A symmetry-derived mechanism for atomic resolution imaging

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We introduce a new image contrast mechanism for scanning transmission electron microscopy that derives from the local symmetry within the specimen. For a given position of the electron probe on the specimen, the image intensity is determined by the degree of similarity between the exit electron intensity distribution and a chosen symmetry operation applied to that distribution. The contrast is robust with respect to specimen thickness, electron probe energy and defocus, and it reveals atomic positions with a precision that can significantly exceed that defined by the resolving aperture.

In scanning transmission electron microscopy (STEM), images can be generated by scanning an electron beam across the object and, at each position of the electron beam, detecting the scattered electron intensity distribution after transmission through the specimen. The most common STEM imaging modes integrate the scattered intensity in the diffraction plane across a particular angular range, using either disc or annular detector geometries, to generate phase-contrast Bright Field (BF) and adsorptive-contrast High Angle Annular Dark Field (HAADF) images, respectively, (or a mix of phase and adsorptive contrast in the case of Annular Bright Field (ABF)). Recently, the advent of fast read-out, high dynamic range detectors [1–6] has enabled the full angular distribution of scattered intensity to be recorded at each beam position. This represents a revolution in STEM, providing access to a vast and rich palate of additional specimen information. Fast detectors have already been applied, for example, to improve STEM spatial resolution using ptychography [7–10], to map electric [11, 12] and magnetic fields [4, 13–15], strain [16–18], polarization domains [19], and octahedral tilts [20], representing just the beginning of this powerful new era in STEM.

Here we propose a new image contrast mechanism for atomic resolution STEM based on a measurement of the degree of symmetry in the scattered intensity distribution at each point of a scan: Symmetry STEM (S-STEM). By virtue of the strong electron-specimen interaction and resultant dynamical scattering, the symmetry of the illuminated specimen volume is encoded in the symmetry of the scattered intensity distribution, independent of the specimen thickness and accelerating voltage [21–23]. In this paper, the scattered intensity distribution is in the form of a convergent beam electron diffraction (CBED) pattern [Fig. 1(a)], the most common case for STEM, but the approach can be applied in principle to any form of scattered intensity distribution in any optical plane and also to other scanning microscopy techniques.

The ‘degree’ of symmetry in a pattern can be analysed by a comparison of the scattered intensity distribution with itself after an applied symmetry operation [24]. For a given two dimensional (2D) pattern A, the symmetry

![Diagram](image-url)

FIG. 1. Schematic of a Symmetry STEM experiment. (a) An atomic-scale electron probe scans the specimen. A convergent beam electron diffraction pattern is imaged in the far field by a fast pixelated detector (green box) for each point of the scan. (b) Grid of simulated CBED patterns resulting from a scan across a Ce atomic column (red box) in a CeB$_6$ crystal. Each CBED pattern is arranged according to the position of the electron probe in real space, scan step size is 20 picometres. (c) CBED patterns for the probe on the centre of the Ce column (middle figure) and shifted 20 picometres either side of the centre (but still on the atomic column) (top and bottom figures), showing the rapid change in pattern symmetry (4mm to m).
intensity $I$ is given by:

$$I = \max \left[ A \ast \text{symmetry operation} (A) \right],$$

(1)

where $\ast$ is a normalised cross-correlation and the symmetry operation can be chosen (for example, a rotation or a mirror). If $A$ is invariant under the symmetry operation, then the intensity will be maximum, $I = 1$, and $I < 1$ if the symmetry is not matched [25].

In the case of Symmetry STEM, the intensity will be calculated from each CBED pattern at each point of a 2D scan, $(x, y)$, which can be plotted as an image $I = I_{x,y}$. Each CBED pattern resolves the electron distribution in reciprocal space at a particular point, $(x, y)$, of a scan $A = A_{x,y}(k_x, k_y)$, denoting one point in a so called four dimensional (4D) STEM dataset [26]. Similar algorithms have been applied in biological imaging [27, 28] and also in measurements of local polarization domains [29, 30].

Data processing was based on methodology introduced in [13, 31, 32] and implemented in GPU accelerated ArrayFire library [33].

The application of equation 1 to this dataset generates an entirely different image contrast mechanism, neither phase-contrast nor adsorption-contrast, which provides access to new specimen information at the atomic level.

The sensitivity to local symmetry that underpins the Symmetry STEM contrast mechanism is illustrated with a STEM simulation. Figure 1(b) shows an array of simulated CBED patterns corresponding to a 0.5 Å FWHM probe scanning across the Ce column in (100) oriented CeB$_6$. Each simulated CBED pattern is arranged according to the corresponding real space position, $(x, y)$, of the probe, with the Ce column position located at the centre of the array. The GPU accelerated parallel implementation of the multislice simulations were performed using the Prismatic software package [34–36] using parameters given in [37]. The arranged CBED patterns give a clear sense of how the symmetry changes as the probe is scanned across the Ce atomic column in 0.2 Å steps [Fig. 1(c)]. For example, there is an immediate shift from 4-fold and multiple mirrors (4mm) to a single mirror symmetry (m) as the probe centre moves from the absolute centre of the atomic column to just 20 picometres off-centre (but is nevertheless still located on the atomic column). This highlights the acute sensitivity to local specimen symmetry that is delivered by dynamical scattering [22, 23, 38] and forms the basis of image contrast in Symmetry STEM.

Equation 1 provides an extremely efficient method for distilling the local symmetry information present in the pattern. This is illustrated generically in Fig. 2 for an intensity distribution, $A_p$, in the form of a palm tree.

Two classes of symmetry are tested, a rotation and mirror symmetry. These symmetry operations can be tested for an arbitrary angle on this arbitrary pattern, $A_p$ [e.g. Figs 2(a) and 2(b), test a 20° rotation for the palm tree pattern]. The cross-correlation is calculated for 0° to 360° [Fig. 2(c)]. It can be seen that even if the pattern does not possess a perfect symmetry element, there still exist local maxima in the analysis. For the palm tree pattern this signal arises when the leaves are overlapping after application of a given symmetry operation. [25]

To demonstrate Symmetry STEM, the symmetry analysis of Eq. 1 will now be applied to the simulated scanning CBED dataset [Fig. 1] across a field of view slightly larger than a CeB$_6$ unit cell [Fig. 3(a)], again using the parameters in [37]. The size of the dataset was 2.5 GB and included 124x124 probe positions. Symmetry STEM images corresponding to 180° rotation, 90° mirror and 1° rotation are generated [Figs 3(b)-(d), respectively]. For comparison, standard STEM BF, ABF and HAADF images were also reconstructed [Figs 3(e)-(g), respectively] by integrating across the angular ranges indicated in [Fig. 3(h)] for each probe position.

As anticipated, the S-STEM images exhibit atomic-scale contrast, revealing local maxima wherever some degree of the applied symmetry element is present, reaching a maximum value near one when there is an identity, such as on the Ce column at 180° rotation and 90° mirror. 180° rotation symmetry shows exceptionally intense and sharp contrast for Ce, small local maxima at all B positions, and also it highlights the 180° symmetry with a broad maxima half way between Ce atomic columns along (100) [Fig. 3(b)]. 90° mirror symmetry shows bright contrast where mirror planes within the lattice match the symmetry [Fig. 3(c)]. An interesting contrast arises when a small rotation symmetry of 1° is measured [Fig. 3(d)] giving extra sensitivity to the rate of change of the local symmetry. Strong ‘intensity’ appears where the CBED pattern varies slowly with angle $(k_x, k_y)$, namely at the position of asymmetric B atom sites, with weak intensity at the Ce sites where the pattern varies rapidly.

Importantly, local maxima at atomic sites in the Symmetry STEM images are exceptionally sharp with low intensity ‘moats’ around them. This is particularly the case...
for the peaks at Ce columns with a FWHM of \( \sim 0.25 \text{ Å} \) [Fig. 3(i)]. This greatly exceeds the diffraction limit defined by the numerical aperture of the lens, namely 0.8 Å (FWHM of idealised probe 0.5 Å), and the corresponding resolution of conventional BF, ABF and HAADF STEM (for 15 mrad at 300 kV) [Fig 3(i)].

Local maxima due to the presence of a symmetry element but in the absence of an atomic column do not show the ‘moat’ because the rate of change of specimen symmetry is more slowly varying than in the presence of an atomic column. This enables atom sites to be distinguished from atom-free symmetry sites. This distinction can be further checked when S-STEM images derived from different symmetry elements are compared.

We examine further the imaging properties of Symmetry STEM by calculating a line scan over Ce–B–Ce atomic columns in \{100\} CeB₆ for different defocus and probe size and sample thickness and tilt [Fig. 4]. The symmetry element is chosen to be a 180° rotation and the base parameters are as specified in [37] and kept constant unless otherwise noted.

**Dependence on Thickness and Defocus:** As S-STEM probes local symmetry within the sample, there is a no change of contrast at the centre of atomic sites due to the change of defocus nor thickness [Figs 4(a) and (b) respectively]. This is a consequence of the fact that, while the scattered intensity distribution can vary rapidly with thickness, accelerating voltage and defocus, its symmetry remains invariant. This is a great advantage over standard methods for which the sign and magnitude of the signal can change. For all thicknesses, the intensity peaks at atomic sites are an almost constant, extremely narrow width (\( \sim 0.25 \text{ Å} \)), with some local variations in the vicinity of the B octahedra likely due to scattering onto nearby high symmetry sites. Nevertheless, even in the presence of this “cross-talk”, the narrow peak persists [inset Fig. 4(b)]

**Resolution and Probe size:** The ability to resolve two features in S-STEM depends on the ability of the probe to detect a symmetry change. In other words, it depends on the probe size relative to the rate of change with position of the symmetry of the local specimen potential. The resolution of Symmetry STEM images is therefore only a function of the electron-optical imaging system in so far as this system defines a probe size (which is set by the collective effect of the spatial coherence function, probe-forming aberrations and aperture size [39]). Beyond this, it is not meaningful to describe resolution in S-STEM in terms of concepts used in traditional optics.

In the aberration-free, spatially coherent calculations, the convergence semi-angle defines the probe size and was varied from 0.5–40 mrad [Fig. 4(c)]. The signal is constant and identity at \(< 4 \text{ mrad} (\sim 2.4 \text{ Å probe FWHM}) because the probe is greater than the unit cell in this perfect crystal, so no change in symmetry can be detected. Put another way, the CBED patterns have non-overlapping CBED discs and hence do not change with position.

From the calculations, it can be seen that when the convergence semi-angle generates a probe FWHM comparable to the “non-bonded” atomic radius (>4 mrad; >2.4 Å probe) we start to resolve clearly the two Ce columns. With higher convergence, the probe FWHM
approaches half the non-bonded atomic radius (∼7 mrad; ∼1.7 Å probe) and the image peaks sharpen significantly to ∼0.3Å FWHM, increasing the resolution well beyond the diffraction limit set by this convergence angle [Fig. 4(c)]. Interestingly, around 22 mrad, where the probe FWHM is 0.44 Å, less than a quarter of the atomic radius there is a step-function reduction in peak width to <0.085 Å, beating the “diffraction limit” defined by the probe-forming aperture by a factor of 5. The sharpness of the peak reduces for convergence angles larger than ∼25 mrad, possibly due to scattering onto nearby atomic sites promoted by the larger transverse momentum of the incident probe.

Dependence on Tilt: Tilt of a sample is a crucial parameter to study as it changes the excitation errors and hence the symmetry of the scattering matrix and resulting CBED pattern. The sample was tilted from the (100) zone axis by −1° to 3° in the plane of the line scan [Fig. 4(d)]. When the sample is tilted more than 0.05° the symmetry peak related to Ce starts to disappear. This can be an advantage of Symmetry STEM, as it allows for an extremely sensitive tilt calibration, beyond that easily detectable by eye in the corresponding diffraction pattern and with minimum problems due to the defocus change with tilt [Fig. 4(a)]. Even when the tilt is such that the symmetry peak is lost, the presence of the atomic column is highlighted by a symmetry break (dark areas present at the location of the Ce). Hence, the Symmetry STEM algorithm could potentially be automated in the detector hardware (something analogous to [40]) to provide an easy, on-line and ultrahigh precision alignment of the specimen for any imaging or spectroscopy experiment in STEM.

Finally, we compare the Symmetry STEM analysis of simulated data with experimental data from CeB6 [Fig. 5]. The scanned CBED data was collected at 300 kV on an early generation double-spherical aberration corrected FEI Titan3 80–300 FEGTEM equipped with a pixelated EMPAD detector [4]. Aberrations were largely corrected within the convergence semi-angle of 15 mrad. Conventional BF and ABF STEM images were reconstructed [Fig. 5(a) and (b), respectively] to compare with the Symmetry STEM signals with the same symmetry operations as applied in Fig. 3 [Fig. 5]. (The HAADF signal was not collected here because the large angular field of view required would constrain the symmetry measurement from the central disk area.) The 1° and 180° rotation images [Figs 5(d) and (e), respectively], show contrast closely related to the theoretical calculations [Fig. 3]. In particular, the 180° rotation image shows extremely sharp peaks surrounded by dark ‘moats’, corresponding to the symmetry maxima when the probe is positioned at the absolute centre of the Ce atomic column and the break of symmetry as soon as the beam shifts slightly from the centre but remains on the column, as seen in the calculations [Fig. 3(f)]. We can also see the symmetric position of the central B atomic column, however the asymmetric sites of other B atomic positions are not as clear, most likely due to imperfect instrument and specimen stability and a lack of local 180° rotation symmetry in the position. The 1° rotation image shows very strong signal to noise at the Ce columns and also shows some residual specimen tilt effects. An average unit cell image is compared with the calculated image in Fig. 5(f).

In summary, in Symmetry STEM, the symmetry of the local specimen potential defines the mathematical symmetry of the dynamical N-dimensional scattering matrix [22, 23] and this, in turn, defines the symmetry of the scattered intensity distribution (CBED pattern) which is extracted using equation 1. This very different contrast mechanism can generate images with exceptional spatial resolution (several times better than conventional diffraction-limited STEM techniques). For perfect crystals, a reduction in symmetry can be detected as soon as an atomic-scale probe shifts a few picometres from the centre of an atomic column, generating extremely sharp image peaks and enabling the identification of atomic positions with extraordinary precision. In experimental data, the practical limit is the stability of the instrument and specimen. The proof-of-concept examples in Fig. 5 were taken on a decade-old instrument. The ongoing improvements in instrument stability bode well for the further development of this technique. The remarkable robustness of S-STEM images to thickness, accelerating voltage and defocus is because the mathematical symmetry of the scattering matrix and hence the CBED

![Figure 5](image_url)
pattern symmetry does not depend on these quantities. The acute sensitivity to tilt is because it does. The latter provides an opportunity for an automated, high precision tilt alignment. The potential of Symmetry STEM to obtain atomic resolution images of light and heavy atoms, from thick and thin crystals across a wide selection of accelerating voltages opens up a range of applications in material science that are otherwise challenging to image, including thick and beam sensitive specimens. The ability to image defects due to the change in symmetry they induce, also opens new opportunities, including the imaging of dopant atoms and dislocations. The method could also be applied to the imaging of atomic magnetic fields in electromagnetic circular dichroism [41]. Furthermore, the precision in locating atomic positions delivered by the exceptionally sharp image peaks offers the potential for the accurate measurement of strain, octahedral tilts and other types of atomic displacements.

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Supplemental material: A symmetry-derived mechanism for atomic resolution imaging

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Additional characteristics of Symmetry STEM

Intensity $I$ in S-STEM is defined by:

$$I = \max \left[ A * \text{symmetry operation} (A) \right], \quad (1)$$

where $*$ is a normalised cross-correlation and the symmetry operation can be chosen (a rotation or a mirror). If $A$ is invariant under the symmetry operation, then the intensity will be maximum, $I = 1$, and $I < 1$ if symmetry is not matched. This operation is applied to the recorded scattered electron intensity for each point of a scan. In calculating S-STEM images the following points are worth noting:

- Unlike traditional STEM, in S-STEM the CBED pattern does not need to be centred on the detector, because the intensity is based on a maximum value of the cross-correlation and it is therefore shift invariant \cite{1}.

- Only $0^\circ$ to $180^\circ$ angles need to be calculated because only the relative rotation matters. $1^\circ$ clockwise and $1^\circ$ anti-clockwise rotations have the same maximum of cross-correlation (ignoring interpolation errors).

- The result of $0^\circ$ rotation will always be $I = 1$ due to Eq. 1 becoming an auto-correlation. Due to the presence of noise in any real image and due to interpolation errors in the image rotation algorithm, the symmetry value will reach $I = 1$ in only very specific cases. Contrast value will be $I = 1$ if image rotation is 4-fold and image has the same symmetry without presence of any noise. This is due to the discrete nature of pixelated images.

Animation of rotation and mirror symmetry

Animations S1 and S2 show experimental S-STEM images for rotation and mirror symmetries between angles $1^\circ$ to $180^\circ$ for CeB$_6$ specimen. The $1^\circ$ is used as a start because $0^\circ$ rotation is an identity with intensity equal $I = 1$ for the whole image. The field of view is $\sim 57 \times 57 \text{Å}$. The convergence semi-angle was 15 mrad and acceleration voltage 300 kV.

Additional experimental example - wedge-shaped sample

In a wedge-shaped sample, the mirror symmetry in the CBED pattern will be broken perpendicular to the thickness gradient due to the change in the number of atoms per atomic column [Fig. 1]. This has potential for counting the number of atoms in the atomic columns with high precision.

FIG. 1. Mirror Symmetry STEM image from an edge of the wedge-shaped CeB$_6$ sample. Inset shows that Symmetry STEM detects the break in symmetry due to the thickness gradient. The text shows details about thickness variation and the orientation of the chosen mirror symmetry used in the analysis. The convergence semi-angle was 15 mrad and acceleration voltage 300 kV.

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