Quantum Imaging with Incoherently Scattered Light from a Free-Electron Laser

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The advent of accelerator-driven hard x-ray free-electron lasers (FEL) has opened new avenues for high-resolution structure determination via diffraction methods. With the extremely intense, ultrashort coherent pulses from these sources it has become possible to record diffraction patterns of nanoparticles, viruses, microcrystals and non-periodic objects in a single shot, opening new possibilities for structural studies that go beyond conventional x-ray crystallography methods\textsuperscript{1-7}. The ultimate goal is to reveal the elementary steps of structural and electronic dynamics of single molecules with atomic resolution\textsuperscript{8-10}. Since all present-day imaging methods using FELs rely on coherent scattering processes, great efforts are undertaken to maintain the 1\textsuperscript{st}-order coherence.
of the radiation field upon propagation from the source through the optical system towards the sample. On the other hand, inevitable causes of incoherence in the interaction of x-rays with matter\textsuperscript{11,12} often lead to background that completely obstructs the coherent diffraction signal. Here we show that higher-order intensity correlations of incoherently scattered x-rays can reveal the full information about the arrangement of the scatterers. It is achieved via analysis of photon correlations in the detector plane to eventually reach a spatial resolution even below the Abbe diffraction limit. Our method is an extension of the intensity correlation measurements of Hanbury-Brown and Twiss (HBT)\textsuperscript{13} to higher than 2\textsuperscript{nd}-order and is applied here to image a non-periodic two-dimensional (2D) distribution of incoherent scatterers resembling an artificial molecule. The well-known phase problem of coherent diffraction imaging\textsuperscript{14} is bypassed as the spatial frequency vectors directly appear in the higher-order correlations. Our method constitutes a new approach towards x-ray structure determination, exploiting higher degrees of coherence rather than the fragile 1\textsuperscript{st}-order coherence of the light field, thus enabling structure determination under conditions of incoherent scattering processes that are generally considered detrimental for imaging applications.

The discovery by Hanbury Brown and Twiss of photon bunching of thermal light, that is the enhanced probability of pairs of photons arriving within a time span short compared to the coherence time of the source\textsuperscript{15}, and its use for the determination of the angular diameter of stars by measuring spatial photon correlations\textsuperscript{13} was a hallmark experiment for the development of modern quantum optics\textsuperscript{16}. The quantum mechanical description of photon correlations by Glauber paved the way for a generalised concept of optical coherence\textsuperscript{17} that is founded on the analysis of correlation functions of order \(m\) rather than the 1\textsuperscript{st}-order coherence. For example, the spatial 2\textsuperscript{nd}-order photon correlation function \(g^{(2)}(r_1, r_2)\) relates the probability to detect a photon at position \(r_1\) given that a photon is recorded at position \(r_2\). In the case of two incoherent thermal point sources one obtains a cosine modulation which oscillates at a spatial frequency depending on the source separation\textsuperscript{18,19}. Thus interference fringes show up even in the case of complete absence of 1\textsuperscript{st}-order coherence, allowing for the extraction of structural information for incoherently emitting objects. This has been applied in earth-bound stellar interferometry to measure the angular diameter of stars with 100-fold increased resolution\textsuperscript{13} or to reveal the statistical properties of pulsed FEL
sources\textsuperscript{20,21}, essentially bypassing the phase fluctuations imposed by imperfections along the path from the source to the detector.

Extending this concept to arbitrary arrangements of incoherently scattering emitters enables one to use intensity correlations for imaging applications. This has been demonstrated recently for one-dimensional distributions of emitters in the visible range of the spectrum\textsuperscript{22-24} where a spatial resolution even below the canonical Abbe limit has been achieved. To open up a new approach for x-ray structure determination we go still further and employ the method to image arbitrary two-dimensional arrangements of objects which incoherently scatter XUV radiation.

The experiment has been performed at the PG2 beamline of the FLASH FEL facility at DESY, Hamburg\textsuperscript{25}. The scheme of the setup is shown in Fig. 1. The FEL runs in a 10 Hz pulsed mode ($\tau_{\text{pulse}} \approx 60$ fs) at a center wavelength of $\lambda = 13.2$ nm (for experimental details see\textsuperscript{21}). After passing a monochromator to limit the bandwidth to 0.1% ($\pm 0.013$ nm), the laser beam impinges on a slowly moving diffusor converting the spatially highly coherent light of the FEL to quasi-monochromatic pseudo-thermal light with a far field speckle pattern that fluctuates randomly from shot to shot but is stationary for each pulse\textsuperscript{26}. A two-dimensional object mask is placed behind the diffusor, consisting of six square-cut holes in a hexagonal arrangement to resemble the carbon atoms in a benzene molecule. The structure of this object is characterised by a set of nine spatial frequency vectors $\zeta = \{f_i = (f_{ix}, f_{iy}) = \{h_{i,x}/\lambda L, h_{i,y}/\lambda L\}, i = 1, \ldots, 9\}$, determined by the nine different source separation vectors $h_i = (h_{ix}, h_{iy}), \ i = 1, \ldots, 9$, connecting the artificial atoms (holes in the mask) of the molecule (see Fig. 1).

When illuminated with the light scattered by the diffusor the artificial atoms represented by the holes in the mask act as pseudo-thermal sources that emit incoherently and independently from each other. The resulting intensity pattern is recorded in the far-field for each pulse individually (see Fig. 2a) where a stationary field distribution is ensured due to the extremely short pulse lengths of the FEL (see Methods section). Correlating the intensities $I(r_i)$ at different positions $r_i, \ i = 1, \ldots, m$, for each pulse and averaging the result over many pulses produces the wanted $m^{th}$-order intensity correlation function

$$g^{(m)}(r_1, \ldots, r_m) = \frac{\langle I(r_1) \cdots I(r_m) \rangle}{\langle I(r_1) \rangle \cdots \langle I(r_m) \rangle}$$  \quad (1)
Our imaging algorithm using higher-order intensity correlations is based on extracting the set of spatial frequency vectors $\zeta$ of the unknown source distribution – assumed to be placed on a 2D lattice –, from which the source geometry in real space can be reconstructed (see Method section). As in our technique two-dimensional spatial frequency vectors directly appear in the higher-order correlations we overcome the phase problem of coherent diffraction imaging [14]. In the intensity correlations of 2$^{\text{nd}}$ order the entire set of spatial frequency vectors appears. This means that the amount of parameters to be fitted and extracted from the 2$^{\text{nd}}$-order correlation function is unfeasibly large, especially in the case of a data set with low signal to noise ratio. Measuring intensity correlations of higher than 2$^{\text{nd}}$ order allows to split the information, as only a subset of spatial frequencies appears within a given correlation order. This significantly reduces the number of fit parameters to be determined within each

Figure 1: Scheme of the experiment. a): The FEL beam created in the undulator (UN) passes a monochromator consisting of a grating (GR) and an exit slit (ES) such that quasi-monochromatic coherent radiation hits the moving diffusor (D). The chaotic light scattered by the diffusor is used to illuminate a benzene-like hole mask (HM) generating six quasi-monochromatic independently radiating thermal light sources. The resulting light field is measured at a distance $L$ behind HM by a CCD. b): The nine different source separation vectors $h_i$, $i = 1, \ldots, 9$, of HM are shown in different colours for a better readability. c): To
determine \( g^{(m)}(\mathbf{r}_i; MP_x) \), for each CCD image \( m - 1 \) pixels of the CCD camera are selected as detectors at the magic positions along the \( x \)-axis (red) and correlated with all other pixels (blue) of the CCD; thereafter an average over all images is taken.

correlation order, allowing one to reveal \( \zeta \) with substantially increased accuracy\(^{24} \). In certain cases a numerical aperture smaller than required by the canonical Abbe limit suffices to determine \( \zeta \), allowing for a spatial resolution below the classical diffraction limit (see Methods Section).

The light scattered by the artificial atoms is measured by a CCD camera in the far field at a distance \( L \) behind the source plane, where each pixel of the CCD serves as an independent detector. From the observed speckle patterns the \( m \)-th order spatial intensity correlation functions \( g^{(m)}(\mathbf{r}_1, ..., \mathbf{r}_m) \) (1) are computed by correlating the intensities measured by \( m \) pixel detectors at positions \( \mathbf{r}_i \), \( i = 1, ..., m \), for each CCD image individually and then averaging over all CCD images (see Methods Section).

In general \( g^{(m)}(\mathbf{r}_1, ..., \mathbf{r}_m) \) takes a complicated form, depending on the detector positions \( \mathbf{r}_1, ..., \mathbf{r}_m \) and the source geometry. However, when placing all but one detectors at the so-called magic positions (MP), only specific spatial frequency vectors of the object appear within a given intensity correlation function of order \( m \). In particular, if \( m - 1 \) detectors are placed along the \( x \)-axis such that \( f_x x_j = \frac{j-2}{m-1} \), \( j = 2, ..., m \), where \( f_x = d_x/(\lambda L) \) is the spatial frequency associated with the lattice constant \( d_x \), only those spatial frequency vectors \( f_i \) of the benzene molecule appear in \( g^{(m)}(\mathbf{r}_i; MP_x) \) where the \( x \)-component fulfils the filtering condition \( f_{ix} = \kappa (m-1) f_x \), with \( \kappa \in \mathbb{N}_0 \); an analogue filtering condition holds for the \( y \)-direction. As in 1D\(^{24} \), the filtering process in 2D is a result of sum rules which hold for the \((m-1)\)-th roots of unity (see Methods section).

As an example, we consider \( g^{(4)}(\mathbf{r}_1; MP_x) \) with three detectors located at the MP along the \( x \)-direction (see Fig. 1). In this case only the spatial frequency vectors \( \zeta_x^{(4)} = \{0, 3, 6\}/f_x \) (in units of \( f_x \) and \( f_y \)) appear in \( g^{(4)}(\mathbf{r}_1; MP_x) \), as only \( f_x = 3f_x \) (with \( \kappa = 1 \)) and \( f_x = 0 \) match the filtering condition. Note that even though the fixed detectors are aligned at the MP only along the \( x \)-direction, the corresponding components \( f_y \) of the filtered \( f_x \) also show up in \( g^{(4)}(\mathbf{r}_1; MP_x) \).
After evaluating $g^{(m)}(r; MP)$ for $m = 3, 4, 5$, with the fixed detectors aligned at the MP along the x-axis as well as along the y-axis (resulting in a total of six 2D correlation functions), the set of experimentally obtained spatial frequency vectors $\zeta_{\text{exp}}$ of the benzene ring structure is derived from a best 2D fit to the experimental data (see Methods section). As an example, the correlation function $g^{(4)}(r; MP_x)$, experimentally determined after evaluating 10,800 single shot CCD images with three pixel detectors located at the MP along the x-direction, and corresponding fit functions are shown in Fig. 2(b-d).

The set of spatial frequency vectors $\zeta_{\text{exp}}$ thus obtained is shown in Tab. 1. Note that since some spatial frequencies match the filtering condition for more than one correlation order.
they can be accessed from correlation functions $g^{(m)}(\mathbf{r}; MP)$ of different orders $m$. However, it turns out, that the values for $f_x$ and $f_y$ obtained in this way deviate from each other by less than 1%.

Table 1: Theoretically expected and experimentally obtained spatial frequency vectors (in units of $f_x$ and $f_y$); colors correspond to the colors of the spatial frequency vectors shown in Fig. 1

<table>
<thead>
<tr>
<th>$\zeta$</th>
<th>$(\frac{1}{1})$</th>
<th>$(-\frac{1}{1})$</th>
<th>$(\frac{2}{0})$</th>
<th>$(\frac{2}{-2})$</th>
<th>$(\frac{3}{1})$</th>
<th>$(\frac{3}{-1})$</th>
<th>$(\frac{4}{0})$</th>
<th>$(\frac{0}{2})$</th>
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<tr>
<td>$\zeta_{exp}$</td>
<td>$(-1.94 \pm 0.02)$</td>
<td>$(-1.96 \pm 0.01)$</td>
<td>$(-1.97 \pm 0.02)$</td>
<td>$(2.97 \pm 0.01)$</td>
<td>$(2.97 \pm 0.01)$</td>
<td>$(3.90 \pm 0.04)$</td>
<td>$(0.01 \pm 0.18)$</td>
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A systematical error for the experimentally derived spatial frequencies originates from the finite size of the pixel detectors, as this prevents the $m$-1 fixed pixel detectors from being located exactly at the MP (assumed to be point-like). In our experiment, using a CCD camera with pixel size 13.5 $\times$ 13.5 $\mu$m$^2$ at a distance of $L = 275$ mm behind the benzene-mask, this results in a systematical error for $f_x$ and $f_y$ of 3.6% and 6.3%, respectively.

Note that according to the filtering condition the two spatial frequency vectors $(\frac{1}{1})$ and $(\frac{-1}{-1})$ (in units of $f_x$ and $f_y$) cannot be determined from $g^{(m)}(\mathbf{r}; MP)$ if $m \geq 3$. Hence, without measuring $g^{(2)}(\mathbf{r}; MP)$, four different possibilities to complete the set $\zeta$ from the set $\zeta_{exp}$ are possible, containing either none, one of the two, or both of the two spatial frequencies. However, for the investigated benzene ring structure only the set containing both spatial frequency vectors provides a valid solution for the source arrangement in real space, resulting in a unique set $\zeta$ (see Methods section).

Besides the geometry of the sources, the correlation signals also contain information about the size of the sources as their finite extension translates into sinc-shaped envelopes in x- and y-direction of $g^{(m)}(\mathbf{r}; MP)$ (see Methods section). Merging the extracted fit parameters, i.e., the spatial frequencies of $g^{(m)}(\mathbf{r}; MP)$ for $m = 3, 4, 5$, as well as their envelopes, we are able to determine the entire benzene ring structure. The result is shown in Fig. 3.
Figure 3: Geometry of the artificial benzene structure. a) Scanning electron microscopy image of the hole mask. b) Image of the hole mask reconstructed after evaluating $g^{(m)}(r; MP)$ for $m = 3, 4, 5$. The uncertainties of the source positions and sizes are displayed by a Gaussian shaped color gradient with a standard deviation equal to the corresponding error (for details see Methods Section).

The reconstruction of the source geometry using our approach proves to be extremely robust with respect to intensity and frequency fluctuations of the scattered light\textsuperscript{24}. In particular, the method allows to reconstruct the source arrangement in complete absence of 1\textsuperscript{st-order} coherence, e.g., due to imperfect optics that leads to wavefront distortions or vanishing coherent diffraction signals. Such aspects become increasingly important with decreasing wavelength. Imaging applications in the regime of hard x-rays, where 1\textsuperscript{st}-order coherence might be compromised by imperfect x-ray optical elements, will therefore particularly benefit from our imaging technique.

The method has in particular great potential for imaging with intense sources of x-rays where most of the emission and scattering occurs incoherently due to inelastic processes like Compton scattering or those that are associated with the strong electromagnetic driving\textsuperscript{8}. The approach might thus become increasingly relevant and applicable for imaging with existing and future laser sources in the regime of hard x-rays, in particular with respect to the ultimate goal of single-molecule imaging\textsuperscript{9,10}. In this spirit it could be combined with novel techniques like macromolecular imaging from imperfect crystals\textsuperscript{28} where the single-
molecule scattering signal is discriminated with high signal-to-noise ratio against the coherent Bragg peaks from the crystalline structure. Moreover, applying our correlation imaging technique to the detection of resonance fluorescence as an intrinsically incoherent scattering process will enable element-specific imaging applications. Finally, it should be noted that the role of photons can be taken by any other bosonic particle since the statistical properties of the correlation functions are based on the bosonic nature. Using fermionic statistics and corresponding correlation functions, our method can be extended to imaging with pulsed beams of electrons and possibly also pulsed neutrons from spallation neutron sources.

References


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**Author contributions**

J.v.Z., R.R. and I.A.V. conceived the experiment and coordinated the experimental efforts. R.S. and T.M. designed the experimental layout and provided post-measurement analysis and evaluation of experimental data. G.M. designed and built the experimental setup. G.M., L.W., R.S., T.M., O.G., S.L., P.S. and I.Z. carried out the experiment at FLASH/DESY. A.C. and F.W. developed idea of sequential spatial frequency filtering in 1D and 2D using higher-order intensity correlations. A.C. and D.B. developed quantum path analysis of the method and mathematical explanation for sub-Abbe resolution. A.B. installed the motor control of the diffusor stage. L.B. coordinated efforts to characterize diffusors and samples. B.F. provided silica particles for production of diffusors. K.S. prepared diffusors and thin film coatings. J.W. prepared the hole mask of the artificial benzene molecule used as sample. S.W. participated in preparation and characterization of diffusors. G.B. provided the operation of the beamline PG2 at FLASH. Y.O. participated in discussion of the theoretical basis of quantum imaging. W.W. provided the end station for measurements at the PG2 beamline. R.S., T.M., J.v.Z., R.R., and I.A.V. wrote the manuscript with contributions and improvements from all authors.

**Author information**

The authors declare no competing financial interests.
Methods

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, was below 60 fs (FWHM) and the average photon pulse energy 25 µJ. The central wavelength $\lambda$, 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). 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 ring 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 $\approx 340$ 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 × 13.5 \( \mu m \)\(^2\) was placed behind the hole mask at a distance of \( L = 275 \) 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 \( g^{(m)}(r_1, ..., r_m) = \frac{I(r_1) ... I(r_m)}{I(r_1) ... I(r_m)} \) the intensities \( I(r_i) \) (grey values) measured by \( m \) pixels of the CCD at positions \( r_i, \ i = 1, ..., m \), 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\(^{27}\). The normalisation factor appearing in the denominator of \( g^{(m)}(r_1, ..., r_m) \) was obtained by taking into account the average grey value for each individual detector pixel at position \( r_i, \ i = 1, ..., m \).

**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 \( f_x x_j = \frac{j-2}{m-1} \) for \( j = 2, ..., m \), where \( f_x = d_x / (\lambda L) \) 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 $g^{(m)}(x_1, x_2, x_3)$. We exploit the fact that if $m-1$ equidistantly separated pixels match the MP all spatial frequencies $f_x$ not fulfilling the filtering condition $f_x = \kappa (m-1) f_x$ are suppressed\(^{24}\). Extracting the corresponding pixel positions for $m = 3$ we are able to 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 $g^{(m)}(r_i; MP_\alpha)$ were evaluated for $m = 3, 4, 5$. Employing the filtering condition $f_{i,x} = \kappa (m-1) f_x$, $\kappa \in \mathbb{N}_0$, this allows to access all spatial frequency vectors $f_i$ of the benzene molecule of which the x-component $f_{i,x}$ is a multiple of 0, 2, 3, 4. Equally, $g^{(m)}(r_i; MP_\beta)$ 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 $f_i$ 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 $f_i = (f_{i,x}, f_{i,y})$ of our benzene ring structure from the measured correlation signals, two-dimensional functions of the form

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

\[
\text{env}(x, y, w_x, w_y) := \text{sinc} \left( \left( \pi f_x x - \frac{m-2}{m-1} \pi \right) \frac{w_x}{2 d_x} \right)^2 \times \text{sinc} \left( \left( \pi f_y y \right) \frac{w_y}{2 d_y} \right)^2
\]

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 $\text{env}(x, y, w_x, w_y)$, 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 \( u_\phi \cdot env(x, y, u_x, u_y) \), which approximates the change of \( g^{(m)} \) due to different source intensities. The fit parameters \( u_\phi, u_x, u_y \) 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 \( \{ f_{lx}, f_{ly} \} \), representing \( \zeta_{\text{exp}} \). Note that the two spatial frequencies \( \{1\} \) and \( \{-1\} \) cannot be determined from \( g^{(m)}(\mathbf{r}_1) \) if \( m \geq 3 \). Hence, there is a four-fold ambiguity to obtain the complete set of spatial frequencies \( \zeta \) of the benzene ring structure from the set \( \zeta_{\text{exp}} \), as either none, one of the two, or both of the two vectors could be contained in \( \zeta \). However, there is only one physically valid solution for completing this set, namely adding the two frequencies \( \{1\} \) and \( \{-1\} \), as none of the other three possibilities to complete \( \zeta \) leads to a valid solution of the source arrangement in real space. Here, the arrangement of the sources 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 \( \zeta \).

**Error discussion**

The statistical error of the experimentally obtained spatial frequencies was estimated by comparing measurements \( g^{(m)}(\mathbf{r}_1; MP) \) 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 \( f_{lx} \) and \( f_{ly} \)) we estimate a corresponding systematic uncertainty of 3.6% and 6.3%, respectively. Moreover, the width and height of the sources are derived 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 ring 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 $\Delta w_y = 0.20 \, \mu m$, respectively.

**Explanation of the frequency filtering process**

To illustrate the frequency filtering process we consider the quantum mechanical calculation of the $m$th-order correlation functions $g^{(m)}(r_1, ..., r_m) = \langle \hat{E}^{(-)}(r_1) ... \hat{E}^{(-)}(r_m)\hat{E}^{(+)}(r_m) ... \hat{E}^{(+)}(r_1) \rangle$ in the far field of N sources, where the positive frequency part of the field operator is given by $\hat{E}^{(+)}(r_j) = \sum_{i=1}^{N} e^{i k n_j \cdot R_i} \hat{a}_i$. Here, $n_j$ is the normalised vector pointing towards the detector at $r_j$, and $R_i$ and $\hat{a}_i$ are the position (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 relative phase by $k n_j \cdot R_i \approx 2\pi f_{j,x} x_j + 2\pi f_{j,y} y_j$. This leads to a separation in x- and y-components of the exponential terms in $g^{(m)}(r_1, ..., r_m)$. By placing the $m-1$ fixed detectors along the y-axis at $y_2 = ... = y_m = 0$ the y-dependent phase components vanish, reducing the further calculations to one dimension. With the fixed detectors placed along the x-axis at the MP one can show that all phase terms vanish but those fulfilling the filtering condition$^{24}$. 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 the x-components of the spatial frequencies, the y-components appear alongside with the x-components in the two-dimensional correlation function $g^{(m)}(r_1; MP_2)$.

**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 least numerical aperture required to image a source arrangement with spatial frequency set $\zeta$ is given by $\mathcal{A}_{\text{min}}^{(1)} \geq \frac{\lambda}{2d_{\text{min}}}$, with $d_{\text{min}} = \min_{f \in \zeta} \sqrt{|f_x \cdot d_x|^2 + |f_y \cdot d_y|^2}$, where $d_x$ and $d_y$ are the lattice constants along the x- and y-direction, respectively. Obviously, $\mathcal{A}_{\text{min}}^{(1)}$ is governed 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 $\zeta$ within a given correlation order $m$, that is we resolve either the smallest spatial frequency, associated with $d_{\text{min}}$, or larger frequencies. Therefore, the moving detector $D_1$ will always require a numerical aperture $\mathcal{A}_{1}^{(m)} \leq \mathcal{A}_{1}^{(1)}$.

The numerical aperture $\mathcal{A}_{1,...,m}^{(m)}$, required not only by the moving detector $D_1$ but by all detectors $D_1,...,D_m$ including the m-1 detectors at the MP, depends on the grid constants $d_x$ and $d_y$. $\mathcal{A}_{1,...,m}^{(m)}$ is smaller than $\mathcal{A}_{1}^{(1)}$ if the frequency $(0_1)$ or $(1_0)$, corresponding to the smaller of the grid constants $d_x$ and $d_y$, is part of the complete set of spatial frequencies $\zeta$. Under these conditions our imaging algorithm provides a resolution below the canonical Abbe limit of classical optics. If this condition is not met a larger aperture is needed and our algorithm will not overcome the Abbe limit. In our experimental setup the source geometry does not exhibit one of the spatial frequency vectors $(0_1)$ or $(1_0)$, so in this case we do not beat the classical resolution limit.

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