Ptychographic coherent diffractive imaging for attosecond pulses

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Attosecond science has been transforming our understanding of electron dynamics in atoms, molecules and solids. However, to date almost all of the attoscience experiments have been based on spectroscopic measurements because attosecond pulses have intrinsically very broad spectra due to the uncertainty principle and are incompatible with conventional imaging systems. Here we report a major advance towards achieving attosecond coherent diffractive imaging. Using simulated attosecond pulses, we simultaneously reconstruct the spectrum, 17 probes and 17 spectral images of extended objects from a set of ptychographic diffraction patterns. We further confirm the principle and feasibility of this method by successfully performing a ptychographic coherent diffractive imaging experiment using a light-emitting diode with a broad spectrum. We believe this work clear the way to an important unexplored domain of attosecond imaging science, which could have a far-reaching impact across different disciplines.
The first demonstration of attosecond pulses in 2001 has opened a new window to probe electron dynamics in atoms and molecules with unprecedented time resolution\textsuperscript{1-9}. With the continuing decrease of the temporal pulse duration and the increase of the photon energy range to the x-ray regime\textsuperscript{10-14}, the potential applications of attoscience could be even broader. However, attoscience experiments have been mostly limited to spectroscopic techniques due to the broad spectrum of the attosecond source\textsuperscript{3-5}. For an attosecond pulse, its energy bandwidth ($\Delta E$) and temporal pulse duration ($\Delta t$) are fundamentally set by the uncertainty principle ($\Delta E \cdot \Delta t \geq \hbar/2$). For example, the recent experimental demonstration of 53-attosecond soft x-ray pulses reaches the carbon K-absorption edge (284 eV) with $\Delta E/E \approx 100\%$\textsuperscript{13}. Such broad spectrum pulses cause severe chromatic aberration for any lens-based imaging systems. Chromatic aberration, first discovered by Newton more than 300 years ago\textsuperscript{15}, is a failure of a lens to focus all colors to the same focal spot due to the change of the refractive index of the lens with the wavelength of light. A classical method to overcome chromatic aberration is the use of achromatic lenses\textsuperscript{16}. But this method does not work for the full spectrum of electromagnetic radiation. For example, design and manufacture of achromatic lenses are extremely challenging in the x-ray regime, where lens design is difficult even in the monochromatic case\textsuperscript{17}. Here we develop a more general computational method based on coherent diffractive imaging (CDI), which can not only eliminate chromatic aberration across the full spectrum of electromagnetic radiation, but also take advantage of the broad spectrum to simultaneously reconstruct the spectrum, probes and images at 17 different wavelengths.

CDI is a lensless imaging or computational microscopy method, where the diffraction patterns of an object are first measured and then directly phased to obtain high-resolution images\textsuperscript{18}. Since the first experimental demonstration in 1999\textsuperscript{19}, various forms of CDI such as plane-wave
CDI, ptychography and Bragg CDI have been developed and applied to a broad range of samples in the physical and biological sciences using synchrotron radiation, x-ray free electron lasers, high harmonic generation, optical laser and electrons. With advanced computational algorithms, broadband CDI has also been developed to deal with the low temporal coherence of the illumination source. Ptychography, a powerful scanning CDI method, is particularly suitable for broadband imaging, which relies on collecting a series of diffraction patterns using a spatially confined probe to scan over an extensive object. By partially overlapping the probe between adjacent scan positions, advanced algorithms can reconstruct both the probe and the complex exit wave of the object. More recently, multimode and multiplex ptychographic methods have been developed to deal with broadband data. Here we make a significant advance to merge CDI with attosecond science for the first time, allowing the simultaneous reconstruction of 17 probes, 17 spectral images of an extended object and the spectrum of attosecond pulses with $\Delta E/E \approx 100\%$.

For attosecond light with a broad spectral bandwidth, its diffraction pattern is composed of an incoherent sum of diffracted intensity from all wavelengths,

$$I_j(k) = \int \left| \Psi_{\lambda,j}(k) \right|^2 d\lambda,$$

where $I_j(k)$ is the diffraction pattern of the $j^{th}$ scan position and $\Psi_{\lambda,j}(k)$ is the diffracted wave with wavelength $\lambda$. For simplicity, we ignore the constant in front of the integral of the equation. In practice we have to discretize Eq. (1), which introduces a discretization error. In addition, noise is present in the diffraction intensity of each wavelength. By introducing a new term, $N_{\lambda,j}(k)$, to absorb the discretization error and noise, we re-write Eq. (1),

$$I_j(k) = \sum_{\lambda=1}^{M} \left[ \left| \Psi_{\lambda,j,e}(k) \right|^2 + N_{\lambda,j}(k) \right] = \sum_{\lambda=1}^{M} \left[ \left| \Psi_{\lambda,j,e}(k) \right|^2 + \left| \Psi_{\lambda,j,g}(k) \right|^2 \right],$$

where $\Psi_{\lambda,j,e}(k)$ and $\Psi_{\lambda,j,g}(k)$ represent the electron and ground state amplitudes, respectively.
where $M$ is the number of spectral images to be reconstructed, $\Psi_{\lambda,j,e}(\vec{k})$ and $\Psi_{\lambda,j,g}(\vec{k})$ represent real and ghost modes of the diffracted wave, respectively. The introduction of ghost modes is to separate the signal from the error and noise. To simultaneously reconstruct the spectrum, probes and spectral images, we develop the Spectrum, Probe and Image Reconstruction (SPIR) algorithm by incorporating the ghost mode and probe replacement constraint, whereas probe replacement identifies the best probe among all the wavelengths in each iteration and uses it to refine the probes of other wavelengths. To implement SPIR, a set of diffraction patterns are first collected by performing a 2D scan of a confined probe relative to a sample with the probe at each position overlapped with its adjacent ones. The algorithm then iterates between real and reciprocal space (Supplementary text). In real space, the exit wave of the real and ghost modes at different wavelengths is obtained by multiplying probes by the object functions of the sample. For ghost modes, a 2D reflection transformation is applied to the coordinate of each object function. Taking the Fourier transform of the exit wave generates the diffracted wave of real and ghost modes. An updated diffracted wave is obtained by constraining it to the diffraction patterns. By applying the inverse Fourier transform, a new exit wave is created from the diffracted wave, which is used to create the next-iteration probes and object functions of real and ghost modes. To apply probe replacement, in each iteration the best probe is identified among all the wavelengths and is propagated back to construct the probes of other wavelengths. By incorporating ghost modes and probe replacement, SPIR can simultaneously reconstruct the spectrum and 17 probes and 17 spectral images. The algorithm is robust as it is not sensitive to the initial input and converges after several hundreds of iterations.

To validate the SPIR algorithm, we first performed numerical simulations using attosecond pulses with a spectrum ranging from 4.1 nm to 12.4 nm$^{13}$. The probe was confined by a 3-µm-
diameter pinhole that was placed 100 μm upstream of the sample and Fresnel propagated to the sample plane. Two samples were used in the simulations. The first is a resolution pattern, composed of 200-nm-thick aluminum structure with bar widths ranging from 30 nm to 1.2 μm. The second sample is a letter pattern with “atto” made of 200-nm-thick aluminum and “CDI” of 200-nm-thick boron. Boron was chosen because its K absorption edge (6.6 nm) is within the simulated spectrum, providing a contrast difference to assess the reconstruction quality of the spectrum and spectral images. Each sample was scanned in a randomly perturbed 2D raster grid scheme with 94% overlap between adjacent scan positions. A flux of $1 \times 10^7$ photons per scan position was used in the simulations and Poisson noise was added to the diffraction intensity. Each diffraction pattern was collected by a CCD camera, positioned 10 cm downstream of the sample to satisfy the oversampling requirement for all wavelengths. The quantum efficiency of the detector as a function of the spectrum was taken into account in each diffraction pattern. Figure 1a and b show a representative diffraction pattern from the resolution and letter pattern, respectively. The most noticeable feature of these broadband diffraction patterns is the absence of strong speckles that are presented in monochromatic diffraction patterns.

From the diffraction patterns, we used the SPIR algorithm to reconstruct the spectrum, probes and spectral images. All reconstructions consist of two runs of 250 iterations each. In the first run, the initial guesses of the probes and spectral images were binary masks and random arrays, respectively. The second run was initialized with new random arrays of the images while retaining the reconstructed probes from the first run. In the second run, only the images were allowed to update while the probes are fixed. In all reconstructions, we chose 17 probes and 17 spectral images that span the simulated spectrum in equal wavelength intervals. The number of probes and spectral images was heuristically chosen in a manner to reconstruct the spectrum with
high accuracy while also minimizing the crosstalk between adjacent images. Figure 1d-e show the reconstructed spectra (blue) of the resolution and letter pattern, which are in good agreement with the true ones (red). In comparison, a state-of-the-art broadband algorithm known as ptychographic information multiplexing (PIM) failed to reconstruct the spectra in both cases (Supplementary Fig. 1). Figure 2 shows the SPIR reconstruction of the probes and spectral images of the resolution pattern at three representative wavelengths (9.3 nm, 6.72 nm, 6.2 nm), in which the spatial resolution is increased with the decrease of the wavelength. The full reconstructions of 17 probes and 17 spectral images are shown in Supplementary Figs. 2a and 3a, respectively. SPIR faithfully reconstructed all the probes and spectral images except the first and last image due to the low incident flux at these two wavelengths (Fig. 1d). Figure 3, Supplementary Figs. 4a and 5a, and Video 1 show the reconstructed probes and spectral images of the letter pattern. While the absorption of aluminum is relatively flat across the spectrum, the K-edge of boron at 6.6 nm causes a jump in the absorption contrast of the “CDI” letters (Fig. 3). In comparison, PIM failed to reconstruct several probes and spectral images of both the resolution and letter patterns (Supplementary Figs. 2b, 3b, 4b and 5b). To account for the spectral instability of attosecond pulses, we conducted another simulation with a shot-to-shot spectral fluctuation of 10%. With all the other parameters kept the same, SPIR successfully reconstructed the average spectrum, probes and spectral images at 17 different wavelengths (Supplementary Figs. 6 and 7).

Next, we performed a broadband light-emitting diode (LED) experiment to validate the method. A collimated white LED was used to illuminate a test pattern with a 200 μm pinhole placed approximately 6 mm upstream of the sample to confine the probe. A CCD camera from Princeton Instruments was placed 26 cm downstream of the sample to collect diffraction patterns while fulfilling the oversampling requirement for all wavelengths of the LED. A field of view...
approximately 600 x 600 µm was scanned using a 2D raster grid with 94% overlap between adjacent probe positions. A small random offset was applied to the scan positions to avoid gridding artifacts in the reconstructions. Three exposures of different duration were collected at each scan position and merged to improve the dynamic range of the diffraction patterns. Diffraction patterns were cropped to be square and binned by a factor of 2 in each dimension to increase the signal-to-noise ratio and reduce the computation time. After two runs of 250 iterations each, SPIR reconstructed the spectrum, 17 probes and 17 spectral images from the diffraction patterns. The spectrum agrees with the experimental measurement using a spectrometer (Fig. 1f). Figure 4 shows three representative probes and spectral images at 662.5 nm, 550 nm and 437.5 nm. All 17 probes and images are shown in Supplementary Fig. 8. The successful reconstruction of the spectrum, probes and spectral images from the experimental diffraction patterns is consistent with the numerical simulation results and further corroborates the method.

In conclusion, we have developed a powerful algorithm (SPIR) by introducing two new constraints - ghost modes and probe replacement. Using a simulated attosecond source and an LED with a broad spectrum, we validated the SPIR algorithm and demonstrated the potential of achieving attosecond CDI by simultaneously reconstructing the spectrum, probes and spectral images at 17 different wavelengths. The significance of this work is threefold. First, our method is in principle applicable to any photon and electron source with a broad spectrum such as synchrotron radiation pink beams, HHG and electron sources, allowing the recovery of chemically specific contrast information without the use of monochromatic optics. Second, in this work we not only just overcome the broadband imaging obstacle, but also turn it into a strength by harnessing all the flux from a source. This would significantly reduce the data acquisition time in performing spectro-ptychography experiments, whereas the conventional method requires serial
repetition of ptychographic scans as a function of the energy. Finally, this work potentially unifies two important fields - attosecond science and CDI - into a single frame. As the spectrum of the current state-of-the-art attosecond sources extends to the x-ray regime\textsuperscript{13,14}, the ability to simultaneously reconstruct the spectrum, probes and images at multiple wavelengths could find broad application ranging from visualizing attosecond electron dynamics to imaging materials and biological samples at the nanometer scale with attosecond x-ray pulses.

References

1. Paul, P. M. Observation of a Train of Attosecond Pulses from High Harmonic Generation. *Science* **292**, 1689–1692 (2001).
2. Hentschel, M. *et al.* Attosecond metrology. *Nature* **414**, 509–513 (2001).
3. Krausz, F. & Ivanov, M. Attosecond physics. *Rev. Mod. Phys.* **81**, 163–234 (2009).
4. Corkum, P. B. & Krausz, F. Attosecond science. *Nature Phys.* **3**, 381–387 (2007).
5. Agostini, P. & DiMauro, L. F. The physics of attosecond light pulses. *Rep. Prog. Phys.* **67**, 1563–1563 (2004).
6. Baltuška, A. *et al.* Attosecond control of electronic processes by intense light fields. *Nature* **421**, 611 (2003).
7. Itatani, J. *et al.* Tomographic imaging of molecular orbitals. *Nature* **432**, 867–871 (2004).
8. Cavalieri, A. L. *et al.* Attosecond spectroscopy in condensed matter. *Nature* **449**, 1029–1032 (2007).
9. Sansone, G. *et al.* Electron localization following attosecond molecular photoionization. *Nature* **465**, 763 (2010).
10. Sansone, G. *et al.* Isolated Single-Cycle Attosecond Pulses. *Science* **314**, 443–446 (2006).
11. Kapteyn, H., Cohen, O., Christov, I. & Murnane, M. Harnessing Attosecond Science in the Quest for Coherent X-rays. *Science* **317**, 775 (2007).

12. Popmintchev, T. *et al.* Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers. *Science* **336**, 1287 (2012).

13. Li, J. *et al.* 53-attosecond X-ray pulses reach the carbon K-edge. *Nature Commun.* **8**, (2017).

14. Gaumnitz, T. *et al.* Streaking of 43-attosecond soft-X-ray pulses generated by a passively CEP-stable mid-infrared driver. *Opt. Express* **25**, 27506–27518 (2017).

15. Newton, I. *Opticks* (Royal Society, London, 1704).

16. Daumas, M. *Scientific Instruments of the Seventeenth and Eighteenth Centuries and Their Makers*. (Portman Books, London, 1989).

17. Chao, W., Harteneck, B. D., Liddle, J. A., Anderson, E. H. & Attwood, D. T. Soft X-ray microscopy at a spatial resolution better than 15 nm. *Nature* **435**, 1210 (2005).

18. Miao, J., Ishikawa, T., Robinson, I. K. & Murnane, M. Beyond crystallography: Diffractive imaging using coherent x-ray light sources. *Science* **7**, 545–549 (2015).

19. Miao, J., Charalambous, P., Kirz, J. & Sayre, D. Extending the methodology of X-ray crystallography to allow imaging of micrometre-sized non-crystalline specimens. *Nature* **400**, 342 (1999).

20. Chapman, H. N. *et al.* Femtosecond diffractive imaging with a soft-X-ray free-electron laser. *Nat. Phys.* **2**, 839–843 (2006)

21. Gaffney, K. J. & Chapman, H. N. Imaging Atomic Structure and Dynamics with Ultrafast X-ray Scattering. *Science* **316**, 1444 (2007).
22. Rodenburg, J. M. *et al.* Hard-X-Ray Lensless Imaging of Extended Objects. *Phys. Rev. Lett.* **98**, 034801 (2007).

23. Thibault, P. *et al.* High-resolution scanning X-ray diffraction microscopy. *Science* **321**, 379–382 (2008).

24. Nishino, Y., Takahashi, Y., Imamoto, N., Ishikawa, T. & Maeshima, K. Three-dimensional visualization of a human chromosome using coherent X-ray diffraction. *Phys. Rev. Lett.* **102**, 018101 (2009).

25. Robinson, I. & Harder, R. Coherent X-ray diffraction imaging of strain at the nanoscale. *Nature Mater.* **8**, 291–298 (2009).

26. Seibert, M. M. *et al.* Single mimivirus particles intercepted and imaged with an X-ray laser. *Nature* **470**, 78–81 (2011).

27. Zheng, G., Horstmeyer, R. & Yang, C. Wide-field, high-resolution Fourier ptychographic microscopy. *Nature Photon.* **7**, 739–745 (2013).

28. Gardner, D. F. *et al.* Subwavelength coherent imaging of periodic samples using a 13.5 nm tabletop high-harmonic light source. *Nature Photon.* **11**, 259–263 (2017).

29. Holler, M. *et al.* High-resolution non-destructive three-dimensional imaging of integrated circuits. *Nature* **543**, 402–406 (2017).

30. Jiang, Y. *et al.* Electron ptychography of 2D materials to deep sub-Ångström resolution. *Nature* **559**, 343–349 (2018).

31. F. Pfeiffer, X-ray ptychography. *Nature Photon.* **12**, 9–17 (2018).

32. Whitehead, L. W. *et al.* Diffractive Imaging Using Partially Coherent X Rays. *Phys. Rev. Lett.* **103**, (2009).
33. Abbey, B. et al. Lensless imaging using broadband X-ray sources. *Nature Photon.* **5**, 420–424 (2011).

34. Witte, S., Tenner, V. T., Noom, D. W. & Eikema, K. S. Lensless diffractive imaging with ultra-broadband table-top sources: from infrared to extreme-ultraviolet wavelengths. *Light Sci. Appl.* **3**, e163 (2014).

35. Thibault, P. & Menzel, A. Reconstructing state mixtures from diffraction measurements. *Nature* **494**, 68–71 (2013).

36. Batey, D. J., Claus, D. & Rodenburg, J. M. Information multiplexing in ptychography. *Ultramicroscopy* **138**, 13–21 (2014).

37. Enders, B. *Development and Application of Decoherence Models in Ptychographic Diffraction Imaging.* (Technical University of Munich, 2015).

38. Zhang, B. et al. Ptychographic hyperspectral spectromicroscopy with an extreme ultraviolet high harmonic comb. *Opt. Express* **24**, 18745 (2016).

39. Chen, B. K., Sidorenko, P., Lahav, O., Peleg, O. & Cohen, O. Multiplexed single-shot ptychography. *Opt. Lett.* **43**, 5379–5382 (2018).

40. Miao, J., Sayre, D. & Chapman, H. N. Phase retrieval from the magnitude of the Fourier transforms of nonperiodic objects. *J. Opt. Soc. Am. A.* **15**, 1662 (1998).

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Figure 1. Representative diffraction patterns and reconstructed spectra by SPIR. **a** and **b**, Representative diffraction patterns measured from a resolution and letter pattern, respectively, using simulated attosecond pulses. **c**, Representative diffraction pattern of a test pattern using an LED. **d**-**f**, Reconstructed spectra (in blue) of the resolution and letter pattern with simulated attosecond pulses and of the test pattern with the LED, respectively, where the true spectra are in red. The true spectrum of the LED in (**f**) was measured by a spectrometer.
Figure 2. Probe and spectral image reconstructions of a resolution pattern with simulated attosecond pulses. 

a-c, The structure of the resolution pattern at 9.3 nm, 6.72 nm and 6.2 nm, respectively. 

d-i, Three representative probes and spectral images at different wavelengths reconstructed by SPIR, respectively, where the spatial resolution is increased with the decrease of the wavelength. The full 17 probes and 17 spectral images are shown in Supplementary Figs. 2a and 3a, respectively. Scale bar, 2 μm.
Figure 3. Probe and spectral image reconstructions of a letter pattern with simulated attosecond pulses. a-c, Absorption contrast images of the letter pattern at 7.75 nm, 6.72 nm and 6.2 nm, respectively, where the K-edge of boron is at 6.6 nm. d-i, Three representative probes and spectral images at different wavelengths reconstructed by SPIR, respectively, where the image contrast of the “CDI” letters changes across the absorption edge. The full 17 probes and 17 spectral images are shown in Supplementary Figs. 4a and 5a, respectively. Scale bar, 2 μm.
Figure 4. Probe and spectral image reconstructions of a test pattern from a broadband LED experiment. a-f, Three representative probes and spectral images reconstructed by SPIR at 662.5 nm, 550 nm and 437.5 nm, respectively. The full 17 probes and 17 spectral images are shown in Supplementary Fig. 8. Scale bar, 200 μm.
Supplementary Text

The SPIR algorithm with ghost mode and probe replacement constraints

SPIR iterates between real and reciprocal space. The $i^{th}$ iteration exit wave of the real and ghost mode, $\psi_{\lambda,j,e}^i(\vec{r})$ and $\psi_{\lambda,j,g}^i(\vec{r})$, is calculated by

$$
\begin{pmatrix}
\psi_{\lambda,j,e}^i(\vec{r}) \\
\psi_{\lambda,j,g}^i(\vec{r})
\end{pmatrix} =
\begin{pmatrix}
P_{\lambda,e}^i(\vec{r}) \\
P_{\lambda,g}^i(\vec{r})
\end{pmatrix}
\cdot
\begin{pmatrix}
O_{\lambda,j,e}^i(\vec{r}) \\
O_{\lambda,j,g}^i(\vec{T}\vec{r})
\end{pmatrix},
$$

where $i$ and $j$ index the $i^{th}$ iteration and $j^{th}$ scan position, $P_{\lambda,e}^i(\vec{r})$ and $P_{\lambda,g}^i(\vec{r})$ are the probes, $O_{\lambda,j,e}^i(\vec{r})$ and $O_{\lambda,j,g}^i(\vec{T}\vec{r})$ are the object function of the sample with the subscripts $e$ and $g$ representing the real and ghost modes, respectively, and $T$ is a transformation matrix. After testing several different types of the transformation matrix, we found a 2D coordinate reflection matrix works the best in this work. By taking the Fourier transform ($\mathcal{F}$) of the exit wave, we obtain the diffracted wave of the real and ghost modes,

$$
\begin{pmatrix}
\psi_{\lambda,j,e}^i(\vec{k}) \\
\psi_{\lambda,j,g}^i(\vec{k})
\end{pmatrix} = \mathcal{F}
\begin{pmatrix}
\psi_{\lambda,j,e}^i(\vec{r}) \\
\psi_{\lambda,j,g}^i(\vec{r})
\end{pmatrix}.
$$

The diffracted wave is updated by being constrained to the measured intensity, $I_j(\vec{k})$.

$$
\begin{pmatrix}
\psi_{\lambda,j,e}^{i+1}(\vec{k}) \\
\psi_{\lambda,j,g}^{i+1}(\vec{k})
\end{pmatrix} = \frac{\sqrt{I_j(\vec{k})}}{\sqrt{\sum_{k=1}^{M} |\psi_{\lambda,j,e}^i(\vec{k})|^2 + |\psi_{\lambda,j,g}^i(\vec{k})|^2}}
\begin{pmatrix}
\psi_{\lambda,j,e}^i(\vec{k}) \\
\psi_{\lambda,j,g}^i(\vec{k})
\end{pmatrix}.
$$

The corresponding exit wave of the real and ghost modes is calculated by

$$
\begin{pmatrix}
\psi_{\lambda,j,e}^{i+1}(\vec{r}) \\
\psi_{\lambda,j,g}^{i+1}(\vec{r})
\end{pmatrix} = \mathcal{F}^{-1}
\begin{pmatrix}
\psi_{\lambda,j,e}^i(\vec{k}) \\
\psi_{\lambda,j,g}^i(\vec{k})
\end{pmatrix},
$$

where $\mathcal{F}^{-1}$ is the inverse Fourier transform. The probe and object function of the $(i+1)^{th}$ iteration are updated by

$$
P_{\lambda,e}^{i+1}(\vec{r}) = P_{\lambda,e}^i(\vec{r}) + \beta_1 \cdot \frac{O_{\lambda,j,e}^i(\vec{r})}{|O_{\lambda,j,e}^i(\vec{r})|_{max}^2} \left[ \psi_{\lambda,j,e}^{i+1}(\vec{r}) - \psi_{\lambda,j,e}^i(\vec{r}) \right]
$$

and

$$
O_{\lambda,e}^{i+1}(\vec{r}) = O_{\lambda,e}^i(\vec{r}) + \beta_1 \cdot \frac{P_{\lambda,j,e}^i(\vec{r})}{|P_{\lambda,j,e}^i(\vec{r})|_{max}^2} \left[ \psi_{\lambda,j,e}^{i+1}(\vec{r}) - \psi_{\lambda,j,e}^i(\vec{r}) \right]
$$

where $\beta_1$ is set to 0.9. Equations (5) and (6) are also used to calculate the $(i+1)^{th}$ iteration probe and object function of the ghost mode by replacing the subscript $e$ with $g$.

Next we implement the probe replacement constraint into the SPIR algorithm, which takes into account the wavelength-dependent propagation of the probe. In the $i^{th}$ iteration, we first locate the best probe, $P_{\lambda^*,e}^{i+1}(\vec{r})$, among all the reconstructed probes, where $\lambda^*$ represents the wavelength
of the best probe. This probe is Fresnel propagated back to the pinhole plane. The best probe in the pinhole plane is then Fresnel propagated to the sample plane at other wavelengths ($\lambda'$), defined as $P_{\lambda',e}^{i+1}(\vec{r})$. The $(i+1)^{th}$ iteration probe is further updated by

$$
P_{\lambda,e}^{i+1}(\vec{r}) = \begin{cases} 
P_{\lambda',e}^{i+1}(\vec{r}) + \beta_2 \left( P_{\lambda',e}^{i+1}(\vec{r}) - P_{\lambda,e}^{i+1}(\vec{r}) \right) & \lambda = \lambda' \\
p_{\lambda,e}^{i+1}(\vec{r}) & \lambda = \lambda^* \end{cases}$$

(7)

where $\beta_2$ is set to 0.1. Eq. (7) is also used to calculate the probe of the ghost mode by replacing the subscript $e$ with $g$. The updated probe and object function of the real and ghost modes from Eqs. (6) and (7) are used for the next iteration.

**Spectrum recovery**

We recover the spectrum from the exit wave of the real mode,

$$
\psi_{\lambda,j,e}(\vec{r}) = O_{\lambda,j,e}(\vec{r}) P_{\lambda}(\vec{r}) = \left[ \frac{1}{c_\lambda} O_{\lambda,j,e}(\vec{r}) \right] \left[ c_\lambda P_{\lambda,e}(\vec{r}) \right]
$$

(8)

where $\psi_{\lambda,j,e}(\vec{r})$ and $O_{\lambda,j,e}(\vec{r})$ are the reconstructed exit wave and object function of the sample, $P_{\lambda}(\vec{r})$ is the reconstructed probe at wavelength $\lambda$, and $c_\lambda$ is a wavelength-dependent constant. As the exit wave is constrained to the measured intensity by Eq. (3), it can be uniquely reconstructed by SPIR, but the probe and object function are not unique up to a multiplicative constant (Eq. (8)). Thus we use the reconstructed exit wave instead of the probe to recover the spectrum,

$$
S_{\lambda,j} = \sum_{x,y} |O_{\lambda,j,e}(x,y) P_{\lambda}(x,y)|^2
$$

(9)

where $S_{\lambda,j}$ is the spectrum of the $j^{th}$ scan position and $\vec{r} = (x,y)$. By choosing an empty region in the sample and averaging the spectra of all the scan positions in the region, we obtain the final recovered spectrum, $S_{\lambda}$, which removes the effect of object function on the spectrum recovery. By incorporating the ghost mode and probe replacement constraints, SPIR reconstructs more accurate probes across the entire spectrum than other algorithms$^{35-39}$, which leads to more accurate spectrum recovery.
Supplementary Fig. 1. Reconstructed spectra of the resolution (a) and letter pattern (b) with simulated attosecond pulses and of the test pattern with a LED experiment (c). The true spectrum is in red and the spectra reconstructed by PIM and SPIR are in blue and green, respectively. The true spectrum of the LED was measured by a spectrometer.

Supplementary Fig. 2. SPIR and PIM reconstruction of the probes for the resolution pattern using simulated attosecond pulses. (a) 17 probes reconstructed by SPIR that span the simulated spectrum in equal wavelength intervals, with good recovery across the whole spectrum. (b) 17 probes reconstructed by PIM, exhibiting artifacts in the reconstruction at several wavelengths.
Supplementary Fig. 3. SPIR and PIM reconstruction of the spectral images of the resolution pattern using simulated attosecond pulses. (a) 17 spectral images reconstructed by SPIR. The first and last images were not successfully reconstructed due to the low incident flux at these two wavelengths. (b) 17 spectral images reconstructed by PIM, where the poorly reconstructed images are innately linked to the poorly reconstructed probes (Supplementary Fig. 2b). Scale bar, 2 μm.
Supplementary Fig. 4. SPIR and PIM reconstruction of the probes for the letter pattern using simulated attosecond pulses. (a) 17 probes reconstructed by SPIR that span the simulated spectrum in equal wavelength intervals. (b) 17 probes reconstructed by PIM, exhibiting significant artifacts in the reconstruction.
Supplementary Fig. 5. SPIR and PIM reconstruction of the spectral images of the letter pattern using simulated attosecond pulses. (a) 17 spectral images reconstructed by SPIR, where the boron K-absorption edge is at 6.6 nm (between modes 12 and 13). (b) 17 spectral images reconstructed by PIM, in which 10 out of 17 modes were poorly reconstructed. Scale bar, 2 µm.
Supplementary Fig. 6. SPIR reconstruction of the spectra with a shot-to-shot spectral fluctuation of 10%. (a) Reconstructed spectra from all the scan positions for the resolution pattern. The fluctuation of the reconstructed spectra is consistent with that used to calculate the ptychographic diffraction patterns. (b) Reconstructed spectrum (blue) by averaging all the spectra in (a), which is in good agreement with the average true spectrum (red).

Supplementary Fig. 7. SPIR probe and spectral image reconstruction of the resolution pattern with a shot-to-shot spectral fluctuation of 10%. The reconstructed probes (a) and spectral images (b) at 17 different wavelengths.
Supplementary Fig. 8. SPIR reconstruction of 17 probes (a) and spectral images (b) of a test pattern from a broadband LED experiment. Scale bar, 200 μm.

Supplementary Video 1. SPIR reconstruction of 17 spectral images of the letter pattern using simulated attosecond pulses, where the boron K-absorption edge is at 6.6 nm.

References

41. Maiden, A. M. & Rodenburg, J. M. An improved psychogaphical phase retrieval algorithm for diffractive imaging. Ultramicroscopy 109, 1256–1262 (2009).