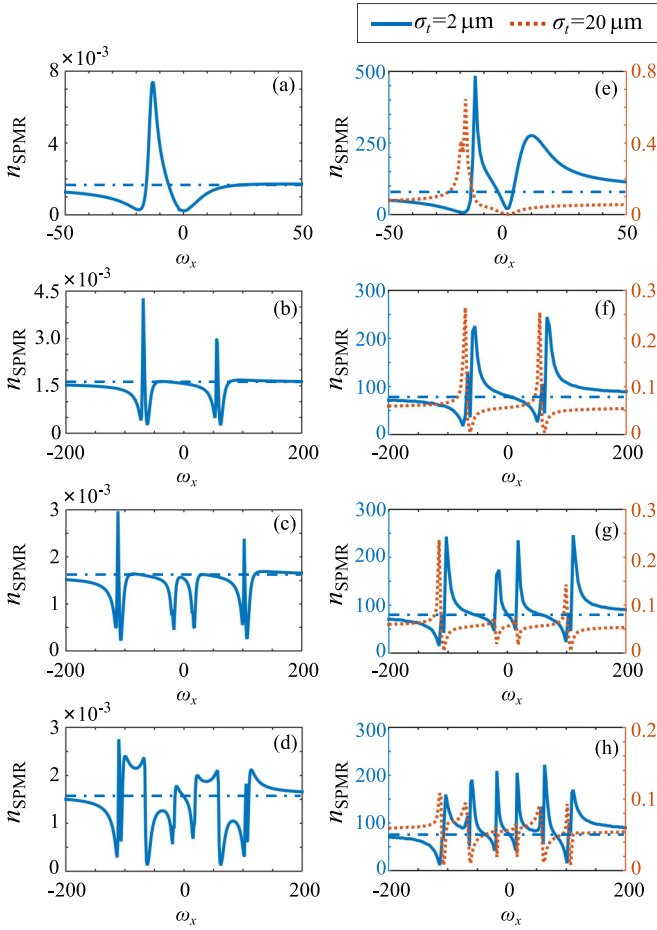


FIG. 10. SPMR emission spectra around the Mössbauer resonance, plotted as a function of the dimensionless frequency $\omega_x = (\omega - \omega_0)/(\gamma_0/2)$. Panels (a)–(d) show the emission spectra for the geometric parameters $(\Theta_0, a_0) = (45^\circ, 0)$, with the electron-beam transverse size $\sigma_t = 2 \mu\text{m}$. Panels (e)–(f) show the corresponding results in panels (a)–(d) in the grazing SD geometry at larger cutting angles. Panels (a) and (e) represent the degenerate cases without nuclear hyperfine splitting, while the remaining panels represent the realistic cases including nuclear hyperfine splitting with different orientations of the hyperfine magnetic field $\mathbf{B}_{\text{hf}}$ with respect to the SPMR emission direction $\mathbf{k}_0$: (b), (f) $\xi_B = \pi/2$, (c), (g) $\xi_B = 0$, and (d), (h) $\xi_B = \pi/4$. In panels (e)–(h), the blue solid lines correspond to $(\Theta_0, a_0) = (89.45^\circ, 0.4 \mu\text{m})$ with $\sigma_t = 2 \mu\text{m}$, and the red dashed lines correspond to $(\Theta_0, a_0) = (89.9^\circ, 0.4 \mu\text{m})$ with $\sigma_t = 20 \mu\text{m}$. Results are calculated for the (011) reflection of the α -iron crystal with the SPMR polarization $s = \sigma$. The pure electronic-scattering contributions n_{SPMR} calculated only by χ_{0e} and χ_{ge} are also indicated. In panels (a)–(d), $n_{\text{SPMR}} \approx 1.58 \times 10^{-3}$ (dot-dashed lines), and in panels (e)–(h), $n_{\text{SPMR}} \approx 79.8$ (dot-dashed lines) for the blue solid lines and $n_{\text{SPMR}} \approx 0.056$ for the red dashed lines.

beam position $a_0 = 0$ and the electron-beam size $\sigma_t = 2 \mu\text{m}$, for different hyperfine configurations. Figure 10(a) shows the single-line spectrum in the degenerate case, while Figs. 10(b)–10(d) show the spectra with hyperfine splitting, corresponding to the hyperfine magnetic field $\mathbf{B}_{\text{hf}}$ parallel to the diffraction plane but with different orientations: $\xi_B = \pi/2$ (doublet), $\xi_B = 0$ (quartet), and $\xi_B = \pi/4$ (sextet). Without hyperfine splitting, as shown in Fig. 10(a), a single peak appears near the

Mössbauer resonance, clearly above the electronic-scattering background. In contrast, for the nondegenerate cases shown in Figs. 10(b)–10(d), the peak-to-background ratio decreases with the increasing number of hyperfine spectral lines. This trend is roughly consistent with the dependence of the peak values on the number of hyperfine spectral lines in the nuclear-to-electronic susceptibility ratio $|\chi_{gn}/\chi_{ge}|$, and hence in $|\chi_g/\chi_0|$ illustrated in Fig. 4. Notably, Fig. 10(c) shows that only the outer hyperfine sidebands ($\omega_x \approx \pm 106.9$) produce sharp peaks emerging from the spectral dips, while the inner narrow peaks ($\omega_x \approx \pm 17.3$) are nearly invisible. This reflects the low ratio $|\chi_g/\chi_0|$ for the inner sidebands. In Fig. 4(b), the outer sidebands correspond to $|\chi_g/\chi_0| \approx 1.28$, whereas the corresponding ratio $|\chi_g/\chi_0|$ for the inner sidebands is below unity. In addition, although the more complex spectral profile in Fig. 10(d) cannot be predicted by the ratio $|\chi_g/\chi_0|$ due to the multiple-electron transverse distribution, the overall envelope of the spectrum still reflects the shape of $|\chi_g/\chi_0|$ shown in Fig. 4(d). Furthermore, as the electron beam becomes narrower, the spectral profile more closely maps onto the shape of $|\chi_g/\chi_0|$.

Figures 10(e)–10(h) present the corresponding spectra, but for the grazing SD geometry. Results are given for the geometric parameters $(\Theta_0, a_0) = (89.9^\circ, 0.4 \mu\text{m})$ with a larger electron-beam size of $\sigma_t = 20 \mu\text{m}$ (typical for EuXFEL undulator), and for $(\Theta_0, a_0) = (89.45^\circ, 0.4 \mu\text{m})$ with a smaller beam size of $\sigma_t = 2 \mu\text{m}$. A comparison of the spectra in different diffraction geometries shows that the spectral profiles are strongly modified by the grazing SD geometry at high cutting angles. In this extreme diffraction geometry, the approximate solution for the single dominant diffracted field E_{g1s} , which can be used to predict the spectral profile via the ratio $|\chi_g/\chi_0|$, is no longer valid. Instead, both diffracted fields, E_{g1s} and E_{g2s} , associated with the physical solutions ε_{1s} and ε_{2s} , contribute significantly to the resulting spectra. Nevertheless, the hyperfine-splitting features remain clearly identifiable, while increasing the electron-beam size from 2 to 20 μm reduces the spectral intensity by approximately three orders of magnitude.

Another noteworthy feature of the SPMR emission spectra is the resonance shifts of spectral lines relative to the corresponding Mössbauer resonance frequencies. The clearest example can be observed in the degenerate case shown in Fig. 8(a), the spectral resonance at $\omega_x \approx -12.8$ is shifted by nearly $6.4\gamma_0$ from ω_0 . Even if the nuclear susceptibility $\chi_{gn}(\omega)$ is resonant at ω_0 , the interference with the electronic susceptibility χ_{ge} can lead to non-Lorentzian line shapes accompanied by a resonance shift. The resulting scattering amplitude is therefore the coherent sum of a broad, nonresonant channel (electronic scattering) and a narrow resonant channel (nuclear scattering), i.e., $\chi_g(\omega) = \chi_{gn}(\omega) + \chi_{ge}(\omega_0)$. This situation is characteristic of the Fano-type interference [52], well known to lead to non-Lorentzian line shapes and frequency shifts. At low cutting angles, where the approximate electric-field solutions of the two-wave dynamical diffraction problem in Eq. (24) are valid, the maximal (S)PMR intensity occurs near the frequency at which the amplitude of the Bragg-diffracted field E_{g1s} is maximal [30]. The frequency dependence of this amplitude can be estimated by the ratio $|\chi_g(\omega)/\chi_0(\omega)|$, of which the resonance deviates from that of the purely nuclear-electronic susceptibility ratio $|\chi_{gn}(\omega)/\chi_{ge}(\omega_0)|$, as shown in

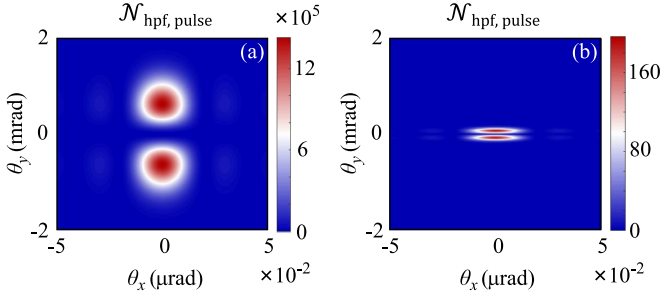


FIG. 11. Angular distribution of SPMR photons around the Mössbauer hyperfine resonance, $\mathcal{N}_{\text{hpf,pulse}}(\theta_x, \theta_y)$, generated per pulse, per unit solid angle, within a single Mössbauer natural linewidth $\Delta\omega = \gamma_0$ of a single hyperfine spectral line. The results correspond to the grazing SD geometry $(\Theta_0, a_0) = (89.2^\circ, 1 \mu\text{m})$ for two different electron-beam sizes (a) $\sigma_t = 2 \mu\text{m}$ and (b) $\sigma_t = 20 \mu\text{m}$. The angular variables θ_x and θ_y denote the deviations from the ideal Mössbauer emission direction $\mathbf{k}_0$ within the diffraction plane and perpendicular to it, respectively. Calculations are shown for the (011) reflection of the α -iron crystal, with the hyperfine configuration $\mathbf{B}_{\text{hf}} \parallel \mathbf{k}_0$ and the relevant Mössbauer hyperfine resonance $\omega_x \approx -95$.

Fig. 4(e). In fact, the maximum of $|\chi_g(\omega)/\chi_0(\omega)|$ typically occurs near the minimum of $|\chi_0(\omega)|$, consistent with the behavior shown in Fig. 4(f).

2. Angular distribution

Based on the angular-spectral distribution of SPMR photons, we can also briefly discuss the features of the angular density of Mössbauer photons, which also serves as a useful figure of merit for characterizing x-ray sources. Figure 11 presents the angular distribution of $\mathcal{N}_{\text{hpf,pulse}}$, defined as the number of photons generated per pulse, per unit solid angle in the (θ_x, θ_y) direction, and contributed to by a single hyperfine spectral line within one Mössbauer natural linewidth $\Delta\omega = \gamma_0$. The number of Mössbauer photons per pulse, N_{pulse} , is related to the angular density by $N_{\text{pulse}} = \iint \mathcal{N}_{\text{pulse}} d\theta_x d\theta_y$. Figures 11(a) and 11(b) correspond to different transverse sizes of electron beam, $\sigma_t = 2 \mu\text{m}$ and $\sigma_t = 20 \mu\text{m}$, respectively, under the geometric parameters in the grazing SD case $(\Theta_0, a_0) = (89.2^\circ, 1 \mu\text{m})$, and with the hyperfine configuration $\mathbf{B}_{\text{hf}} \parallel \mathbf{k}_0$, of which we consider the hyperfine spectral resonance at $\omega_x \approx -95$. The angular width along the θ_y direction is sensitive to the electron-beam size σ_t , since the divergence of the emitted radiation perpendicular to the diffraction plane scales as $(k_0\sigma_t)^{-1}$. As a result, the angular width $\Delta\theta_y$ in Fig. 11(b) is about an order of magnitude smaller than in Fig. 11(a). Although a larger transverse beam size generally substantially reduces the PXR intensity, as shown by the values of $\mathcal{N}_{\text{pulse}}$ in Fig. 11 and also by the SPMR results in the companion paper [39], on the other hand, it also effectively acts as a transversely larger coherent source, resulting in a more collimated emission pattern. An interesting feature of Fig. 11 is the pronounced angular anisotropy, in which the angular width confined to the diffraction plane, $\Delta\theta_x \sim (k_0L)^{-1} \approx 10^{-8} \text{ rad}$, is significantly smaller than the angular width $\Delta\theta_y$ in the perpendicular direction. This smaller value is attributed to the long-range coherence established by the grazing SD geometry of the electron beam over the entire

crystal length L [30]. Thus, the substantial enhancement of PMR angular density could be achieved not only by the combined mechanism of the superradiant amplification and the grazing SD geometry but also by the partial advantage of the grazing EAD configuration, namely, the grazing SD trajectory of the electron beam along the crystal surface, which preserves high transverse coherence along the θ_x direction in the diffraction plane and thus enables extreme angular confinement of the photons.

3. Nuclear dynamical diffraction effects

It is well known that nuclear dynamical diffraction effects may lead, for example, to spectral-line broadening or shifts of the nuclear resonances in the spectra [2,61–64]. In general, the relevance of dynamical diffraction phenomena can be assessed by comparing the extinction distance with the propagation length of the relevant waves inside the crystal [23,65,66]. When the x-ray propagation length within the crystal greatly exceeds the extinction length, multiple scattering becomes significant, and hence also dynamical diffraction effects. In the previously proposed grazing EAD geometry, the Bragg-diffracted wave $\mathbf{E}_{g\mu s}$ propagates along the crystal surface over the entire crystal length L , whereas the forward-transmitted wave $\mathbf{E}_{\mu s}$ is restricted by its absorption length L_{abs} , as illustrated in Fig. 5(b).

For the grazing-incidence geometry of the electron beam employed in the grazing EAD configuration, but away from the grazing SD regime, e.g., for a lower cutting angle $\Theta_0 \lesssim 89^\circ$, the forward-transmitted wave amplitude E_{1s} remains approximately equal to the incident-field amplitude E_{0s} , and the spectral response is mainly determined by the ratio $|\chi_g(\omega)/\chi_0(\omega)|$. In this regime, only a Fano-like spectral profile appears due to the interference between electronic scattering and nuclear scattering, and thus the intrinsic linewidth remains the natural nuclear linewidth γ_0 . This indicates that the dynamical diffraction regime is not strong enough to induce significant nuclear dynamical diffraction effects such as line broadenings (decay speedup) and resonance shifts [2,61–64].

However, this situation qualitatively changes in the grazing SD regime. When Θ_0 approaches 90° , the diffraction plane becomes nearly parallel to the crystal surface, resulting in much longer propagation lengths for the diffracted waves $\mathbf{E}_{g1s}$ and $\mathbf{E}_{g2s}$. In this extreme regime, both solutions ε_{1s} and ε_{2s} play significant roles in the field amplitudes. This corresponds to a situation in which the multiple energy exchanges between the transmitted and the diffracted waves inside the crystal lead to substantial modifications of the field amplitudes, as shown in Eqs. (19)–(21). As a result, a parameter regime of strong dynamical diffraction emerges, in which considerable spectral broadenings or Mössbauer resonance shifts may appear.

To quantify the dynamical diffraction effects, we examine in Fig. 12(a) the spectral distribution of the specularly reflected diffracted field as an illustrative example with Θ_0 near 90° . The hyperfine geometric configuration corresponds to the general case where all six hyperfine nuclear transitions contribute to the response. As Θ_0 approaches 90° , the spectral profile of the field amplitude is strongly modified, and the spectral broadenings and resonance shifts appear as a direct

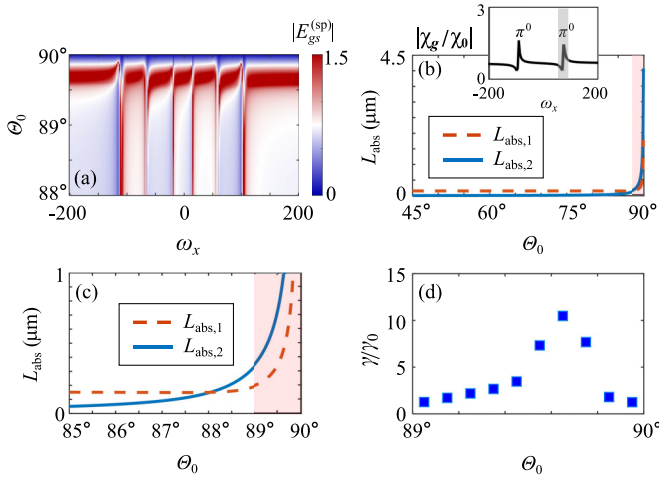


FIG. 12. (a) Spectral distribution of the amplitude of the specularly reflected diffracted field $|E_{gs}^{(sp)}(\omega)|$ for the cutting angle $88^\circ \leq \Theta_0 \leq 90^\circ$. The result corresponds to the (011) reflection of the α -iron crystal. The hyperfine magnetic field $\mathbf{B}_{hf}$ lies in the diffraction plane with the orientation angle $\xi_B = \pi/4$ such that all six hyperfine transitions contribute. (b) Absorption lengths $L_{abs,1} = (k_0 \text{Im}[\varepsilon_{1s}] \sin \theta_{in})^{-1}$ (red dashed line) and $L_{abs,2} = (k_0 \text{Im}[\varepsilon_{2s}] \sin \theta_{in})^{-1}$ (blue solid line) as functions of Θ_0 , ranging from the grazing EAD geometry $\Theta_0 = 45^\circ$ up to 90° . For panel (b), the hyperfine configuration is $\xi_B = \pi/2$, involving only two linearly polarized transitions at $\omega_x \approx \pm 62$, as indicated by the spectral distribution of the susceptibility ratio $|\chi_g(\omega)/\chi_0(\omega)|$. Panel (c) shows the magnification of the region of higher Θ_0 for panel (b). (d) Ratio of the spectral linewidth Γ of the indicated hyperfine spectral band in panel (b) to the single-nucleus natural linewidth γ_0 , the linewidth Γ is extracted from the spectral band around $\omega_x \approx 62$.

consequence of the rapid onset of the dynamical diffraction regime. In Figs. 12(b) and 12(c), we examine the absorption lengths L_{abs} of the two forward-transmitted waves inside the crystal. It is shown clearly that the interaction lengths of these electromagnetic modes inside the crystal become significantly extended as Θ_0 approaches 90° , allowing for the multiple scattering of the parametric x-ray photons within the crystal and thus the enhanced nuclear dynamical diffraction effects. In Fig. 12(d), we further plot the ratio of the fitted spectral linewidth Γ of the π polarization ($\xi_B = \pi/4$) hyperfine spectral band to the natural nuclear linewidth γ_0 as a function of Θ_0 . Pronounced superradiant spectral broadenings are observed in the range $89.4^\circ \lesssim \Theta_0 \lesssim 89.8^\circ$, reaching a maximum $\Gamma/\gamma_0 \approx 10$ at $\Theta_0 \approx 89.65^\circ$. However, at lower cutting angles, such as in the grazing EAD geometry, the linewidth remains close to the natural linewidth due to the predominantly Fano-like spectral profile.

VIII. SUMMARY

We have developed a general dynamical diffraction theory that is capable of modeling (S)PXR and (S)PMR in general diffraction geometries, including all possible x-ray emission directions. The analysis includes a broad range of physical effects related to the Mössbauer crystal and the electron beam. In particular, we extended the established two-wave boundary formulation to the four-wave interaction regime, considered a nuclear hyperfine splitting and the resulting polarization

dependence of PMR, discussed new regimes of electron-beam parameters provided by XFELs, extended the optical reciprocity theorem used in the analysis to also include nuclear electromagnetic multipoles in PMR, and analyzed nuclear dynamical diffraction effects.

In a related companion paper [39], we generalize PMR to the superradiant case and utilize this framework to optimize the nuclear-resonant photon production. Interestingly, we find that optimum conditions are not achieved in the previously considered grazing EAD geometry, but rather in the grazing SD geometry opened up by our analysis.

Using our approach, we analyzed in detail the physical mechanism underlying this result. Due to an intricate interplay of the specular diffraction, the depth-dependent absorption, and the effect of the transverse profile of the electron beam, the radiation from electrons moving closely to but outside of the crystal surface may dominate over that of the electrons inside the crystal, unlike in the grazing EAD geometry. The qualitatively different nature of this diffraction geometry is further highlighted by the clear signatures of dynamical diffraction effects in the emitted x-ray radiation. Although we focused the detailed discussion on SPMR, our approach for general diffraction geometries also applies to PXR, SPXR, and PMR. We envision that it will open up further exploration of the grazing SD geometry, also experimentally using state-of-the-art accelerator-based electron sources.

As an alternative Mössbauer radiation source, PMR requires an enhanced photon yield near the Mössbauer resonance to enable coherent nuclear excitation. However, under the Mössbauer resonance condition, the conventional grazing EAD geometry—though favorable for increasing PXR intensity—becomes less effective for PMR. Beyond developing the existing PMR theory, we identified the grazing SD geometry as particularly advantageous for enhancing PMR intensity. When the electron beam propagates slightly above the crystal surface, the virtual photons associated with the incident electron beam undergo specular diffraction at the crystal surface, effectively compensating for the stronger absorption of PMR generated inside the crystal. We have investigated, in the reverse picture, the properties and the attenuation characteristics of the diffracted waves in this extreme diffraction geometry, as well as the effect of transverse modulation caused by the electron-beam transverse distribution, providing a physical understanding for the resulting geometry-dependent intensity enhancement. Furthermore, we have analyzed the characteristics of the near-resonant parametric Mössbauer photons, including their spectral profile, angular distribution, and nuclear dynamical diffraction effects, and have demonstrated that the grazing SD geometry drives PMR into a strong dynamical diffraction regime. This regime is fundamentally distinct from that of the conventional grazing EAD geometry and could provide a novel x-ray quantum optical platform for engineering the coherent interactions between x-ray photons and Mössbauer nuclei in future studies.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: RADIATION FROM A CHARGED PARTICLE MOVING IN A MEDIUM

In this Appendix, we review the electromagnetic radiation problem of a charged particle moving in a medium and present the derivation of the angular-spectral distribution of the emitted photons.

The electric field at the detection point $\mathbf{r}$ generated by a charged particle moving in a medium with an arbitrary dispersion can be solved by the dyadic Green's function, which is defined via

$$\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - k^2 \int d^3r'' \boldsymbol{\epsilon}(\mathbf{r}, \mathbf{r}'', \omega) \cdot \mathbf{G}(\mathbf{r}'', \mathbf{r}', \omega) = \mathbf{I} \delta(\mathbf{r} - \mathbf{r}'), \quad (\text{A1})$$

with $k = \omega/c$. In Eq. (A1), we have considered the general constitutive relation involving the relative dielectric permittivity tensor $\boldsymbol{\epsilon}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{I} \delta(\mathbf{r} - \mathbf{r}') + \boldsymbol{\chi}(\mathbf{r}, \mathbf{r}', \omega)$ defined in the whole space with $\boldsymbol{\chi}(\mathbf{r}, \mathbf{r}', \omega)$ being the electric susceptibility tensor. Recalling the free-space dyadic Green's function $\mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega)$ and considering the usual case of far-field detection, $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ in the far-field zone can be solved from Eq. (A1) as

$$\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \frac{e^{ikr}}{4\pi r} \sum_{s=\sigma, \pi} \mathbf{e}_{ks} \mathbf{E}_{ks}^{(-)*}(\mathbf{r}', \omega), \quad (\text{A2})$$

where $\mathbf{k} = k\mathbf{r}/r$, and $\mathbf{e}_{ks}$ is the s polarization unit vector of mode $\mathbf{k}$. The electric field $\mathbf{E}_{ks}^{(-)}(\mathbf{r}', \omega)$ is related to the solution of the homogeneous Maxwell's wave equation

$$\nabla \times \nabla \times \mathbf{E}_{ks}^{(+)}(\mathbf{r}, \omega) - k^2 \int d^3r' \boldsymbol{\epsilon}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{E}_{ks}^{(+)}(\mathbf{r}', \omega) = 0 \quad (\text{A3})$$

via the optical reciprocity theorem $\mathbf{E}_{-ks}^{(+)} = \mathbf{E}_{ks}^{(-)*}$ [41]. With the dyadic Green's function, the radiation field in the far-field zone is given by

$$\mathbf{E}(\mathbf{r}, \omega) = i\omega\mu_0 \int d^3r' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{J}(\mathbf{r}', \omega), \quad (\text{A4})$$

where $\mathbf{J}(\mathbf{r}, \omega)$ is the temporal Fourier transform of the source current. From the Poynting vector, the radiation energy emitted into the solid angle $d\Omega$ around the detection direction $\mathbf{n} = \mathbf{k}/k$ and in the frequency interval $d\omega$ around the frequency ω is then obtained as

$$\frac{\partial^2 W_{\mathbf{n}, \omega}}{\partial \omega \partial \Omega} = 4\pi \epsilon_0 c r^2 |\mathbf{E}(\mathbf{r}, \omega)|^2. \quad (\text{A5})$$

For the case of a single particle, the angular-spectral distribution of the number of photons with s polarization is found to be [23]

$$\frac{\partial^2 N_{\mathbf{n}, \omega s}}{\partial \omega \partial \Omega} = \frac{q^2 \omega}{16\pi^3 \hbar \epsilon_0 c^3} \left| \int \mathbf{E}_{ks}^{(-)*}(\mathbf{r}(t), \omega) \cdot \mathbf{v}(t) e^{i\omega t} dt \right|^2, \quad (\text{A6})$$

where q is the single-particle charge, and the vectors $\mathbf{r}(t)$ and $\mathbf{v}(t)$ are the trajectory and velocity of the charged particle, respectively. One can see that the electric field $\mathbf{E}_{ks}^{(-)}(\mathbf{r}, \omega)$ can now be replaced via the reciprocity theorem by the solution

$\mathbf{E}_{-ks}^{(+)}(\mathbf{r}, \omega)$ of the scattering problem of a plane wave with the wave vector $-\mathbf{k}$ incident on the source-free medium (reverse picture).

APPENDIX B: EXPRESSIONS FOR NUCLEAR SUSCEPTIBILITIES OF ^{57}Fe CRYSTAL

We first briefly outline how the incident and diffracted waves of the same polarizations are coupled through the crystal susceptibilities. For this, we return to the two-wave coupling equations in their original vector form:

$$(k_{\mu s}^2 - k^2) \mathbf{E}_{\mu s} = k^2 (\boldsymbol{\chi}_0 \cdot \mathbf{E}_{\mu s} + \boldsymbol{\chi}_g \cdot \mathbf{E}_{g\mu s}),$$

$$(k_{g\mu s}^2 - k^2) \mathbf{E}_{g\mu s} = k^2 (\boldsymbol{\chi}_0 \cdot \mathbf{E}_{g\mu s} + \boldsymbol{\chi}_g \cdot \mathbf{E}_{\mu s}), \quad (\text{B1})$$

with $s = \sigma$ or π . In principle, Eq. (B1) can be solved by projecting the electric-field vectors $\mathbf{E}_{\mu s}$, $\mathbf{E}_{g\mu s}$, and the susceptibility tensors $\boldsymbol{\chi}_0$, $\boldsymbol{\chi}_g$ onto any coordinate axes. However, in the polarization Cartesian coordinate system spanned by $(\mathbf{e}_k, \mathbf{e}_\sigma, \mathbf{e}_\pi)$, as shown in Figs. 2(b) and 2(c), only the susceptibility components $\chi_{0,g}^{(\sigma)} = \mathbf{e}_\sigma \cdot \boldsymbol{\chi}_{0,g} \cdot \mathbf{e}_\sigma$ and $\chi_{0,g}^{(\pi)} = \mathbf{e}_\pi \cdot \boldsymbol{\chi}_{0,g} \cdot \mathbf{e}_\pi$ enter, i.e.,

$$(k_{\mu s}^2 - k^2) E_{\mu s} = k^2 [\chi_0^{(s)} E_{\mu s} + c_s \chi_g^{(s)} E_{g\mu s}],$$

$$(k_{g\mu s}^2 - k^2) E_{g\mu s} = k^2 [\chi_0^{(s)} E_{g\mu s} + c_s \chi_g^{(s)} E_{\mu s}], \quad (\text{B2})$$

where $c_\sigma = 1$ and $c_\pi = \cos(2\theta_B)$.

The specific expression for the components of the nuclear susceptibility tensor of ^{57}Fe crystal is given by [2]

$$\chi_n^{ij}(\mathbf{k}, \mathbf{k}', \omega) = \tilde{\chi}_{kk'} \left\{ \mathcal{P}_{-1}^{ij} \left[\frac{1}{4} \mathcal{L}_{-1}^-(\omega) + \frac{1}{12} \mathcal{L}_{-1}^+(\omega) \right] \right. \\ \left. + \mathcal{P}_0^{ij} \left[\frac{1}{6} \mathcal{L}_0^-(\omega) + \frac{1}{6} \mathcal{L}_0^+(\omega) \right] \right. \\ \left. + \mathcal{P}_{+1}^{ij} \left[\frac{1}{12} \mathcal{L}_{+1}^-(\omega) + \frac{1}{4} \mathcal{L}_{+1}^+(\omega) \right] \right\}, \quad (\text{B3})$$

where

$$\tilde{\chi}_{kk'} = 3 \left[-\frac{4\pi c^3}{\omega^3 \Omega_0} \frac{\eta e^{-W_{LM}}}{1 + \alpha_c} S(\mathbf{k}, \mathbf{k}') \right],$$

$$\mathcal{L}_0^\pm(\omega) = \frac{\gamma_0/2}{\omega - (\omega_0 + \delta_0^\pm) + i\gamma_0/2},$$

$$\mathcal{L}_{+1}^\pm(\omega) = \frac{\gamma_0/2}{\omega - (\omega_0 + \delta_{+1}^\pm) + i\gamma_0/2},$$

$$\mathcal{L}_{-1}^\pm(\omega) = \frac{\gamma_0/2}{\omega - (\omega_0 + \delta_{-1}^\pm) + i\gamma_0/2}, \quad (\text{B4})$$

with the transition frequency detunings with respect to the two-level resonance frequency ω_0 ($\hbar\omega_0 = 14.41$ keV):

$$\delta_{-1}^- = -(3/2)\delta_e - (1/2)\delta_g, \quad (\sigma^-),$$

$$\delta_{-1}^+ = -(1/2)\delta_e + (1/2)\delta_g, \quad (\sigma^-),$$

$$\delta_0^- = -(1/2)\delta_e - (1/2)\delta_g, \quad (\pi^0),$$

$$\delta_0^+ = (1/2)\delta_e + (1/2)\delta_g, \quad (\pi^0),$$

$$\delta_{+1}^- = (1/2)\delta_e - (1/2)\delta_g, \quad (\sigma^+),$$

$$\delta_{+1}^+ = (3/2)\delta_e + (1/2)\delta_g, \quad (\sigma^+). \quad (\text{B5})$$

It is easy to check that Eq. (B3) can be simplified to the expression for the nuclear susceptibility of a ^{57}Fe crystal in

the absence of $\mathbf{B}_{\text{hf}}$, which is given by [30]

$$\chi_n^{ij}(\mathbf{k}, \mathbf{k}', \omega) = -\frac{4\pi c^3}{\omega^3 \Omega_0} \frac{\eta e^{-W_{\text{LM}}}}{1 + \alpha_c} S(\mathbf{k}, \mathbf{k}') \mathcal{P}^{ij} \frac{\gamma_0/2}{\omega - \omega_0 + i\gamma_0/2}. \quad (\text{B6})$$

By explicitly evaluating the polarization factors in Eq. (15), one can readily derive the specific expressions for $\chi_n^{(\sigma)}$ and $\chi_n^{(\pi)}$ of a ^{57}Fe crystal in the following two hyperfine geometric configurations:

(1) $\mathbf{B}_{\text{hf}} \parallel (\mathbf{k} \times \mathbf{k}_g)$. The expressions for $\chi_n^{(\sigma)}(\mathbf{k}, \mathbf{k}', \omega)$ and $\chi_n^{(\pi)}(\mathbf{k}, \mathbf{k}', \omega)$ are given by

$$\chi_n^{(\sigma)}(\mathbf{k}, \mathbf{k}', \omega) = \tilde{\chi}_{kk'} \left\{ \frac{1}{2} e^{i\alpha_{kk'}} \left[\frac{1}{12} \mathcal{L}_{+1}^-(\omega) + \frac{1}{4} \mathcal{L}_{+1}^+(\omega) \right] + \frac{1}{2} e^{-i\alpha_{kk'}} \left[\frac{1}{4} \mathcal{L}_{-1}^-(\omega) + \frac{1}{12} \mathcal{L}_{-1}^+(\omega) \right] \right\}, \quad (\text{B7})$$

and

$$\chi_n^{(\pi)}(\mathbf{k}, \mathbf{k}', \omega) = \tilde{\chi}_{kk'} \cos(\alpha_{kk'}) \left[\frac{1}{6} \mathcal{L}_0^+(\omega) + \frac{1}{6} \mathcal{L}_0^-(\omega) \right], \quad (\text{B8})$$

respectively. Here $\alpha_{kk'}$ is the angle between $\mathbf{k}$ and $\mathbf{k}'$, i.e., $\alpha_{kk'} = 0$ for $\mathbf{k}' = \mathbf{k}$ and $\alpha_{kk'} = 2\theta_B$ for $\mathbf{k}' = \mathbf{k}_g$. One can see that, in this geometry, $\sigma^\pm$ Zeeman transitions are coupled with the PMR of σ polarization, and π^0 Zeeman transitions are coupled with the PMR of π polarization. It can be easily checked that, if the hyperfine splitting is absent, both $\chi_n^{(\sigma)}$ and $\chi_n^{(\pi)}$ will be reduced to

$$\chi_n^{(\sigma)}(\mathbf{k}, \mathbf{k}', \omega) = \chi_n^{(\pi)}(\mathbf{k}, \mathbf{k}', \omega) = \tilde{\chi}_{kk'} \cos(\alpha_{kk'}) \frac{\gamma_0/2}{\omega - \omega_0 + i\gamma_0/2}, \quad (\text{B9})$$

which can also be reproduced from Eq. (B3) with $\mathcal{P}^{ij} = (\mathbf{k} \cdot \mathbf{k}')/kk' = \cos(\alpha_{kk'})$. In other words, the parametric radiations of both σ and π polarizations are generated via the same nuclear susceptibility $\chi_n^{(\sigma)} = \chi_n^{(\pi)} = \chi_n$.

(2) $\mathbf{B}_{\text{hf}} \perp (\mathbf{k} \times \mathbf{k}_g)$. The expressions for $\chi_n^{(\sigma)}(\mathbf{k}, \mathbf{k}', \omega)$ and $\chi_n^{(\pi)}(\mathbf{k}, \mathbf{k}', \omega)$ are given by

$$\begin{aligned} \chi_n^{(\sigma)}(\mathbf{k}, \mathbf{k}', \omega) &= \tilde{\chi}_{kk'} \left\{ \frac{1}{2} \cos \xi_B \cos(\xi_B - \alpha_{kk'}) \left[\frac{1}{4} \mathcal{L}_{-1}^-(\omega) + \frac{1}{12} \mathcal{L}_{-1}^+(\omega) \right] \right. \\ &\quad + \sin \xi_B \sin(\xi_B - \alpha_{kk'}) \left[\frac{1}{6} \mathcal{L}_0^+(\omega) + \frac{1}{6} \mathcal{L}_0^-(\omega) \right] \\ &\quad \left. + \frac{1}{2} \cos \xi_B \cos(\xi_B + \alpha_{kk'}) \left[\frac{1}{12} \mathcal{L}_{+1}^-(\omega) + \frac{1}{4} \mathcal{L}_{+1}^+(\omega) \right] \right\}, \end{aligned} \quad (\text{B10})$$

and

$$\begin{aligned} \chi_n^{(\pi)}(\mathbf{k}, \mathbf{k}', \omega) &= \tilde{\chi}_{kk'} \left\{ \frac{1}{2} \cos(\alpha_{kk'}) \left[\frac{1}{12} \mathcal{L}_{+1}^-(\omega) + \frac{1}{4} \mathcal{L}_{+1}^+(\omega) \right] \right. \\ &\quad \left. + \frac{1}{2} \cos(\alpha_{kk'}) \left[\frac{1}{4} \mathcal{L}_{-1}^-(\omega) + \frac{1}{12} \mathcal{L}_{-1}^+(\omega) \right] \right\}, \end{aligned} \quad (\text{B11})$$

respectively. In this hyperfine geometry, the orientation of $\mathbf{B}_{\text{hf}}$ in the diffraction plane is parametrized by the angle ξ_B between $\mathbf{B}_{\text{hf}}$ and the incident wave vector, i.e., the Mössbauer wave vector $\mathbf{k}$. Also $\chi_n^{(\sigma)}(\mathbf{k}, \mathbf{k}', \omega)$ and $\chi_n^{(\pi)}(\mathbf{k}, \mathbf{k}', \omega)$ give the same expression in Eq. (B9) if $\mathbf{B}_{\text{hf}}$ is absent.

APPENDIX C: RECIPROCITY THEOREM WITH NUCLEAR MULTIPOLE

The optical reciprocity theorem plays an important role in x-ray optics with Mössbauer crystals. Since most Mössbauer nuclear transitions correspond to higher-order electromagnetic multipoles, the quantum-mechanical optical reciprocity theorem was generalized to account for the higher multipolarity of Mössbauer sources [42,43,67]. The Born-Wolf optical reciprocity theorem is primarily based on the assumption of electric-dipole ($E1$) scatterers [41]. Therefore, in this Appendix, we generalize the Born-Wolf formalism to incorporate higher electromagnetic multipoles for PMR involving Mössbauer crystals.

The x-ray scattering dynamics with crystals is dominated by the wave equation in terms of the electric field $\mathbf{E}(\mathbf{r}, \omega)$ of the radiation [23]

$$\begin{aligned} \nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) - k^2 \mathbf{D}(\mathbf{r}, \omega) &= 0, \\ \mathbf{D}(\mathbf{r}, \omega) &= \int d^3\mathbf{r}' \boldsymbol{\epsilon}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{E}(\mathbf{r}', \omega). \end{aligned} \quad (\text{C1})$$

Note that the effects of electromagnetic multipoles are included in the effective relative dielectric permittivity tensor $\boldsymbol{\epsilon}(\mathbf{r}, \mathbf{r}', \omega)$ via the relation $\boldsymbol{\chi}(\mathbf{k}, \mathbf{k}', \omega) = \boldsymbol{\chi}_e(\mathbf{k}, \mathbf{k}', \omega) + \boldsymbol{\chi}_n(\mathbf{k}, \mathbf{k}', \omega)$ in the reciprocal space. Specifically, for Mössbauer crystals, it incorporates the electronic and nuclear electric-dipole ($E1$) contributions $\boldsymbol{\chi}_{e,E1}$ and $\boldsymbol{\chi}_{n,E1}$, the nuclear magnetic-dipole ($M1$) contribution $\boldsymbol{\chi}_{n,M1}$, as well as the nuclear electric-quadrupole ($E2$) contribution $\boldsymbol{\chi}_{n,E2}$, specified by the different polarization factors, as shown in Eqs. (11)–(15). While $\boldsymbol{\epsilon}(\mathbf{k}, \mathbf{k}', \omega)$ is a macroscopic electrodynamical parameter, its specific value and frequency—as well as wave vector—dependencies are related to the quantum-mechanical behavior of electrons and nuclei within the crystal. Considering two solutions of Eq. (C1), denoted by the subscripts 1 and 2, we obtain the differential form of the Lorentz reciprocity relation

$$\begin{aligned} \nabla \cdot [\mathbf{E}_1 \times (\nabla \times \mathbf{E}_2) - \mathbf{E}_2 \times (\nabla \times \mathbf{E}_1)] \\ = k^2 (\mathbf{E}_2 \cdot \mathbf{D}_1 - \mathbf{E}_1 \cdot \mathbf{D}_2), \end{aligned} \quad (\text{C2})$$

where all the vectors are in the space-frequency domain $(\mathbf{r}, \omega)$. Integrating both sides over a volume V where the scattering crystal is located and applying Gauss's divergence theorem to the left-hand side, we arrive at the integral form of the Lorentz reciprocity theorem:

$$\begin{aligned} \oint_S (\mathbf{E}_1 \times \mathbf{H}_2 - \mathbf{E}_2 \times \mathbf{H}_1) \cdot d\mathbf{S} \\ = i\omega\epsilon_0 \int_V (\mathbf{E}_1 \cdot \mathbf{D}_2 - \mathbf{E}_2 \cdot \mathbf{D}_1) dV, \end{aligned} \quad (\text{C3})$$

where the integral on the left-hand side is performed over the surface S that encloses the scattering volume V , and this surface is placed in the far-field region in vacuum for which the vacuum constitutive relation of the magnetic field $\mathbf{B} = \mu_0 \mathbf{H}$ is applied. We note here that, compared with the conventional form of the Lorentz reciprocity theorem with both the electric part $\mathbf{E}_1 \cdot \mathbf{D}_2 - \mathbf{E}_2 \cdot \mathbf{D}_1$ and the magnetic part $\mathbf{H}_1 \cdot \mathbf{B}_2 - \mathbf{H}_2 \cdot \mathbf{B}_1$ being integrated over the scattering volume, in Eq. (C3), the effects of magnetic multipoles are

absorbed into the crystal electric susceptibility $\chi(\mathbf{k}, \mathbf{k}', \omega) = \chi_e(\mathbf{k}, \mathbf{k}', \omega) + \chi_n(\mathbf{k}, \mathbf{k}', \omega)$, giving rise to the effective macroscopic electric displacement vector in Eq. (C1). For the simple case of only $E1$ scattering, it is easy to prove the source-free Lorentz reciprocity theorem

$$\oint_S (\mathbf{E}_1 \times \mathbf{H}_2 - \mathbf{E}_2 \times \mathbf{H}_1) \cdot d\mathbf{S} = 0. \quad (\text{C4})$$

Next, let us focus on the case of a Mössbauer crystal with nuclear electromagnetic multipoles. Taking the periodicity of the dielectric permittivity tensor with respect to spatial coordinates into account, the spatial Fourier components of the electric fields and the dielectric permittivity tensor can be written as [23,46]

$$\begin{aligned} \mathbf{E}_l(\mathbf{r}, \omega) &= \int \mathbf{E}_l(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot \mathbf{r}} d^3\mathbf{k}, \\ \boldsymbol{\varepsilon}(\mathbf{r}, \mathbf{r}', \omega) &= \sum_g \int \boldsymbol{\varepsilon}(\mathbf{k}, \mathbf{k}_g, \omega) e^{i\mathbf{k} \cdot \mathbf{r}} e^{-i(\mathbf{k}+\mathbf{g}) \cdot \mathbf{r}'} d^3\mathbf{k}, \end{aligned} \quad (\text{C5})$$

with $\mathbf{k}_g = \mathbf{k} + \mathbf{g}$ and $l \in \{1, 2\}$. From this, we obtain the constitutive relation for the Fourier components with spatial dispersion [23,46]

$$\mathbf{D}_l(\mathbf{k}, \omega) = \sum_g \boldsymbol{\varepsilon}(\mathbf{k}, \mathbf{k}_g, \omega) \cdot \mathbf{E}_l(\mathbf{k}_g, \omega). \quad (\text{C6})$$

Thus, the volume integral on the right-hand side of Eq. (C3) is written as

$$\begin{aligned} &\int [\mathbf{E}_1(\mathbf{r}, \omega) \cdot \mathbf{D}_2(\mathbf{r}, \omega) - \mathbf{E}_2(\mathbf{r}, \omega) \cdot \mathbf{D}_1(\mathbf{r}, \omega)] d^3\mathbf{r} \\ &= \sum_g \int [\mathcal{I}_{12}(\mathbf{k}, \mathbf{k}_g, \omega) - \mathcal{I}_{21}(-\mathbf{k}_g, -\mathbf{k}, \omega)] d^3\mathbf{k}, \end{aligned} \quad (\text{C7})$$

with the Fourier components of the Lorentz reciprocity integral kernels

$$\begin{aligned} \mathcal{I}_{12}(\mathbf{k}, \mathbf{k}_g, \omega) &= \sum_{i,j} E_1^i(-\mathbf{k}, \omega) \varepsilon^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) E_2^j(\mathbf{k}_g, \omega), \\ \mathcal{I}_{21}(-\mathbf{k}_g, -\mathbf{k}, \omega) &= \sum_{i,j} E_2^j(\mathbf{k}_g, \omega) \varepsilon^{ji}(-\mathbf{k}_g, -\mathbf{k}, \omega) \\ &\quad \times E_1^i(-\mathbf{k}, \omega), \end{aligned} \quad (\text{C8})$$

where the superscript i (j) represents the i th (j th) component of the electric-field vectors, and the superscript ij is the index of the component of the dielectric permittivity tensors. These two Lorentz reciprocity integral kernels correspond to two mutually reversed scattering processes represented as the forward diffraction $\mathbf{k} \rightarrow \mathbf{k}_g$ and the backward diffraction $-\mathbf{k}_g \rightarrow -\mathbf{k}$, respectively. Obviously, whether these two scattering processes are reciprocal is determined by the reciprocal symmetry between the dielectric permittivities $\varepsilon^{ij}(\mathbf{k}, \mathbf{k}_g, \omega)$ and $\varepsilon^{ji}(-\mathbf{k}_g, -\mathbf{k}, \omega)$. The spatial Fourier component of the dielectric susceptibility is $\varepsilon^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) = \delta_{ij} \delta_{\mathbf{k}, \mathbf{k}_g} + \chi^{ij}(\mathbf{k}, \mathbf{k}_g, \omega)$ [23], in which the total electric susceptibility of the Mössbauer crystal is split in terms of nuclear

electromagnetic multipoles as

$$\begin{aligned} \chi^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) &= \chi_e^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) + \chi_n^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) \\ &= \chi_{e,E1}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) + \chi_{n,E1}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) \\ &\quad + \chi_{n,M1}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) + \chi_{n,E2}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) + \cdots, \end{aligned} \quad (\text{C9})$$

where the ellipsis represents the contributions of the other higher-order nuclear electromagnetic multipoles. It is obvious that the electric susceptibility tensor of the $E1$ interaction is reciprocally symmetric, i.e.,

$$\chi_{E1}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) = \chi_{E1}^{ji}(-\mathbf{k}_g, -\mathbf{k}, \omega), \quad (\text{C10})$$

due to the simple $E1$ -polarization factor $\mathcal{P}_{E1}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) = \delta_{ij}$, which leads to $\mathcal{I}_{12}(\mathbf{k}, \mathbf{k}_g, \omega) = \mathcal{I}_{21}(-\mathbf{k}_g, -\mathbf{k}, \omega)$ and thus ensures the source-free Lorentz reciprocity theorem (C4). The reciprocal symmetry of the $M1$ and $E2$ susceptibilities $\chi_{M1}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega)$ and $\chi_{E2}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega)$ are determined by their respective polarization factors in Eq. (15). Then one can also easily prove that the reciprocal relations of the $M1$ - and $E2$ -polarization factors $\mathcal{P}_{M1}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) = \mathcal{P}_{M1}^{ji}(-\mathbf{k}_g, -\mathbf{k}, \omega) = [(\mathbf{k} \cdot \mathbf{k}_g) \delta_{ij} - k_{gi} k_{kj}] / k k_g$ and $\mathcal{P}_{E2}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) = \mathcal{P}_{E2}^{ji}(-\mathbf{k}_g, -\mathbf{k}, \omega) = [(\mathbf{k} \cdot \mathbf{k}_g) \delta_{ij} + k_j k_{gi} - 2k_i k_{gj}] / k k_g$ hold with the direction of the hyperfine magnetic field $\mathbf{B}_{\text{hf}}$ (or the quantization axis) being reversed from forward diffraction to backward diffraction, i.e., $\mathbf{B}_{\text{hf}} \rightarrow -\mathbf{B}_{\text{hf}}$ [42,51]. This is due to the fact that, for the reciprocity theorem to hold true in a medium where the conventional time-reversal symmetry is broken, $\chi^{ij}(\mathbf{k}, \mathbf{k}_g, \mathbf{B}_{\text{hf}}) \neq \chi^{ji}(-\mathbf{k}_g, -\mathbf{k}, \mathbf{B}_{\text{hf}})$ (due to the hyperfine magnetic field), all time-odd variables should be reversed in the backward-scattering process [42,51]. Then we can also get the generalized reciprocal symmetry of the electric susceptibilities of higher-order multipolarity:

$$\begin{aligned} \chi_{M1}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) &= \chi_{M1}^{ji}(-\mathbf{k}_g, -\mathbf{k}, \omega), \\ \chi_{E2}^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) &= \chi_{E2}^{ji}(-\mathbf{k}_g, -\mathbf{k}, \omega), \end{aligned} \quad (\text{C11})$$

which also leads to the reciprocal symmetry of the dielectric permittivity $\varepsilon^{ij}(\mathbf{k}, \mathbf{k}_g, \omega) = \varepsilon^{ji}(-\mathbf{k}_g, -\mathbf{k}, \omega)$ [42,43] and thus $\mathcal{I}_{12}(\mathbf{k}, \mathbf{k}_g, \omega) = \mathcal{I}_{21}(-\mathbf{k}_g, -\mathbf{k}, \omega)$. Then the source-free Lorentz reciprocity theorem (C4) also holds for the case of nuclear electromagnetic multipolarity.

As the application of the Lorentz reciprocity theorem for source-free optical scattering problems, the Born-Wolf optical reciprocity theorem in terms of far-field scattering amplitudes can readily be derived from Eq. (C4), in which the nuclear electromagnetic multipole are included. Based on the standard procedure of the derivation of the optical reciprocity theorem from the Lorentz reciprocity theorem in Ref. [68], one can obtain the Born-Wolf formalism of the optical reciprocity theorem in terms of far-field scattering amplitudes of spherical waves. When the Born-Wolf optical reciprocity theorem is applied to the specific scenario of electromagnetic radiation from a charged particle in a medium, it simply relates the time-reversal radiation field $\mathbf{E}_{\mathbf{k}s}^{(-)}$ in the Green's function in Eq. (A2) with a convergent spherical wave to the radiation field $\mathbf{E}_{-\mathbf{k}s}^{(+)}$ of the normal forward source-free scattering problem dominated by Eq. (A3) [23].

APPENDIX D: DERIVATION OF THE QUANTITIES RELATED TO THE DYNAMICAL DIFFRACTION FORMALISM

1. Cherenkov condition

For the nonideal incident wave vector $\mathbf{k}$ in the reverse picture, we employed the small-angle expansion around the ideal negative Mössbauer wave vector $\mathbf{k}_0$ in Eq. (7), in which the detection frequency ω is, in principle, independent of θ_x and θ_y . However, the radiation with maximal intensity is generated around the Bragg condition, for which the Cherenkov condition [23,34,35]

$$1 + \frac{1}{\omega}(\mathbf{k}_g \cdot \mathbf{v}) = 0 \quad (\text{D1})$$

is a necessary condition. This means that, in the vicinity of the maximal detection signal, the frequency is approximately constrained by both the photon angular spread θ and the electron angular spread θ_e . To analyze this further, we derive from Eq. (D1) the Cherenkov frequency constraint condition up to second-order in the correction $\epsilon^{(2)} \sim \theta^2$,

$$\begin{aligned} \omega_0 &\approx \omega[1 + \theta_x + \epsilon^{(2)}], \\ \epsilon^{(2)} &= \frac{\theta_e^2 - \theta^2 v_{z'x} + 2(\theta_e \cdot \theta) - 2\theta_x(\theta_e \cdot \mathbf{e}'_z)}{2(1 + v_{z'x})}, \end{aligned} \quad (\text{D2})$$

with $v_{z'x} = \mathbf{e}'_z \cdot \mathbf{e}_x$. Note that the first-order condition has the same form as in the regular EAD geometry with $\theta_B = 45^\circ$ ($\vartheta_0 = \pi$), i.e., $\omega_0 \approx \omega(1 + \theta_x) \approx (gc/\sqrt{2})(1 + \theta_x)$, as considered in Refs. [23,34,35].

2. Diffraction wave vectors

In Ref. [36], it is assumed that the virtual-photon beam of the XFEL electrons and the parametric real-photon beam have the same magnitude of angular spreads. Within this assumption, the photon angular spread θ , which should in principle be introduced in the wave vector $\mathbf{k}$ in the reverse picture, is used directly in the diffraction wave vector $\mathbf{k}_g$.

If instead we keep the photon angular spread θ in the incident wave vector $\mathbf{k}$, based on the ideal Bragg equation (4) in the reverse picture together with Eq. (7), we can derive the diffraction wave vector $\mathbf{k}_g = \mathbf{k} + \mathbf{g}$ as

$$\mathbf{k}_g = -\frac{\omega_0}{v}\mathbf{e}_x + \frac{\omega}{c}\left(\theta - \frac{\theta^2}{2}\mathbf{e}'_z\right) + \frac{\omega - \omega_0}{c}\mathbf{e}'_z. \quad (\text{D3})$$

After applying the Cherenkov condition (D2) up to the first-order correction to Eq. (D3) and the Mössbauer resonance condition $\omega = \omega_0$, we obtain the Cherenkov deviation parameter

$$\begin{aligned} q_c &= k_{g,x}v + \omega_0 = \omega_0\theta_x v_{x'x}, \\ v_{x'x} &= \mathbf{e}'_x \cdot \mathbf{e}_x = \sqrt{1 - \left(1 - \frac{g^2}{2k_0^2}\right)^2}. \end{aligned} \quad (\text{D4})$$

Note that for the regular grazing EAD geometry with $\theta_0 = 45^\circ$, we have $g = \sqrt{2}k_0$ and thus $v_{x'x} = 1$. Due to the fact that both the wave vector $\mathbf{k}_{g\mu s} = \mathbf{k}_g - k\epsilon_{\mu s}\mathbf{N}$ of the diffracted wave inside the crystal and the wave vector $\mathbf{k}_{sp}$ of the specularly reflected diffracted wave outside the crystal only differ

from $\mathbf{k}_g$ by their z components, it is straightforward to write

$$\begin{aligned} \mathbf{k}_{g\mu s} &= -\frac{\omega}{v}\mathbf{e}_x + \frac{\omega}{c}\theta_{g,y}\mathbf{e}_y + \frac{\omega}{c}(\theta_{g,z} - \epsilon_{\mu s})\mathbf{e}_z, \\ \mathbf{k}_{sp} &= -\frac{\omega}{v}\mathbf{e}_x + \frac{\omega}{c}\theta_{g,y}\mathbf{e}_y + i\frac{\omega}{c}\left(\sqrt{\gamma^{-2} + \theta_{g,y}^2}\right)\mathbf{e}_z, \end{aligned} \quad (\text{D5})$$

with the angular spreads of $\mathbf{k}_g$,

$$\begin{aligned} \theta_{g,y} &= \frac{-(\sin \vartheta_0 \sin \varphi_0)\theta_x - (\cos \vartheta_0)\theta_y}{\sqrt{\cos^2 \vartheta_0 + \sin^2 \vartheta_0 \sin^2 \varphi_0}}, \\ \theta_{g,z} &= \frac{(\sin \vartheta_0 \sin \varphi_0)\theta_y - (\cos \vartheta_0)\theta_x}{\sqrt{\cos^2 \vartheta_0 + \sin^2 \vartheta_0 \sin^2 \varphi_0}}. \end{aligned} \quad (\text{D6})$$

The evanescence rate in vacuum can be defined from Eq. (D5) as $\eta = (\gamma^{-2} + \theta_{g,y}^2)^{1/2} = -iv_g^{(\text{sp})}$. For the regular grazing EAD-PXR case discussed in Refs. [34,36,37], $\vartheta_0 = \pi$, then we reproduce the relations $\theta_{g,y} = \theta_y$ and $\theta_{g,z} = \theta_x$. However, for more general geometries, these relations no longer hold. Nevertheless, from Eq. (D6), it is easy to verify that the relation $\theta_{g,y}^2 + \theta_{g,z}^2 = \theta_g^2 = \theta^2$ also holds in the general case. Thus, the virtual-photon beam and the real-photon beam have the same magnitude of the angular spreads, as used in Ref. [36].

3. Polarization factors

The polarization factors $\mathbf{e}_s \cdot \mathbf{e}_v$ with $s = \sigma$ and π also need to be generalized for SPMR beyond the grazing EAD case. They are dependent on the photon angular variables θ_x and θ_y . From their definitions [23,30,36]

$$\mathbf{e}_\sigma = \frac{\mathbf{k} \times \mathbf{g}}{|\mathbf{k} \times \mathbf{g}|}, \quad \mathbf{e}_\pi = \frac{\mathbf{k}_g \times \mathbf{e}_\sigma}{|\mathbf{k}_g \times \mathbf{e}_\sigma|}, \quad (\text{D7})$$

one can see that the σ -polarization vector is perpendicular to the diffraction plane spanned by $\mathbf{k}$ (or $\mathbf{v}$) and $\mathbf{g}$. This means that there is no contribution to the polarization factor $\mathbf{e}_\sigma \cdot \mathbf{e}_v$ from the second-order correction $(\theta^2/2)\mathbf{e}'_z$ of $\mathbf{k}$ due to $(\mathbf{e}'_z \times \mathbf{g}) \cdot \mathbf{e}_v = 0$. Thus, the wave vector $\mathbf{k}$ only needs to be considered up to the first-order correction θ , and we obtain for the unit vectors of $\mathbf{k}$, $\mathbf{g}$, and $\mathbf{v}$:

$$\begin{aligned} \mathbf{e}_k &= \frac{\mathbf{k}}{k} \approx \mathbf{e}'_z + \theta, \quad \mathbf{e}_v = \frac{\mathbf{v}}{v} \approx \mathbf{e}_x, \\ \mathbf{e}_g &= \frac{\mathbf{g}}{g} \approx -\frac{\omega_0}{gc}(\mathbf{e}'_z + \mathbf{e}_x). \end{aligned} \quad (\text{D8})$$

Here, we ignored the angular spread of the XFEL electrons $\theta_e \sim \sigma_a$ compared with the SPXR photon angular spread θ [36]. From the definition (D7) and after some cumbersome but straightforward derivations, especially with the help of the Cherenkov frequency-constraint condition (D2) for $\mathbf{k}_g$, the polarization factors turn out to be

$$\mathbf{e}_\sigma \cdot \mathbf{e}_v \approx \theta_y, \quad \mathbf{e}_\pi \cdot \mathbf{e}_v \approx -\theta_x, \quad (\text{D9})$$

which has the same form as in the grazing EAD geometry.

4. Bragg deviation parameter

The Bragg deviation parameter α_B also needs to be generalized to the SPMR diffraction geometry. Substituting the explicit form of $\mathbf{k}_g$ in Eq. (D3) into the definition of

$\alpha_B = -(\mathbf{k}_g^2 - \mathbf{k}^2)/k^2$, we obtain the following exact form:

$$\begin{aligned} \alpha_B = & 2(1 + v_{z'x})\left(\frac{\omega_0}{\omega}\right) + 2\theta_x\left(\frac{\omega_0}{\omega}\right) + v_{z'x}\gamma^{-2}\left(\frac{\omega_0}{\omega}\right) \\ & - \theta^2(1 + v_{z'x})\left(\frac{\omega_0}{\omega}\right) - 2(1 + v_{z'x})\left(\frac{\omega_0}{\omega}\right)^2 \\ & - \gamma^{-2}(1 + v_{z'x})\left(\frac{\omega_0}{\omega}\right)^2. \end{aligned} \quad (\text{D10})$$

Considering that the minimal value of $|\alpha_B|$ is of order γ^{-2} , we keep all the terms in Eq. (D10) up to the second-order corrections, which are of the order of γ^{-2} . This means that the orders of correction to which the frequency relations ω_0/ω and $(\omega_0/\omega)^2$ need to be calculated depend on their corresponding prefactors. Applying the Cherenkov frequency constraint condition (D2) to Eq. (D10), we obtain the final form of α_B

$$\alpha_B \approx -[\gamma^{-2} + \theta^2 + \theta_e^2 + 2(\boldsymbol{\theta}_e \cdot \boldsymbol{\theta}) - 2\theta_x(\boldsymbol{\theta}_e \cdot \mathbf{e}'_z)]. \quad (\text{D11})$$

For the regular grazing EAD geometry, we have $\boldsymbol{\theta}_e \cdot \boldsymbol{\theta} = \theta_y\theta_{ey}$ and $\boldsymbol{\theta}_e \cdot \mathbf{e}'_z = -\theta_{ez}$. Then, Eq. (D11) is simplified to [23,34–36]

$$\alpha_B = -[\gamma^{-2} + (\theta_y + \theta_{ey})^2 + (\theta_x + \theta_{ez})^2]. \quad (\text{D12})$$

If we ignore the terms with $\boldsymbol{\theta}_e$ due to $\sigma_a \ll \theta$, Eq. (D11) reduces to $\alpha_B \approx -(\gamma^{-2} + \theta^2)$ [36]. One can see that the simplified form of α_B is still unchanged in the transition from the regular grazing EAD geometry for PXR problem to the SPMR geometry. This is due to the fact that the magnitudes of the angular spreads of $\mathbf{k}_g$ and $\mathbf{k}$ remain the same, i.e., $\theta_g^2 = \theta^2$, as shown in Eq. (D6).

APPENDIX E: NONIDEAL CONDITIONS

The generation of SPMR is inevitably influenced by nonideal conditions, including crystal imperfections, finite

electron-beam quality, and deviations from the ideal parallel grazing-incidence geometry. High-quality crystals are required, characterized by low mosaicity, high isotopic enrichment, low temperature, and clean, flat surfaces. The angular alignment between the electron beam and the crystal surface is another critical factor. While the SPMR intensity exhibits a relatively broad tolerance to the variation of the crystal cutting angle (several tens of milliradians) in the grazing SD geometry, it is far more sensitive to deviations from beam-surface parallelism. The misalignment can lead to a significant reduction in the radiation intensity due to detuning from the Bragg condition. If the electron beam deviates slightly from the ideal parallel incidence by a small angle θ_p , the Bragg deviation parameter α_B , which is associated with the amplitude of the diffracted field via Eq. (20), can be estimated as $\alpha_B \approx 2\theta_p \sin(2\theta_B)$ [56]. When $|\alpha_B| \sim |\chi_{0(g)}|$, the intensity of diffracted wave begins to decrease appreciably. This condition gives an estimate for the order of magnitude of the allowable angular deviation of the electron beam, $|\theta_p| \sim |\chi_{0(g)}|/\sin(2\theta_B) \sim 10^{-5}$ rad for the (S)PMR sources based on the α -iron crystal. Therefore, even a misalignment of only several tens of microradians in the electron-beam parallelism under the grazing-incidence geometry can lead to a substantial reduction of the radiation intensity due to Bragg detuning.

In addition, the energy spread of electrons should also be sufficiently small for (S)PMR sources. For an SPMR source using typical XFEL electrons, an energy spread of $\Delta E_e = (\gamma^2 \Delta v/v)E_e \approx \text{MeV}$ [36] induces a virtual-photon cone shift $\Delta\theta_\gamma \sim \gamma\sigma_a^2 \approx 10^{-6}$ to 10^{-9} rad, which is negligible compared with the intrinsic angular divergence of the electron beam (microradians) and thus has an insignificant effect on SPMR excitation. For the incoherent continuous-wave electron beam employed for the PMR source in Ref. [30], the shift of a virtual-photon cone would be $\Delta\theta_\gamma \approx 10^{-3}$ to 10^{-5} rad, which can be comparable to the corresponding electron-beam angular spread. In this case, the PMR radiation pattern becomes energy-spread dominated.

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