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SUPPLEMENTARY INFORMATION

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Quantum imaging with incoherently scattered light from a free-electron laser

Creation of x-ray photons

To generate XUV photons, the FLASH FEL was operated in a 10 Hz single pulse mode with six undulator modules and total undulator length of 30 m. The electron bunch charge was 0.3 nC, and the electron beam energy 678.0 MeV. The average photon pulse length, deduced from a statistical analysis of the FEL pulses[21], was below 60 fs (FWHM) and the average photon pulse energy 25 μJ. The central wavelength λ, measured at the PG2 beamline in spectrometer mode, was 13.2 nm (93.93 eV) with a spectral bandwidth of 1.1%. With an exit slit opening of 300 μm, the monochromator settings allow a bandpass filtering to 0.096% (0.09 eV). With four C-coated mirrors, one grating and the slit bandpass used, the beamline transmission was 2%.

Generation of pseudo-thermal light

The coherent x-ray beam impinges on a diffusor produced from spin coating a Si_3N_4 substrate of size 10 x 10 mm^2 and 200 μm thickness with a solution of silica (SiO_2) nanospheres of diameter 200 nm in ethanol. The photons from the coherent FEL beam were scattered with a random phase from the homogeneous monolayer of silica spheres, resulting in a speckle pattern with a Gaussian field distribution in the source plane as well as in the detection plane (see Figs. 1 and 2(a) of the main text)[18,19,26]. To ensure that each single shot is an independent realisation of a chaotic light field, the diffusor was constantly moved laterally with a speed of 0.02 mm/s during measurements so that every laser pulse impinges on a different part of the diffuser and creates during each laser pulse a different stationary speckle pattern (see Fig. 2(a)).

A mask with six holes in a hexagonal structure mimicking a benzene molecule was placed behind the diffusor. The mask was made by perforating a Si_3N_4 substrate of 200 nm thickness with focused ion beam technique producing six free-standing square holes with equal side length a = 4.0 μm, arranged in a hexagonal geometry with grid constants d_x = 5.0 μm and d_y = 8.7 μm in x- and y-direction, respectively (see Fig. 1). The opacity of the mask is increased by evaporating ≈ 340 nm thick gold layer onto the mask's surface.

The mask was located at a distance of 10 mm behind the diffusor ensuring that many coherence cells are located within the area of each hole. In this way, each hole acts as an

independent source of pseudo-thermal light, whose lateral coherence length l_c in the far field is defined according to van Cittert-Zernike theorem by the side lengths of the holes[18].

Measurement of higher-order intensity correlations

In order to record higher order intensity correlations, a CCD camera (Andor iKon-L 936) with 2048×2048 pixels of size $13.5 \times 13.5\ \mu m^2$ was placed behind the hole mask at a distance $L = 275$ mm. This yields a lateral coherence length of $l_c \approx 1.3$ mm at the CCD which extends over many pixels, a prerequisite to resolve the higher-order intensity correlations. Due to the limited read out speed of the CCD, a dynamic beam shutter with a frequency of 1 Hz was used, blocking the beam for the whole exposure but the duration of a single pulse. Each pixel of the CCD can be considered to be an individual photon detector. In order to calculate the m[th]-order spatial intensity correlation function $g^{(m)}(\boldsymbol{r}_1, \dots, \boldsymbol{r}_m) = \frac{\langle I(\boldsymbol{r}_1) \cdots I(\boldsymbol{r}_m) \rangle}{\langle I(\boldsymbol{r}_1) \rangle \cdots \langle I(\boldsymbol{r}_m) \rangle}$, the intensities $I(\boldsymbol{r}_i)$ (grey values) measured by m pixels of the CCD at positions $\boldsymbol{r}_i,\ i = 1, \dots, m$, were multiplied and the products thereafter averaged over the whole data set, consisting of the central area (300 x 300 pixels) of all CCD images (10.800 uncompressed 16-bit greyscale images). To enhance the statistics the concept of spatial averaging was applied. This means that each image of 300 x 300 pixels is divided into 441 partially overlapping sub regions of size 200 x 200 pixels. A preliminary correlation function is calculated by processing only one subregion for the whole set of 10.800 images. This is repeated for all subregions. The final correlation function is then obtained by averaging over all 441 preliminary correlation functions, which is possible since $g^{(m)}$ depends only on relative detector positions. Note that applying this method does not affect the spatial frequency determination used in our imaging algorithm[28]. The normalisation factor appearing in the denominator of $g^{(m)}(\boldsymbol{r}_1, \dots, \boldsymbol{r}_m)$ was obtained by taking into account the average grey value for each individual detector pixel at position $\boldsymbol{r}_i,\ i = 1, \dots, m$.

Imaging algorithm

To apply our imaging algorithm we need to translate particular pixel positions on the CCD to detector positions. The phases of the fixed detectors at the magic positions (MP) along the x-axis are given by $\tilde{f}_x\, x_j = \frac{j-2}{m-1}$ for $j = 2, \dots, m$, where $\tilde{f}_x = d_x/(\lambda L)$ is the spatial frequency associated with the lattice constant d_x. To determine d_x experimentally, we investigated the correlation functions of 3[rd] order along the x-axis. We started with the two fixed detector pixels located approximately at the same position and spread them out in subsequent

evaluations of $g^{(3)}(x_1, x_2, x_3)$. We exploit the fact that if $m-1$ equidistantly separated pixels match the MP all spatial frequencies f_x not fulfilling the filtering condition $f_x = \kappa(m-1)\tilde{f}_x$ are suppressed[24]. Extracting the corresponding pixel positions for $m = 3$ we determine the grid constant via $d_x = \frac{\lambda L}{2\, x_3}$ from the position x_3 of the third detector. This process can be applied equally to the y-direction, providing access to d_y. Knowing d_x and d_y, the pixel positions representing the MP for any correlation order *m* can be calculated in the x- and the y-direction.

In the evaluation of the experiment, the m-1 fixed pixels were set to the MP along the x-axis in the center of a given subregion, and two-dimensional correlation functions $g^{(m)}(\boldsymbol{r}_1; MP_x)$ were evaluated for $m = 3,4,5$. Employing the filtering condition $f_{i,x} = \kappa(m-1)\tilde{f}_x$, $\kappa \in \mathbb{N}_0$, this allows to access all spatial frequency vectors $\boldsymbol{f}_i$ of the benzene molecule of which the x-component $f_{i,x}$ is a multiple of 0, 2, 3, 4. Equally, $g^{(m)}(\boldsymbol{r}_1; MP_y)$ were determined for $m = 3,4,5$, with m-1 pixels fixed to the MP along the y-axis in the center of a given subregion, providing information about all spatial frequency vectors $\boldsymbol{f}_i$ of the benzene molecule of which the y-component $f_{i,y}$ is a multiple of 0, 2, 3, 4. To retrieve the spatial frequency vectors $\boldsymbol{f}_i = (f_{i,x}, f_{i,y})$ of our benzene structure from the measured correlation signals, two-dimensional functions of the form

$$g_{fit}^{(m)}(x,y) = c_0 + env(x,y,w_x,w_y) \times \left[\sum_i A_i \times \cos\left(\pi f_{i,x} x + \pi f_{i,y} y\right)\right] + u_0 \times env(x,y,u_x,u_y) \;;$$

$$env(x,y,w_x,w_y) := sinc\left[\left(\pi \tilde{f}_x\, x - \frac{m-2}{m-1}\pi\right)\frac{w_x}{2\, d_x}\right]^2 \times sinc\left[\left(\pi \tilde{f}_y\, y\right)\frac{w_y}{2\, d_y}\right]^2$$

were used to fit the experimental data by use of a least squares fit. The term in square brackets together with the offset c_0 refers to $g^{(m)}$ for point-like sources. Thereby the amplitudes A_i represent the relative strength of the corresponding spatial frequencies, what can be used to remove potential ambiguities in the image reconstruction if necessary. As real sources have a finite extension we multiply this term with an envelope function $env(x,y,w_x,w_y)$, which is an approximation to the exact analytic expression, where the envelope is registered by each fixed detector and is thus slightly shifted from each other[24]. However, the average of squared residuals between the approximation and the exact expression, which is far more complex to be fitted, is below 0.1 %. As in our experiment the six holes are identical, their extensions in the x- and y-directions can be deduced from the fit parameters w_x, w_y. The shift $\frac{m-2}{m-1}\pi$ in the x-

component of the envelope results from the distribution of the fixed detectors at the MP along the x-axis. For correlations with the fixed detectors aligned at the MP along the y-axis, this shift transfers to the y-component of the envelope. Additionally there is a second envelope function $u_0 \times env(x, y, u_x, u_y)$, which approximates the change of $g^{(m)}$ due to different source intensities[24]. The fit parameters u_0, u_x, u_y are needed for the fit, however, they are of no relevance for the image reconstruction.

From the fit curves we extracted a set of seven frequency vectors $\binom{f_{i,x}}{f_{i,y}}$, representing ζ_{exp}. Note that the two spatial frequency vectors $\binom{1}{1}$ and $\binom{1}{-1}$ cannot be determined from $g^{(m)}(\boldsymbol{r}_1)$, if $m \geq 3$. Hence, there is a four-fold ambiguity to obtain the complete set of spatial frequencies ζ of the benzene structure from the set ζ_{exp}, as either none, one of the two, or both spatial frequency vectors $\binom{1}{1}$ and $\binom{1}{-1}$ could be contained in ζ. However, there is only one solution for completing this set, namely adding both frequency vectors $\binom{1}{1}$ and $\binom{1}{-1}$, as none of the other three possibilities to complete ζ leads to a meaningful solution of the source arrangement in real space. The arrangement of the sources in real space is obtained by a computer algorithm that iteratively positions the sources and calculates the corresponding spatial frequencies until a source geometry is obtained that reproduces ζ.

Error discussion

The statistical error of the experimentally obtained spatial frequencies was estimated by comparing measurements $g^{(m)}(\boldsymbol{r}_1; MP)$ of different correlation orders m containing the same spatial frequencies. This leads to a statistical error of less than 1%. The systematical errors can be significantly larger, resulting mainly from the finite size of the CCD pixels ($13.5 \times 13.5\ \mu m^2$)[24]. Since the MP are assumed to be point-like, a finite pixel size results in an integration of the intensity pattern around the MP. Furthermore, the finite pixel size allows only for discrete values of the experimentally determined MP and consequently the lattice constants d_x and d_y. For d_x and d_y (and thus $\tilde{f}_x$ and $\tilde{f}_y$) we derive a corresponding systematic uncertainty of 3.6% and 6.3%, respectively. Moreover, the width and height of the sources are extracted from the envelope function $env(x, y, w_x, w_y)$, which are governed by d_x and d_y as well. Both errors were taken into account in the image reconstruction for the positions of the individual sources (with respect to the center of the benzene structure) as well as the source extension (with respect to the respective source positions). In the final image of Fig. 3, the uncertainties are visualised by Gaussian color gradients in the x- and y-directions, with errors

corresponding to the respective standard deviations. Since the origin of the image is chosen in the center of the benzene structure, a larger distance to the center leads to increasing errors in position, as lengths are determined in units of d_x and d_y and their respective uncertainties. Hence, the errors in position in y-direction for the two top and the two bottom sources are larger than for the left and right sources and vice-versa for the errors in position in the x-direction. The positional uncertainty is given by $(\Delta d_x, \Delta d_y) = (0.18\,\mu m, 0.53\,\mu m)$ for the two top and the two bottom sources and $(\Delta d_x, \Delta d_y) = (0.36\,\mu m, 0\,\mu m)$ for the left and right sources. Additionally the uncertainty for the extension of each source in x- and y-direction is $\Delta w_x = 0.11\,\mu m$ and $\Delta \mathrm{w_y} = 0.20\,\mu m$, respectively.

Explanation of the frequency filtering process

To illustrate the frequency filtering process we consider the quantum mechanical expression of the m[th]-order correlation functions $g^{(m)}(\boldsymbol{r_1}, \dots, \boldsymbol{r_m}) \sim \langle \hat{E}^{(-)}(\boldsymbol{r_1}) \dots \hat{E}^{(-)}(\boldsymbol{r_m}) \hat{E}^{(+)}(\boldsymbol{r_m}) \dots \hat{E}^{(+)}(\boldsymbol{r_1}) \rangle$, where the positive frequency part of the field operator in the far field of N sources is given by $\hat{E}^{(+)}(\boldsymbol{r_j}) \sim \sum_{i=1}^{N} e^{ik\,\boldsymbol{n_j}\cdot\boldsymbol{R_i}}\,\hat{a}_i$. Here, $\boldsymbol{n_j}$ is the normalised vector pointing towards the detector at $\boldsymbol{r_j}$, and $\boldsymbol{R_i}$ and $\hat{a}_i$ are the position of source i (on a lattice with lattice constants (d_x, d_y)) and the annihilation operator of a photon from source i, respectively. Due to the far field condition we can approximate the phase $k\boldsymbol{n_j} \cdot \boldsymbol{R_i} \approx 2\pi \tilde{f}_{i,x} x_j + 2\pi \tilde{f}_{i,y} y_j$. This leads to a separation in x- and y-components of the phase factors appearing in $g^{(m)}(\boldsymbol{r_1}, \dots, \boldsymbol{r_m})$. By placing the $m-1$ fixed detectors along the y-axis at $y_2 = \dots = y_m = 0$, the y-dependent phase components vanish, reducing the further calculations to one dimension. With the $m-1$ fixed detectors placed along the x-axis at the MP, one can show that all phase terms vanish but those fulfilling the filtering condition $f_x = \kappa(m-1)\tilde{f}_x$, $\kappa \in \mathbb{N}_0$.[24] The y-components of the spatial frequencies are not accessed by the fixed detectors, but are included in the phase of the moving detector. Hence, although the filtering process targets only the x-components of the spatial frequencies, the y-components appear alongside with the x-components in the two-dimensional correlation function $g^{(m)}(\boldsymbol{r_1};\, MP_x)$. An analogue argumentation holds for $g^{(m)}(\boldsymbol{r_1};\, MP_y)$, i.e., placing the $m-1$ fixed detectors at the MP along the y-direction.

Resolution power

Our imaging algorithm requires a certain numerical aperture $\mathcal{A}$ in the detection plane to determine the spatial frequencies and thus to reconstruct the unknown emitter geometry. From

classical optics and Abbes resolution limit we know that the numerical aperture required to image a source arrangement with spatial frequency set ζ is given by $\mathcal{A}_1^{(1)} \geq \frac{\lambda}{2 \cdot d_{min}}$, with $d_{min} = min_{\boldsymbol{f} \in \zeta} \sqrt{|f_x \cdot d_x|^2 + |f_y \cdot d_y|^2}$, with d_x and d_y the lattice constants along the x- and y-direction, respectively. Obviously, $\mathcal{A}_1^{(1)}$ is determined by the smallest source-pair distance occurring in the source arrangement.

According to our algorithm, we resolve only a subset of the complete set of spatial frequencies ζ within a given correlation function $g^{(m)}(\boldsymbol{r_1}; MP)$ of order m, that is we resolve either the smallest spatial frequency, associated with d_{min}, or larger frequencies. Therefore, the moving detector D_1 at $\boldsymbol{r_1}$ will always require a numerical aperture $\mathcal{A}_1^{(m)} \leq \mathcal{A}_1^{(1)}$.

The numerical aperture $\mathcal{A}_{1,\ldots,m}^{(m)}$, required not only by the moving detector D_1 alone but by all detectors D_1,.., D_m, including the m-1 detectors at the MP, depends on the grid constants d_x and d_y. $\mathcal{A}_{1,\ldots,m}^{(m)}$ is smaller than $\mathcal{A}_1^{(1)}$ if the frequency vectors $\binom{0}{1}$ or $\binom{1}{0}$, corresponding to the smaller of the grid constants d_x and d_y, is part of the complete set of spatial frequencies ζ[24]. Under these conditions our imaging algorithm provides a resolution below the canonical Abbe limit. If this condition is not met, a larger aperture is needed and our algorithm will not overcome the Abbe limit. In our particular arrangement of sources in the form of a benzene like hexagonal structure, the source geometry does not exhibit one of the spatial frequency vectors $\binom{0}{1}$ or $\binom{1}{0}$, so in this case we do not beat the resolution limit of classical optics.

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.