Quantum imaging with incoherently scattered light from a free electron laser

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

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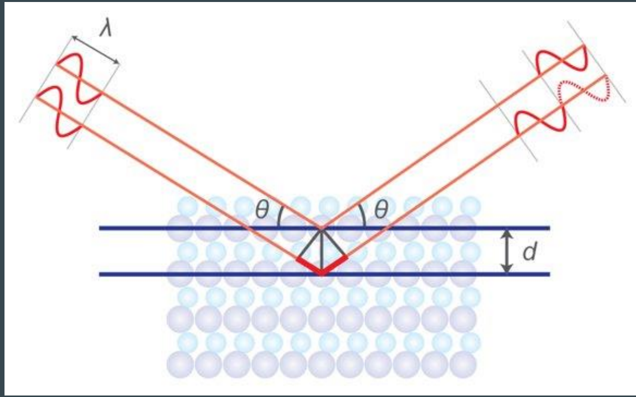
X-Ray Diffraction

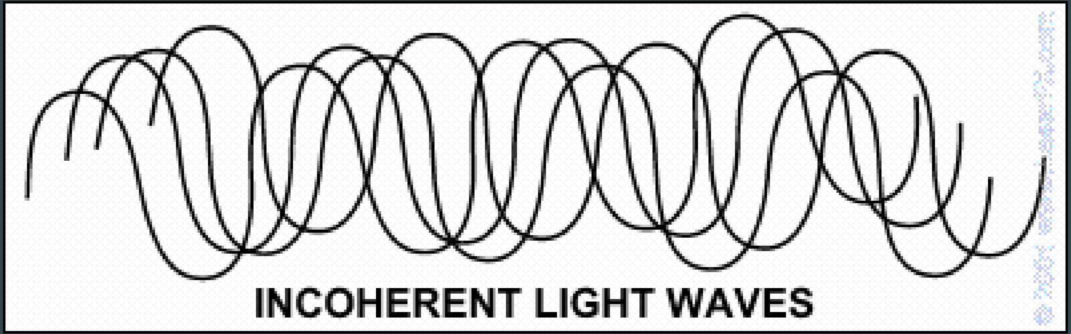


https://www.rigaku.com/en/techniques/xrd

- Traditionally used to image or determine the structure of a sample
- Most algorithms used with this technique require that the first order coherence of the radiation field be maintained
- This is difficult when using hard (high-energy) x-rays
- Methods exist to image samples using hard x-rays that only utilize higher order coherences in only 1D
- This group has extended these methods to image in 2D

Use of incoherent light

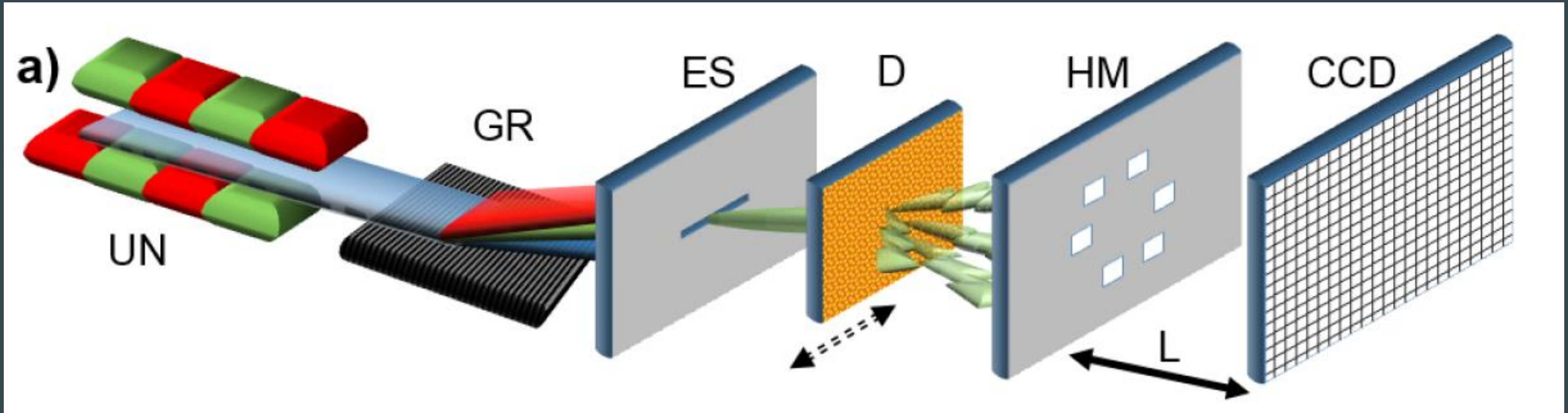
- In general, most scattered light is incoherent
  - Being able to harness this = much more robust technique



http://www.laserfx.com/Works/Works2.html

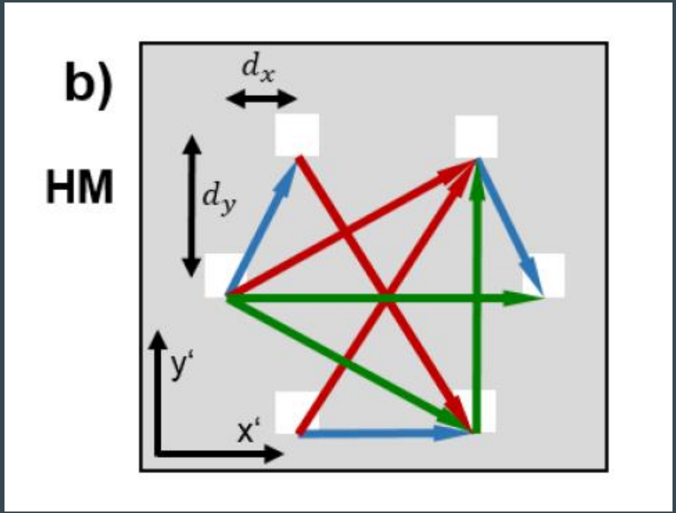
Experimental Imaging Methods Using a Free Electron Laser

- PG2 beamline at the FLASH FEL facility at DESY in Hamburg, Germany
- 60 fs pulses, 10 Hz
- 13.2 ± 0.013 nm monochromatic beam
- L = 275 mm --> lateral coherence length of 1.3 mm (prerequisite to solve higher-order correlation functions)



Sample Structure is determined using correlation functions

- Goal: Image the artificial benzene molecule which is modeled by the hole mask
- The placement of the atoms is described by the 9 source separation vectors
- Intensity patterns collected by the CCD generate an mth-order spatial intensity correlation function, denoted:



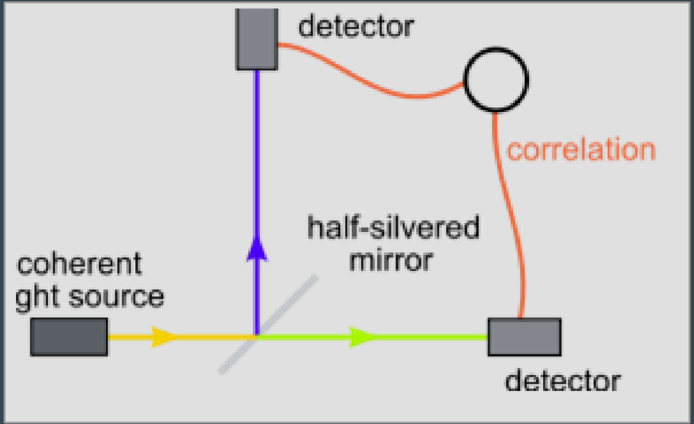
I = Intensity

r_m = position m

$$g^{(m)}(\boldsymbol{r}_1, \ldots, \boldsymbol{r}_m) = \frac{\langle I(\boldsymbol{r}_1) \cdots I(\boldsymbol{r}_m) \rangle}{\langle I(\boldsymbol{r}_1) \rangle \cdots \langle I(\boldsymbol{r}_m) \rangle}.$$

Correlation Functions

$$g^{(m)}(\boldsymbol{r}_1, \ldots, \boldsymbol{r}_m) = \frac{\langle I(\boldsymbol{r}_1) \cdots I(\boldsymbol{r}_m) \rangle}{\langle I(\boldsymbol{r}_1) \rangle \cdots \langle I(\boldsymbol{r}_m) \rangle} .$$

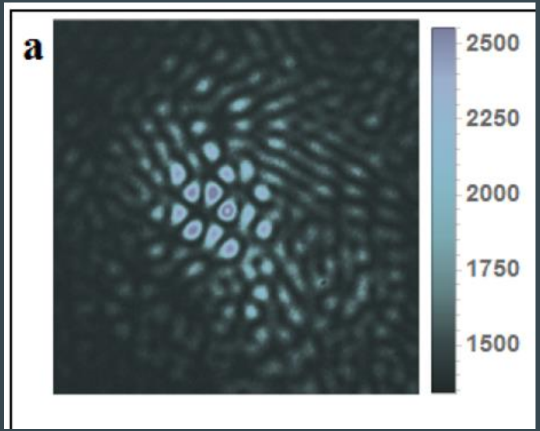


Hanbury Brown and Twiss effect - Wikipedia

- Correlation functions quantify how microscopic variables co-vary with one another on average across space and time.
- With incoherent sources, g(2) oscillates at a spatial frequency depending on the source separation.
- Higher order correlation functions (m=3,4,5) are used to find spatial frequency vectors
- The source separation vectors can be calculated from these spatial frequency vectors

Imaging algorithm vs data

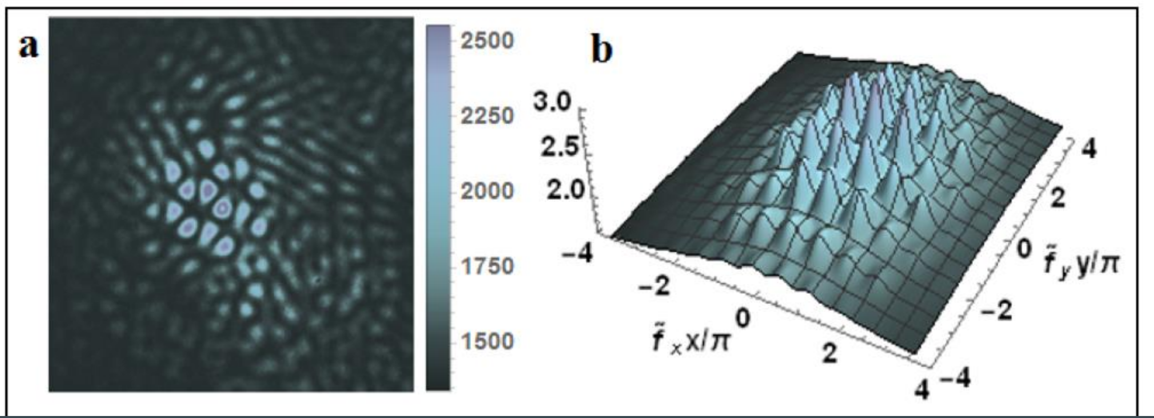
a) A single pulse through the hole mask as measured by the CCD



Imaging algorithm vs data

a) A single pulse through the hole mask as measured by the CCD

b) 10,800 pulses are compiled to obtain g(4)

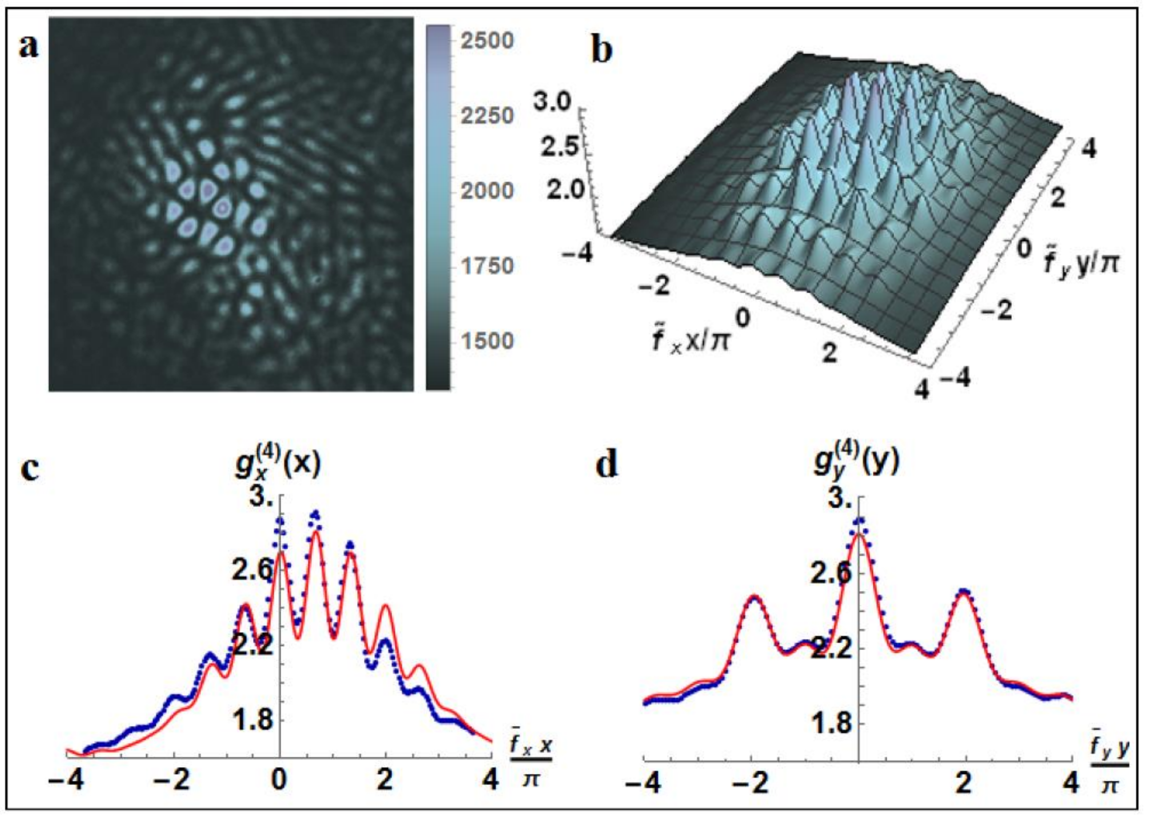


Imaging algorithm vs data

a) A single pulse through the hole mask as measured by the CCD

b) 10,800 pulses are compiled to obtain g(4)

c) & d) Raw g(4) data (blue dots) & fitted (red solid) vs. spatial frequency



Results (Experimental Errors)

- Systematic errors are caused by the assumption that the pixels are points, aka dimensionless
- f_x error: 3.6%
- f_y error: 6.3%

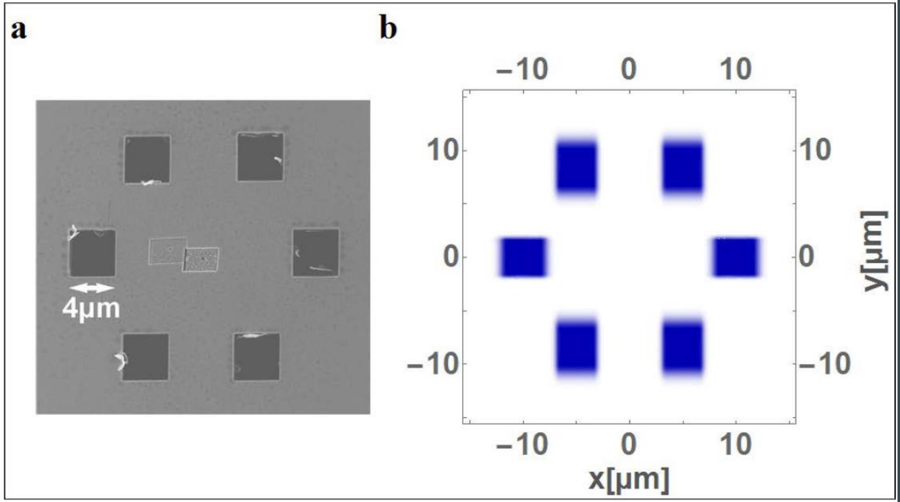


Table 1: Theoretically expected and experimentally obtained spatial frequency vectors (in units of $\tilde{f}_x$ and $\tilde{f}_y$); colors correspond to the colors of the spatial frequency vectors shown in Fig. 1

ζ	$\begin{pmatrix}1\\1\end{pmatrix}$	$\begin{pmatrix}1\\-1\end{pmatrix}$	$\begin{pmatrix}2\\0\end{pmatrix}$	$\begin{pmatrix}2\\2\end{pmatrix}$	$\begin{pmatrix}2\\-2\end{pmatrix}$	$\begin{pmatrix}3\\1\end{pmatrix}$	$\begin{pmatrix}3\\-1\end{pmatrix}$	$\begin{pmatrix}4\\0\end{pmatrix}$	$\begin{pmatrix}0\\2\end{pmatrix}$
ζ_{exp}			$\begin{pmatrix}1.94\\0.02\end{pmatrix}$	$\begin{pmatrix}1.96\\2.00\end{pmatrix}$	$\begin{pmatrix}1.97\\-1.95\end{pmatrix}$	$\begin{pmatrix}2.97\\0.97\end{pmatrix}$	$\begin{pmatrix}2.97\\-1.03\end{pmatrix}$	$\begin{pmatrix}3.90\\0.04\end{pmatrix}$	$\begin{pmatrix}0.01\\1.98\end{pmatrix}$

Other uses of incoherent light in imaging

Incoherent x-ray scattering in single molecule imaging

Jan Malte Slowik, Sang-Kil Son, Gopal Dixit, Zoltan Jurek, Robin Santra

(Submitted on 29 Jul 2014)

"...a background signal from incoherent scattering deteriorates the quality of the coherent scattering signal."

"We find that the coherent scattering pattern suffers from a significant incoherent background signal at high resolution."

- Treats incoherent scattering solely as a problem to be solved
- Recent: 2014

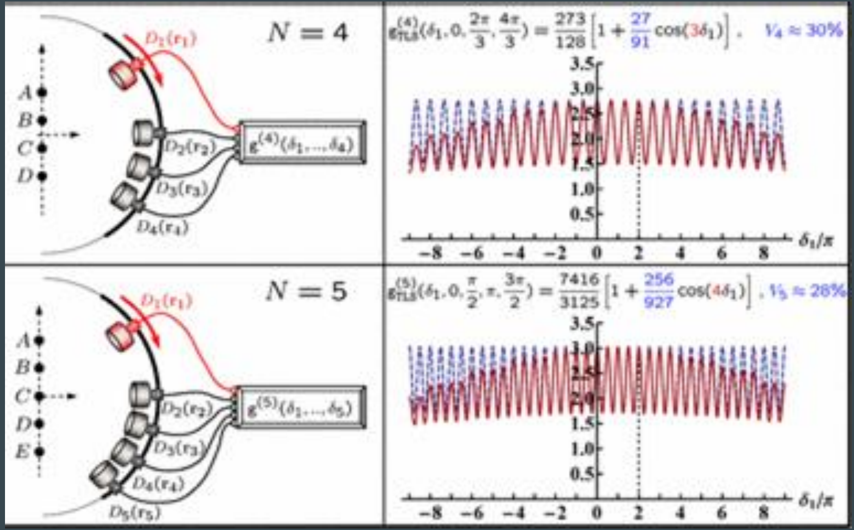
Other uses of incoherent light in imaging

Superresolving Multiphoton Interferences with Independent Light Sources

S. Oppel, T. Büttner, P. Kok, and J. von Zanthier
Phys. Rev. Lett. **109**, 233603 – Published 4 December 2012

- Uses incoherent scattering patterns for 1-D imaging
- Experiment limited to 5 separate sources
- Precedes 2014 paper discounting incoherent scattering

“Experimental results with up to five independent thermal light sources confirm this approach to <u>improve the spatial resolution.</u>” (emphasis added)



https://journals.aps.org/prl/article/10.1103/PhysRevLett.109.233603/figures/1/medium

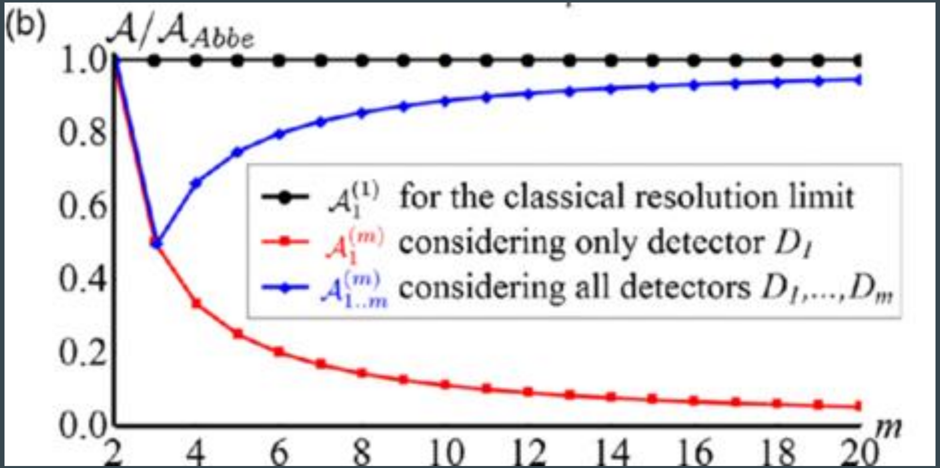
Other uses of incoherent light in imaging

Superresolving Imaging of Irregular Arrays of Thermal Light Sources using Multiphoton Interferences

Anton Classen, Felix Waldmann, Sebastian Giebel, Raimund Schneider, Daniel Bhatti, Thomas Mehringer, Joachim von Zanthier

(Submitted on 11 Aug 2016)

- Extends 2012 paper to use an arbitrary number of detectors at arbitrary separations
- Shows that in 1-D, incoherent imaging always bypasses the Abbe resolution limit
    - In 2-D, the Abbe limit is only bypassed under specific conditions



https://journals.aps.org/prl/article/10.1103/PhysRevLett.117.253601/figures/2/medium

Critique: did not demonstrate sub-Abbe limit resolution

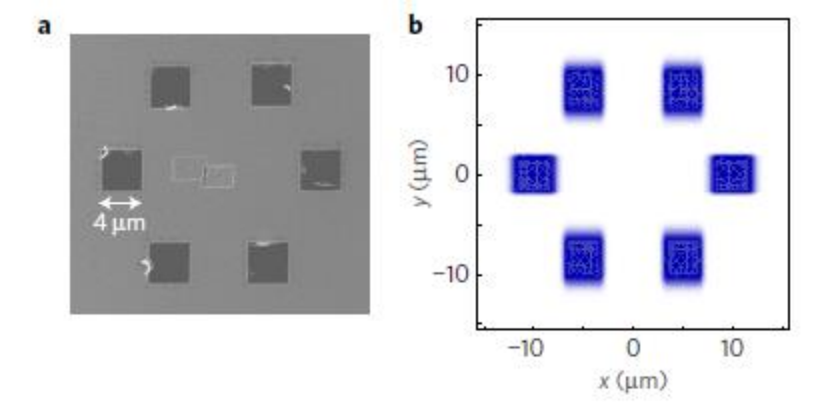
- The imaging algorithm requires $\begin{pmatrix}1\\0\end{pmatrix}$ or $\begin{pmatrix}0\\1\end{pmatrix}$ frequency vectors to breach the Abbe limit.
- The system imaged, a hole mask mimicking the structure, had the listed spatial frequency vectors for m = 3, 4, 5

	Blue			Red			Green		
ζ	$\begin{pmatrix}1\\1\end{pmatrix}$	$\begin{pmatrix}1\\-1\end{pmatrix}$	$\begin{pmatrix}2\\0\end{pmatrix}$	$\begin{pmatrix}2\\2\end{pmatrix}$	$\begin{pmatrix}2\\-2\end{pmatrix}$	$\begin{pmatrix}3\\1\end{pmatrix}$	$\begin{pmatrix}3\\-1\end{pmatrix}$	$\begin{pmatrix}4\\0\end{pmatrix}$	$\begin{pmatrix}0\\2\end{pmatrix}$
ζ_{exp}			$\begin{pmatrix}1.94\\0.02\end{pmatrix}$	$\begin{pmatrix}1.96\\2.00\end{pmatrix}$	$\begin{pmatrix}1.97\\-1.95\end{pmatrix}$	$\begin{pmatrix}2.97\\0.97\end{pmatrix}$	$\begin{pmatrix}2.97\\-1.03\end{pmatrix}$	$\begin{pmatrix}3.90\\0.04\end{pmatrix}$	$\begin{pmatrix}0.01\\1.98\end{pmatrix}$

- Why was benzene chosen? What would provide the correct vectors? Why are these vectors required and under what conditions do they need to appear to achieve sub-classical resolution power?

Critique: justification of hole mask configuration

- No justification given for why gold-coated S_3N_4 ion milled hole mask was chosen
  - Does this properly mimic how a molecule would be imaged?
- The size of the mask is of order 10μ, where as a benzene molecule is of order 10Å
  - Does the imaging process and algorithm scale across order of magnitude?



Conclusion - Authors

- Succeeded in reconstructing ("imaging") <u>2D</u> source structure
- Did not reach sub-classical diffraction limit (Abbe limit)
  - At high photon E, structure info. is more important
- Robust "imaging" method when scattered light's intensity & frequency fluctuate
  - Image w/x 1^{st} order coherence
  - Leads to possible future applications

Possible Future Applications

- Imperfect optics: distortion of coherent diffraction signals
  - Small λ
  - e.g.) hard X-ray (0.01~0.1 nm) 1st order coherence easily compromised
- Intense X-rays: emission, scattering incoherently
  - e.g.) single-particle imaging, macromolecular imaging from imperfect crystals
- Substituting Photons to:
  - → other bosons
  - → fermions: electron, neutron pulsed beams

Conclusion - Our Group

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- Breakthrough in extending the imaging technique: <u>1D → 2D</u>
- Reasonable explanations: sub-Abbe limit is less impt. in <u>high photon E region</u>
- Lots of possible <u>future applications</u> of imaging with incoherent sources

(-)

- A bit overstated/misleading abstract:
    - “image 2D objects close to or even below the Abbe limit”
- Experiment with vacuum UV not X-ray

Citation evaluation

Two papers have cited this paper since its publication according to CrossRef

- “DNA-based construction at the nanoscale”
  - A review of various techniques related to DNA construction
- “Ptychographical intensity interferometry imaging with incoherent light”
  - A different method of 2-D imaging with incoherent light