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Free-electron-driven X-ray caustics from strained van der Waals materials

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Abstract: Tunable nanoscale shaping of X-ray waves remains as an open challenge of critical importance for applications in high-resolution X-ray spectroscopy and imaging. In contrast to this high photon energy regime, shaping of light waves in the visible and infrared regimes is routinely undertaken in a vast range of applications by means of abundantly available optical components. However, analogous optical elements are scarcer for X-rays. Here, we propose a new paradigm based on van der Waals (vdW) materials for shaping X-ray waves directly at the source. By inducing strain and bending on vdW materials, we control their interaction with free electrons in a manner that tunes the emission of X-rays and forms caustic X-ray beams. The geometry of the vdW material alters the caustic beam properties, including its focal length, spot size, and diffraction-free length. The ability to integrate X-ray shaping into the electron-driven emission process bypasses the efficiency limits of current optical techniques in this spectral regime. Looking forward, by shaping X-ray wave interference at the atomic scale we open new horizons in high-resolution X-ray science.

Introduction

Van der Waals (vdW) structures, which are composed of multilayered two-dimensional (2D) materials held together by weak vdW forces, are often subject to various kinds of out-of-plane deformations such as scrolls¹, folds^{2,3}, bubbles^{4–8}, ripples^{9–12}, buckles^{13,14}, crumples¹⁵, tents^{8,16}, etc. These deformations and their physical effects have received a recent surge of interest in topics ranging from precision strain control^{11,17,18} to strain-induced novel phenomena^{19–22}. Strain engineering has emerged as a promising means of manipulating and enhancing light emission from transition metal dichalcogenides (TMDs) and transition metal thiophosphates²³. However, the use of strain engineering in light emission has thus far been confined to the optical regime ($\sim$ eV photon energies), restricted by the optical bandgap of vdW materials²⁴.

These limitations raise the question of whether strain engineering can be used to shape the emitted light at high photon energies including X-rays. The question is especially pertinent given the challenge in shaping X-rays by using traditional optical elements such as mirrors and lenses, which are generally lossy and inefficient at high photon energies. Furthermore, many shaping methods that are commonly employed at visible wavelengths (e.g., phase masks with subwavelength features) become impractical for X-rays. As a result, applications of shaped optical beams are absent from the X-ray regime. An example of potential interest is the Airy beam^{25,26}, a non-diffracting form of light propagation that often manifests itself with a curved trajectory and asymmetric extended side-lobes²⁷. The unique properties of these types of beams have attracted great interest from both fundamental^{27–30} and applied^{31,32} perspectives. An intriguing application of Airy beams is high-contrast, large-field-of-view (FOV) light-sheet imaging^{33–35}, which benefits from the narrow intensity distribution and relatively long non-diffracting axial extent of such beams. Looking at the bigger picture, the Airy beam is just one example of caustics, which are universal phenomena emerging in any wave system in nature. Actually, the concept of caustics had huge impact on the wider field of optics over the past decades^{36,37}. Nevertheless, the lack of optical components in the X-ray regime has so far limited experimental realizations^{38,39} of X-ray caustics and their applications.

In this paper, we propose strain engineering of vdW materials as an effective means of shaping X-ray emission. Specifically, we find that X-rays produced by free electrons interacting with vdW materials can be shaped into caustics by bending the material itself, producing an out-of-plane

strain that we use to realize Airy beams. Importantly, vdW materials can be bent to large curvatures beyond the capability of traditional crystals through different methods, for example by coaxially stacking vdW nanotubes⁴⁰ and by rolling up 2D layers⁴¹. Furthermore, we show that X-ray Airy beams can be tuned in focal length, diffraction-free range, and wavelength by varying the vdW material strain geometry and the electron energy. Our study also reveals that the formation of Airy beams is limited by the quantum uncertainty of the electron itself. Specifically, the coherent momentum range of the incident electron alters the caustic and provides an additional degree of freedom for controlling it by electron wavepacket shaping^{42–47}. Our results pave the way to the development of versatile, compact X-ray sources capable of directly shaping the emitted X-ray photons, mitigating the need for subsequent optical components. The realization of an X-ray Airy beam could be useful in high-resolution imaging and material characterization applications (e.g., nano-ARPES), exploiting the generation of light sheets that can probe selected slices of a target specimen.

Free-electron-driven X-ray emission from vdW materials can arise from two coherent processes: parametric X-ray radiation (PXR) and coherent bremsstrahlung (CBS). vdW materials were demonstrated as controllable platforms to study these phenomena, serving as structures for the generation of relatively monochromatic, high-brightness X-rays, whose frequency can be tuned by controlling the electron energy and material design⁴⁸. Our concept of creating X-ray caustics applies to both mechanisms because the X-ray emission in PXR and CBS follows the same dispersion relation. In particular, PXR⁴⁹ is emitted from polarization currents induced by the incident electrons on the bound electrons of the material. In contrast, CBS⁴⁹ is emitted from the wiggling of the incident-electron trajectories as a result of a periodic series of bremsstrahlung interactions with the nuclei and bound electrons in the material, where each interaction produces some acceleration of an incident electron and causes it to emit radiation. The contribution of PXR to the radiation peak is comparable to that from CBS for slightly relativistic electrons⁵⁰, and usually dominant for ultrarelativistic electrons^{49,51,52}. Consequently, we focus on the PXR process to demonstrate the concept in our simulations below, but the conclusions apply to both emission mechanisms.

We use WSe₂ as the example of platform to study PXR induced by free electrons interacting with strained vdW materials. PXR can be modeled by treating the periodic atomic structure as a

discrete dipole array, with bound electrons around each atom being encapsulated in an effective dipole quantified through an associated X-ray atomic polarizability^{52,53}. We consider a cylindrically bent multilayer vdW material structure produced by out-of-plane strain^{17,18} as shown in Fig. 1b, with the cylinder axis along the y direction.

Figure 1 | Illustration of an Airy beam created from a cylindrically bent multilayer structure.

We use ray optics to compare the monochromatic X-ray emission produced by a free electron passing through flat (a) and cylindrically bent (b) multilayer structures. The emission direction is tunable as a function of the electron incidence angle θ and velocity $\mathbf{v}$, as well as the orientation of the crystal reciprocal lattice vectors ($g_{\parallel}$, $g_{\perp}$) and film bending radius. A collimated X-ray beam can be excited from a flat multilayer structure, as shown in (a). In comparison, when the structure is bent, the electron incidence angle θ varies along the electron trajectory, so the rays emerging from different atomic layers are not parallel and may converge along a caustic line, generating an X-ray Airy beam, as shown in (b). The figures are not to scale.

We consider two crystal deformation mechanisms associated with bending in vdW materials: elastic and plastic bendings^{17,18,54}. In the elastic bending regime, vdW multilayers are perfectly

glued together and bend as a single plate, with a behavior that follows continuum mechanics plate theory. The latter is characterized by a length-invariant neutral surface, so that crystal layers are extended or compressed along the direction normal to such surface in proportion to the distance to it⁴¹. In the plastic bending mechanism, multilayer interfaces are ultralubricated, with each layer bending independently while locally maintaining in-plane atomic distances and interlayer separations nearly intact by shearing and slipping^{17,18}. Recent experimental results show that bending of vdW materials involves both types of mechanisms, although with a dominant contribution of the second one because the multilayers are held together by relatively weak vdW forces^{17,18}. We therefore assume plastic bending and consider invariant lattice constants. Incidentally, no significant differences are found when considering instead elastic bending with in-plane extension and compression taken into consideration (Supplementary section 5).

Results

When an electron is normally impinging on the WSe₂ multilayer structure shown in Fig. 1a, the emitted photons interfere coherently following dispersion relations that are determined by the reciprocal lattice vectors of the crystal^{48,50},

$$\frac{\omega}{v} = \frac{\omega}{c} \cos \varphi + g_{\perp} \cos \theta - g_{\parallel} \sin \theta, \quad (1)$$

where ω is the angular photon frequency; c and v are the free-space light and electron velocities, respectively; $g_{\perp}$ and $g_{\parallel}$ are the out-of-plane and in-plane components of the crystal reciprocal lattice vector, respectively; φ and θ are the photon emission and the electron incidence angles, respectively. A monochromatic collimated X-ray beam is then generated from the flat multilayer structure, as shown in Fig. 1a, where some of the elements of Eq. (1) are indicated. Therefore, the collimated X-ray beam can be tuned by varying the emission angle, crystal lattice constant, electron velocity, and electron incidence angle⁴⁸.

The X-ray beam can be further manipulated by gradually varying the angle θ between the electron and the stacked crystal layers, which can be achieved by using a curved multilayer structure. We consider a cylindrically bent multilayer structure as shown in Fig. 1b to demonstrate the paradigm. This approach can be easily extended to other curved structures provided that the curvature of each layer can be carefully defined. The incidence angle θ in Fig. 1b decreases as the

radius of curvature increases along the electron trajectory in the z direction. Therefore, photons scattered by certain reciprocal lattice vectors $(g_{\parallel}, g_{\perp})$, such as $g_{\parallel}=0$ with $g_{\perp}>0$, would converge. A full-wave simulation identifies that X-ray caustics are created at the convergence area. The most striking feature of the caustic is highlighted by the dashed blue line in Fig. 1b, marking an abrupt change in intensity known as a “catastrophe”, specifically a “fold catastrophe”²⁹ in the present instance. The optical fold catastrophe is always accompanied by an Airy beam pattern around the caustic line, which features a curved profile and asymmetric extended side-lobes.

To exemplify the general concept in Fig. 1, we apply it to the PXR excited by a 1 MeV free electron passing through a WSe₂ multilayer structure of thickness $T=300$ nm. The radiation pattern can be obtained by adding together the electric field scattered by each layer. Figure 2 compares the X-ray intensity pattern from a flat (a) and a bent (b) WSe₂ multilayer structure in the emission angle region $\varphi=80^{\circ}$ - 93° at distances to the middle point in the range of intersection between the electron and the atomic crystal layers in the $\rho=3.5$ - 6.0 μm range. In Fig. 2a, we set the electron incidence angle to $\theta=\sin^{-1}(4/5)$ to compare with the configuration portrayed in Fig. 2b, where the electron passes through a bent crystal. The collimated X-ray beam propagates at an angle $\varphi=86.6^{\circ}$, matching the X-ray dispersion in Eq. (1). In Fig. 2b, we set the crystal bending radius to $R=5$ μm and the electron trajectory displacement relative to the bending axis to $L=4$ μm . The bending radii of the layers lie in the $R-T/2$ to $R+T/2$ range. An Airy beam with spot size ~ 10 nm is generated at a distance $\rho \approx 4.8$ μm . We build the ρ - s frames by rotating the x - z frames anticlockwise by angles of 3.4° (a) and 6.2° (b), so that ρ and s represent the axial and transverse directions of the collimated X-ray beam, respectively. The electric field profiles are plotted in the transverse cross-section planes (y - s) and the longitudinal plane (ρ - s). The latter is not to scale to make the Airy beam profile clearer. Detailed calculation procedures are given in the Methods section.

a Collimated parametric X-ray beam

b Caustic parametric X-ray beam

Figure 2 | Caustic X-ray beam from a bent multilayer WSe₂ heterostructure. We compare the X-ray intensity profiles of a caustic X-ray beam in panel (b) and a collimated X-ray beam in panel (a). The X-ray emission from the bent WSe₂ heterostructure in (b) forms an Airy beam with a spot size of ~ 10 nm that remains diffraction-free over a propagation length of ~ 1 μm ($\rho=4.3$ - 5.3 μm). The ρ - s axes denote the rotated frame from the x - z axes by angles of 3.4° (a) and 6.2° (b), with the ρ axis pointing along the emission direction. The radius of curvature of the WSe₂ heterostructure is $R=5$ μm . The axis of the bending cylinder is shifted by $L=4$ μm from the electron trajectory. Panels (a) and (b) share the same color scale, sample thickness (300 nm), electron kinetic energy (1 MeV), and photon energy (3 keV).

We now describe the radiation produced by electrons traversing the bent structure. Each bent layer is approximated by a flat layer whose position, normal vector, and 2D crystal structure are determined by those at the point where the electron intersects it. This model is accurate under the conditions here considered, in which the evanescent field carried by the electron decays within a small distance compared with the bending radius. We then treat the contribution of each layer to the emission through the dispersion relation (Supplementary section 2)

$$\frac{\omega}{v} = \frac{\omega}{c} \cos \varphi + g_{\perp} \cos \theta - g_{\parallel} \sin \theta + g_{\parallel} \theta \cos \theta, \quad (2)$$

modified from Eq. (1) by the additional term $g_{\parallel} \theta \cos \theta$ to capture the relative layer shifts. In particular, the electron incidence angle varies in the range $\theta = \sin^{-1}(L/(R - T/2)) - \sin^{-1}(L/(R+T/2))$ in Fig. 2b. The numerical results show that photons scattered by the reciprocal lattice vector ($g_{\parallel}=0, g_{\perp}=5 \times 2\pi/d$), where $d=12.98 \text{ \AA}$ is the interlayer distance of the {001} planes in bulk WSe₂, are mainly responsible for the formation of an Airy beam (Fig. 2b). The central point of such beam subtends an angle $\varphi=83.8^{\circ}$ with respect to the electron trajectory. The bottom inset of Fig. 2b shows an enlarged image of the Airy beam, which remains diffraction-free over the $\rho=4.3\text{-}5.3 \text{ }\mu\text{m}$ range (see next paragraph and Supplementary section 3). The top insets depict the electric field intensity profile across several transverse planes, with the extremal values $\rho=4.2 \text{ }\mu\text{m}$ and $5.4 \text{ }\mu\text{m}$ chosen outside the Airy beam diffraction-free region. The Airy beam width determined from Fig. 2b ($\sim 10 \text{ nm}$) is much smaller than the width ($\sim 0.5 \text{ }\mu\text{m}$) of the collimated X-ray beam in Fig. 2a.

The trajectory equation of the Airy beam (i.e., the envelope equation) can be obtained by combining Eq. (2) and its derivative with respect to θ (see detailed derivation in Supplementary section 3). The axial distribution of the caustic profile yields the following equation under the condition $T \ll R$ (small multilayer thickness compared with the bending radius):

$$\rho(r_c) = \frac{\omega}{c g_{\perp}} \frac{R^3}{L^2} \sin^2 \varphi_0 + 3 \left(-\frac{\cos \varphi_0}{\cos \theta_0} - \frac{\omega}{c g_{\perp}} \frac{\sin^2 \varphi_0}{\sin^2 \theta_0} \right) r_c, \quad (3)$$

where $\cos \varphi_0 = \frac{1}{\beta} + \frac{g_{\perp}}{k} \cos \theta_0$, $\sin \theta_0 = \frac{L}{R}$, and $r_c = -\frac{T}{2} - \frac{T}{2}$ is the radial distance of the different layers from the central layer. The first term, $\frac{\omega}{c g_{\perp}} \frac{R^3}{L^2} \sin^2 \varphi_0$, can be interpreted as the focal length, where the center of the Airy beam caustics appears relative to the origin. In Fig. 3b, colored solid

curves represent the focal length, shown as a function of bending radius R and the L/R ratio. The second term in Eq. (3) is the axial distribution of the Airy beam. Colored dashed curves in Fig. 3b represent the axial caustic length, which scale linearly with the thickness T under the assumed condition $T \ll R$. The results in Fig. 3b agree with the ray optics analysis and show that the Airy beam displays a longer axial spatial extent and is farther from the source with a smaller L/R ratio.

Figure 3 | Analysis of the generated X-ray Airy beam: quantifying its focal length, axial caustic length, and conditions for its formation. Panel (a) defines the focal length and the axial caustic length. The image is not to scale, as the focal length is much larger than the axial caustic length. Panel (b) presents the focal length (left vertical axis) and axial caustic length (right vertical axis) as a function of the L/R ratio (see Fig. 2). The axial caustic length scales linearly with the thickness T (colored dashed curves), while the focal length changes when varying the bending radius R (colored solid curves), under the condition $T \ll R$. Panel (c) presents the regime in which an X-ray caustic can develop. Its formation is limited by wave diffraction, which sets a finite (nonzero) focal depth. The latter must be smaller than the focal length for a meaningful caustic to emerge. The ratio of focal depth to focal length is shown as the vertical axis. The regime in which caustics can (cannot) form is highlighted by a blue (red) background. The ratio is a function of bending radius R , thickness T , and L/R ratio. The electron kinetic energy is 1 MeV and the photon energy is 3 keV.

Optical diffraction can however produce a degradation in the formation of the Airy beam that is not captured by ray optics analyses. To study such effect, we adopt optical diffraction theory for lenses (the Airy beam can be treated as lensing with aberrations) and analyze the Airy beam that it predicts. The focal depth⁵⁵, defined as the axial extent of the focal region, scales inversely proportional to the numerical aperture (NA) squared. We find the expression $\frac{2\lambda f^2}{T^2 \sin^2 \varphi_0}$ for the focal depth, where λ is the wavelength, f is the focal length (first term in Eq. (3)), and $T \sin \varphi_0$ is the effective length of the source. To have a distinct focal spot, the axial extent should be much smaller

than the focal length (i.e., $\frac{2\lambda f^2}{\sin^2 \varphi_0 T^2} / f \ll 1$), as otherwise the focal region extends all the way from the source to the focal spot, so that the latter is completely blurred and indistinguishable from the background. Indeed, the ratio between the focal depth and the focal length, plotted in Fig. 3c, shows that both a larger L/R ratio and a larger interaction length favor a distinct focal spot for the Airy beam. The optimal choice of L/R for a given application therefore involves a trade-off between a longer axial extent (favored by a smaller L/R ratio) and a more distinct focal spot (favored by a larger L/R ratio).

An intriguing application of the Airy beam is light-sheet imaging. Two key parameters in light-sheet microscopy are the field of view and the resolution, which are connected to the diffraction-free range and beam width of the Airy beam. The diffraction-free range can be approximated by the axial caustic length, as plotted in Fig. 3b, which is a function of L/R and the thickness T . We compare in Fig. 4 the Airy beams produced by choosing different shifts of the electron trajectory, $L=4\text{ }\mu\text{m}$ (Fig. 4a) and $2.5\text{ }\mu\text{m}$ (Fig. 4b), while keeping other parameters the same as in Fig. 2b. The ρ - s axes denote the anticlockwise rotated frame from the x - z axes by angles of 6.2° (a) and 18.4° (b), with the ρ axis oriented along the emission direction. Figure 4a shows a smaller Airy beam spot size, coming at the price of a shorter axial caustic length ($\sim 1\text{ }\mu\text{m}$, Fig. 3b). In contrast, Fig. 4b shows a bigger Airy beam spot size, but achieving a longer axial caustic length ($\sim 2\text{ }\mu\text{m}$, Fig. 3b).

Figure 4 | Tunability of X-ray caustics and their prospects for light-sheet applications: comparing two X-ray caustics with different parameters. Panel (a) shows the caustic generated

with stronger crystal bending ($L=4.0\ \mu\text{m}$), which creates a smaller Airy beam spot size, coming at the price of a shorter axial caustic length. Panel (b) shows a caustic produced with weaker crystal bending ($L=2.5\ \mu\text{m}$), which results in a larger Airy beam spot size, achieving a longer axial caustic length. These examples emphasize the potential of using X-ray caustics for light-sheet X-ray microscopy. The color scales in (a) and (b) are normalized to their own maximum values. Other parameters are the same as in Fig. 2b.

To demonstrate the robustness of the diffraction-free attribute of the Airy beam, we compare in Fig. 5b the Airy beam width for incident electron beams (e-beams) with different spot sizes and divergence angles. We choose three different locations, $\rho=10.5\ \mu\text{m}$, $11.3\ \mu\text{m}$, and $12.1\ \mu\text{m}$, in Fig. 4b, and plot the transverse intensity profiles in Fig. 5b. The e-beam follows a Gaussian electron density distribution with root-mean-squared (rms) divergence angle $\delta\theta$ and rms spot size δr . The left, middle, and right columns are results obtained from an ideal point electron, an e-beam with divergence angle $\delta\theta$, and an e-beam with spot size δr , respectively. The comparison shows that the width of the Airy beams can be broadened but maintains its basic features within the caustic region.

Figure 5 | The effect of the electron beam parameters on the quality of the generated X-ray caustics. We test the robustness of the X-ray caustic concept by studying its dependence on various (Gaussian) electron-beam parameters (divergence angle $\delta\theta$ and spot size δr), defined in panel (a) from the root-mean-square (rms) divergence angle and spot size, respectively. Panel (b) compares the Airy beam width at three caustic locations (see color labels). The left column shows the intensity profile from an ideal point electron (zero divergence angle and spot size). The middle (right) column shows the effect of a finite divergence angle (spot size). Panel (c) compares the width of the X-ray Airy beam (solid curves) and the collimated X-ray beam (dashed curves) at $\rho=11.3\ \mu\text{m}$. The left column shows results from an ideal point electron. The setup is the same as in Fig. 2, except that the electron incidence angle is $\theta=\sin^{-1}(1/2)$ in the flat multilayer structure, and the electron trajectory shift is $L=2.5\ \mu\text{m}$ in the bent multilayer structure. The s axis is defined by rotating the x axis clockwise by angles of 23.1° and 18.4° for the Airy beams and the collimated beams, respectively.

We then compare the beam widths for the collimated X-ray beam and the light-sheet X-ray beam by plotting the transverse intensity profiles in Fig. 5c. The left, middle, and right columns show results obtained from an ideal point electron, an e-beam with divergence angle $\delta\theta$, and an e-beam with spot size δr , respectively. These plots show that the light-sheet X-ray beam still has smaller beam width even when created by e-beams with finite divergence angles and spot sizes.

Discussion

To better understand the limitations on electron radiation introduced by the quantum nature of the electrons and photons under consideration, it is beneficial to compare our mechanisms of X-ray radiation to the simpler scenario of Cherenkov radiation (CR). There, conservation of momentum causes the free electron to recoil when emitting a photon. Consequently, the coherence of CR and its ability to form a short “shock wave” are bounded by the momentum uncertainty of the incident electrons⁴⁶. In contrast, conservation of momentum in PXR and CBS involves both the crystal and the electron⁴⁹, so part of the recoil is absorbed by the crystal and not only by the electron. Therefore, the coherence of the X-ray emission is bounded by the combination of uncertainties in the electron coherent momentum and in the crystal reciprocal lattice vectors. Figure 6a shows that the gradual change in the crystal reciprocal lattice vectors helps maintain the interference required to create the caustic almost independently of the coherence of the longitudinal momentum of the incident electron. Figure 6b presents the estimated coherence of the electron transverse momentum that is required for the formation of the X-ray caustics. Electron transverse momentum in the keV/ c range can be obtained by focusing the electron beam to a small spot, which can reach sub-nanometer sizes in electron microscopes⁵⁶.

In a complementary picture, a first-principles, rigorous quantum-mechanical treatment of any excitation (including X-ray photons) generated by an electron beam with small incident-divergence and transmitted-scattering angles shows that the result is fully equivalent to the coherent excitation produced by a line dipole (with wave vector ω/v), averaged incoherently (i.e., summing intensities) over the transverse beam directions, with position-dependent weights taken from the electron intensity distribution across the e-beam⁴⁴. In the point-electron limit, only one such line dipole is present, thereby producing maximum coherence in the emission. As the lateral size of the e-beam increases, the generated caustics is blurred by incoherent averaging over the emission of more separated line dipoles.

Figure 6 | Limitations due to quantum effects on the formation of the X-ray beams: the role of electron coherence. Panel (a) illustrates the role of electron coherence and the variation of the reciprocal lattice vectors in the generation of caustics. Constrained by conservation of energy and momentum, the coherent photons are entangled with the corresponding electron final state ($\mathbf{p}'$ or $\mathbf{p}'_1$) from electron initial states of the same energy. The black dashed curves represent electron iso-energy contours. The red lines indicate photon momenta, so they share the same length but have different directions. The yellow lines are reciprocal lattice vectors, which vary both in length and direction. Coherence from the initial momentum distribution of the electron (denoted by the purple ellipse) is transferred to the photons via the connected red and yellow lines. For relativistic electrons, the curvatures of the black dashed curves are very small, making them almost flat in the neighborhood of the p_z axis. Therefore, the quantum coherence of the electron is limited only in the transverse directions. The right inset represents the Feynman diagram of the PXR process, where $\mathbf{p}$, $\mathbf{p}'$, $\mathbf{k}$, and $\mathbf{g}$ denote the momenta of the initial electron, the final electron, the photon, and the reciprocal lattice vector, respectively. Panel (b) presents the required electron coherence (in the p_x momentum component) for the Airy beam to be created with full coherence, shown for different sample radii, thicknesses, and L/R ratios.

In conclusion, we propose a paradigm for generating X-ray caustics from free electrons traversing bent crystal structures. Our work anticipates the application of light-sheet imaging in the X-ray regime. The generation of X-ray caustics is integrated as an intrinsic feature of the X-ray source, eliminating the need for X-ray lenses or masks, which are lossy or non-existent in the X-ray regime. The diffraction-free length and other characteristics of the so-produced Airy beams can be adjusted by tuning the parameters of the incident electron and crystal bending. The analysis of quantum aspects in the generation process shows that entanglement of post-emission electron states and photon states can prevent caustic generation unless the transverse coherent momentum of the electron is properly prepared.

The findings of this work are not limited to cylindrical bending, which approximates any general 2D bending for small thicknesses, as long as the radius of curvature changes monotonously

along the electron trajectory. More general variations of curvature in 3D can be included in Eq. (2) through the functional dependence of the incidence angle θ .

For a slightly-to-moderately relativistic electron, as discussed in this work, only the local curvature of the shaped vdW materials contributes to the X-ray beam shaping. In contrast, ultra-relativistic electrons provide a much broader excitation, and therefore, the curvature over a larger area of the bent vdW material plays a role in their induced X-ray beam shaping. More complex structured X-ray beams can be produced following this concept.

Another interesting possibility is the dynamical control over the produced Airy beams by relying on real-time modulation of the material strain, which can be achieved by applying acoustic waves⁵⁷ or by introducing pressure differences above and below the multilayer crystal^{4,17}.

Methods

Scattering field of an electron by a single flat crystal layer

Consider a crystal layer with Bravais lattice spanning the x - y plane at $z=0$. We use electric dipoles to model the bound electrons at discrete lattice points $\mathbf{R}=m\mathbf{a}_1 + n\mathbf{a}_2 + \mathbf{R}_a$, where $\mathbf{R}=(x, y)$, $\mathbf{a}_1$ and $\mathbf{a}_2$ are 2D primitive vectors, m and n are integers, and $\mathbf{r}_a=(\mathbf{R}_a, \mathbf{0})$ is the position of the atom relative to the center of the 2D unit cell. An electron of velocity $\mathbf{v}$ penetrates the crystal layer at position $\mathbf{r}_e=(\mathbf{R}_e, 0)$, with incidence angle θ relative to the plane normal. The scattering field produced by the interaction of the free electron with the crystal layer can be decomposed into contributions associated with different in-plane reciprocal lattice vectors $\mathbf{g}_\parallel$ as (Supplementary section 1)

$$\mathbf{E}(\mathbf{g}_\parallel, \mathbf{r}, \omega) = \frac{ie\alpha(\omega)}{A\epsilon_0^2 v_z} \frac{e^{ikr' + i\mathbf{g}_\parallel \cdot (\mathbf{R}_a - \mathbf{R}_e)}}{4\pi r'} \frac{(k^2 - \mathbf{k}\mathbf{k}) \cdot (k\boldsymbol{\beta} - \boldsymbol{\kappa})}{k^2 - \kappa^2}, \quad (4)$$

where $\alpha(\omega)$ is the atomic polarizability, A is the unit cell area, ϵ_0 is the vacuum permittivity, v_z is the z component of electron velocity $\mathbf{v}$, $r'=|\mathbf{r} - \mathbf{r}_e|$ is the distance to the impact point of the electron, $k=\omega/c$ is the light wavenumber in vacuum, $\mathbf{k}=\mathbf{k}(\mathbf{r} - \mathbf{r}_e)/r'$, $\boldsymbol{\beta}=\mathbf{v}/c$, c is the light speed in vacuum, $\boldsymbol{\kappa}=(\mathbf{k}_\parallel + \mathbf{g}_\parallel) + \frac{\omega - (\mathbf{k}_\parallel + \mathbf{g}_\parallel) \cdot \mathbf{v}}{v_z} \hat{z}$, and $\mathbf{k}_\parallel$ is the x - y component of $\mathbf{k}$.

The final scattering field is the coherent summation of the fields represented by Eq. (1), arising from different layers for a common $\mathbf{g}_{\parallel}$. However, the contributions from different in-plane reciprocal lattice vectors are summed incoherently, because they contribute to different caustics (i.e., the incoherent average over the electron impact position⁴⁴ $\mathbf{R}_e$ results in the incoherent summation over $\mathbf{g}_{\parallel}$). The strongest Airy beam shown in Fig. 2 comes from $\mathbf{g}_{\parallel} = 0$.

Substituting a flat layer for a bent layer

Consider a 1 MeV free electron penetrating the cylindrically bent WSe₂ structure as shown in Fig. 2b. In the calculation, we substitute the bent layers by flat layers, which are tangent to the former ones at the impacting points of the electron. Importantly, we model the flat layer in a way that the atom positions match those in the bent layer in the neighborhood of the electron impacting point. Details can be found in Supplementary section 2. This approximation is justified by examining the Bohr cutoff⁵⁸ $v\gamma/\omega$ (γ is the Lorentz contraction factor and ω is the angular frequency of the electron field), which gives the characteristic distance for the exponential decay of the evanescent electromagnetic field that accompanies the free electron in vacuum. In our case, this distance is smaller than the dimension of one unit cell (0.18 nm for a 1 MeV electron and 3 keV photons). The range of electron kinetic energies that accommodates this approximation is discussed in Supplementary section 4.

Limitations due to quantum effects

The initial electron-photon state is described as a superposition of electron momentum states $|\mathbf{p}\rangle$ as $|i\rangle = \sum_{\mathbf{p}} \frac{1}{\sqrt{V}} \psi(\mathbf{p}) |\mathbf{p}\rangle \otimes |0\rangle$, where V is the quantization volume, $|0\rangle$ represents the photon vacuum, and $\psi(\mathbf{p})$ is the momentum-space wave function (i.e., the amplitude of each electron momentum). Within first-order perturbation theory, the final electron-photon state is described as $|f\rangle = \sum_{\mathbf{p}', \mathbf{k}} \frac{1}{\sqrt{V}} \psi(\mathbf{k}, \mathbf{p}') |\mathbf{p}'\rangle \otimes |1_{\mathbf{k}}\rangle$, where now the sum includes the emitted photon wave vector $\mathbf{k}$. Here, $\psi(\mathbf{k}, \mathbf{p}')$ is the corresponding amplitude, and $|1_{\mathbf{k}}\rangle$ denotes a one-photon state. Due to energy conservation, all initial electron states that transition to the same final electron state (black dot in Fig. 6a) by the emission of a photon of a fixed energy but arbitrary direction (red lines) must distribute along an isoenergetic surface (black dashed curves). In Supplementary section 6, we

show that the reduced photon-density operator is $\rho_{\text{ph}} = \frac{1}{V} \sum_{\mathbf{p}'} \sum_{\mathbf{k}, \mathbf{k}'} \psi(\mathbf{k}, \mathbf{p}') \psi^*(\mathbf{k}', \mathbf{p}') |1_{\mathbf{k}}\rangle \langle 1_{\mathbf{k}'}|$. Thus, the coherence of the photon emission is determined by the width of $\psi(\mathbf{k}, \mathbf{p}')$, which is fundamentally tied to the initial electron wave function $\psi(\mathbf{p} = \mathbf{p}' + \mathbf{g} + \mathbf{k})$ via Fermi's golden rule. A detailed calculation can be found in Supplementary section 7.

The electron transition from the initial state to the final state is sketched by the yellow and red lines in Fig. 6a. The purple ellipse denotes the coherent momentum range of the initial electron (i.e., the distribution region of $\psi(\mathbf{p})$). Electron coherence is transferred to the emitted photons via the connecting lines. From momentum conservation $\mathbf{p} = \mathbf{p}' + \mathbf{g} + \mathbf{k}$, we know that the electron coherent momentum range $\Delta\mathbf{p}$ required for the Airy beam can be approximated by $\Delta\mathbf{p} = \Delta(\mathbf{g} + \mathbf{k})$. The value of $\Delta(\mathbf{g} + \mathbf{k})$ can be calculated from Eq. (2). We plot in Fig. 6b the maximum difference of $g_x + k_x$ along the caustic line for different sample radii, thicknesses, and L/R ratios, establishing a criterium for the approximate electron coherent momentum range required for generating Airy beams. When the electron coherent momentum range is smaller than this criterium, the generated Airy beam will extend over a shorter axial length.

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