Image simulation in high resolution transmission electron microscopy considering atom as an electrostatic interferometer

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Abstract
An alternative approach to image simulation in high resolution transmission electron microscopy (HRTEM) is introduced after a comparative analysis of the existing image simulation methods. The alternative method is based on considering the atom center as an electrostatic interferometer akin to the conventional off-axis electron biprism within few nanometers of focus variation. Simulation results are compared with the experimental images of 2D materials of MoS2, BN recorded under the optimum combination of third order spherical aberration $C_3 = -35 \mu m$ and defocus $\Delta f = 1, 4, 8$ nm and are found to be in good agreement.

1. Introduction
Imaging of any object both in transmission and reflection geometry is generally carried out by detecting the scattered (incoherent) and diffracted (coherent) radiation on a recording device, e.g., a camera placed at different reference planes away from the object on the optic axis. Image is the replica of the object and not the object itself, and the information about the object is carried to the detector through the complex wave function [1–5]. Maximum spatial details that can be obtained are limited by the diffraction and the microscope performance. The entire topic of quantitative HRTEM falls into two broad categories: (i) object exit wave (OEW) reconstruction to retrieve the missing phase information from the recorded image, and (ii) image simulation to interpret the OEW with the object structure. Various schemes are available for the reconstruction of the OEW function to recover the phase related to the object potential and the crystallography and will not be discussed here, for details see [6–8]. There are several aspects in HRTEM image simulation that need to be considered, e.g., probe electron, interaction between the fast electron and the specimen potential, lens action, and characteristics of the recording device [5, 9]. The probe illumination is typically a plane wave of electrons with relativistic energy in the range of 100–300 kV ($\lambda = 1.97$ pm at 300 kV). The amplitude $A(x, y)$ and the phase $\phi(x, y)$ of the OEW function of the form $\psi = Ae^{i\phi}$ extracted from the recorded intensity pattern are used to interpret the potential information of atoms in solid. The information on the object potential can be used to extract wide range of information such as number of atoms or thickness along the beam propagation direction, identification of atoms, valence electron sharing between the atoms etc [7, 10–14]. However, the change in phase ($\phi$) of the probe electron wave after interaction with the specimen potential and resulting modulation in intensity pattern has been treated in fundamentally distinctive ways, e.g., (i) transmission function based on weak phase object approximation (WPOA) along with Zernike type $\pi/2$ or $\lambda/4$ phase plate equivalent to phase contrast transfer function (PCTF) to account for the lens aberration, where the phase change is incorporated in terms of change in magnitude of momentum vector $k$ of the probe electron due to specimen potential, [4, 5] (ii) phase change according to scattering amplitude in terms of atom scattering $f(k)$ and structure factor $F(q)$ along with PCTF [9, 15, 16], and (iii) self-interference in HRTEM and holographic fringe shift in off-axis electron holography where a phase term ($\pm \phi$) is added inside the trigonometric functions with respect to the reference phase [17, 18]. Kindly note that the phase change due to aberration through PCTF is not added with the object wave.
rather it is applied as a frequency filter and point spread function (psf) in the diffraction and image planes, respectively. Therefore, in the present manuscript, at first, a comparative analysis is provided on the various existing methods for simulating the image of the atom. It is shown that the different ways of considering phase change in the probe electron wave function due to atomic potential result in different magnitude and intensity patterns for the same atom. Subsequently, an alternative method is introduced where the geometry of interference based on the direction of the momentum vector is emphasized. The method is based on atomic potential center as an interferometer akin to the electron biprism within a short range of focus variation (<10 nm) from the reference Gaussian image plane and resembles Abbe’s picture of diffraction. In this alternative method, the aspect of phase change has been treated like off-axis electron holography considering the wave interference at an angle, and its analogy with other approaches can be understood with the help of interference geometry and associated momentum vector direction. Addressing the effect of large defocus together with various coherent aberration envelopes, and thickness on the rich variation in phase and image intensity patterns by the alternative procedure will be part of a separate discussion as this requires a departure from the traditional viewpoint due to unique experimental observation overlooked in the past. Simulation results are compared with the experimental images of 2D materials of MoS₂ and BN recorded under specific settings of third order spherical aberration \( C₃ = -35 \mu m \) and defocus \( \Delta f = 1, 4, \) and 8 nm and are found to be in good agreement.

2. Coherent image formation at near and far field

To begin with, a brief discussion is provided on the existing methodologies involved in the image simulation of atoms. This has an analogy with the slit diffraction pattern in light optics both at near and far field regimes. A typical Fresnel and Fraunhofer diffraction regimes and corresponding patterns are shown in figure 1. In light optics, there exists analytical formula derived from the Fraunhofer integral and Fourier transformation-based method to evaluate the far field diffraction pattern (section S2). The analytical formula embodies various parameters, e.g., wavelength (\( \lambda \)) of the illumination, scattering angle (\( \theta \)), dimension, and periodicity of the slits. The approach is based on the physical picture of path difference and associated constructive and destructive interference between waves having the same momentum vector direction described by the plane wavefront from a pair of spatial points at the slit opening that ensures the distribution of intensity transfer along different scattering angles. This is different compared to diffraction geometry involved in Fresnel zone construction for the image formation at near field (figure 1(c)). In the case of Fresnel diffraction, it is the angular correlation between the wave vectors pointing at different directions lying on the surface of a sphere, and the phase difference between various wave vectors is acquired due to path difference of waves while converging to a point with respect to the outward curvature of the spherical wavefront (more precisely parabolic wavefront in case of Fresnel regime) (Figure S2 &S3). This is the basis of Fresnel zone construction. Kindly note that there is no path
difference between various wave vectors with respect to the emitting point as the radius is the same for a sphere, but not so for a parabolic and plane wavefront geometry. For comparison with the off-axis electron holographic interference geometry and present alternate method, see section 2.3.1. On the other hand, in the Fourier transformation (FT)-based approach, the far field image of the object is the modulus of the FT (abs-FT) of the object function. However, the phase angle calculated for Fourier waves or equivalently Abbe waves (see Abbe’s two step imaging process in sections S2.3&2.4) corresponding to each frequency does not have the information on the scattering angle. Thus, the frequency of Fourier waves needs to be calibrated either with respect to the scattering angle obtained through the analytical method or from an experiment using a standard sample with known lattice parameter (Figure S6).

Now in the context of diffraction from atomic potential, the Fraunhofer pattern is the atom scattering amplitude $f(k)$ and the structure factor $F_g(k)$ for isolated and periodic atoms, respectively that depends on the strength and crystallography of the scattering potential in the object space. Figure 2 shows an example scattering distribution of isolated B and Mo atoms and in monolayer hexagonal periodic BN and MoS$_2$ lattice. In the image plane, the transmitted wave function or object exit wave (OEW) function can be derived in different ways, e.g., weak phase object approximation (WPOA) according to Zernike, Schrodinger integral equation and evaluating the Fraunhofer integral and are discussed next [9, 16].

### 2.1. Image simulation based on Zernike phase object and WPOA

This has origin in Zernike phase contrast theory where for pure phase or weakly scattering object, the object transmission function is represented by a complex function of the form $F(x) = e^{i\phi(x)}$ or $1 + i\phi(x)$ for small $\phi$, where $\phi$ is a real phase function corresponding to the discrete or periodic transparent object. This is known as weak phase object approximation (WPOA) [1]. In fact, the object function is real, it is the replica of the object in the form of object wave that carries the information encoded into its phase and amplitude and is similar in function to the Fourier or Abbe waves and holographic direct and twin image wave components. The effect of
Zernike phase plate modifies the intensity of the object wave that depends linearly on the object phase according to 

\[ I(x) = 1 \pm 2\phi(x) \]

WPOA is a straightforward and widely applied approach to simulate the HRTEM images of thin samples. In HRTEM, WPOA describes the phase shift of probe electron wave due to object electrostatic potential projected along the beam propagation direction, and the transmission function has the following expression after invoking WPOA, i.e., ignoring the terms with \( \alpha^2 \) and higher order,

\[ t(x) = 1 - i\sigma V_i(x, y) \]  \( (1) \)

Where, \( V_i(x, y) \) is the projected specimen potential and \( \sigma = \frac{2|e|m\lambda}{f} \) is the interaction constant. The approximation in equation 1 is essential to retain the object information in the image plane. The accompanied transmitted wave function is derived considering the change in magnitude of electron wavelength from \( \lambda \) to \( \lambda' \) due to attractive positive potential \( V_i \) of the specimen. Refraction of electron through atomic potential is associated with the change in momentum vector direction as well and is addressed in the description of the alternative method (section 2.3). \( \lambda' \) can be an average change corresponding to a mean inner potential (MIP) for a given spatial extent or as a function of spatial position from the center of the atom at medium and atomic resolution, respectively. At medium resolution, for average projected potential \( V_i \), the transmitted wave function of the electron within kinematical scattering in 1D is given by

\[ \psi_i(x) \sim t(x) \exp(2\pi ikz) \]  \( (2) \)

Equation 2 is equivalent to the reading component of Gabor’s in-line holography (section S1 (available online at stacks.iop.org/JPCC/5/085004/mmedia)). The plane wave component in equation 2 contributes to the background as DC component and poses difficulty in in-line holography along with twin image components.

The image intensity after considering the lens effect is given by \([4, 5]\)

\[ I(x, y) = \psi_i(x, y)\psi_i^*(x, y) \approx 1 + 2\pi\phi(x, y) \]  \( (3) \)

Figure 3 shows the image intensity calculated by using equation 3 with and without considering the lens response for isolated Mo, S, N, and B atoms \([7]\). Not considering lens response is similar to Zernike like phase transfer and as the potential function is asymptotic, peak intensity value will remain undefined with a background value of 1 (figure 3(b)). Considering aberration through optimum PCTF \((C_S = -35 \mu m \text{ and } \Delta f = 8 \text{ nm})\), peak values (and FWHM) of \( \sim 22000 \text{ (0.25 Å)} \) and \( 3500 \text{ (0.25 Å)} \) are obtained for Mo and B atoms, respectively (figure 3(a)). This gives a Mo/B peak intensity ratio of \( \sim 6.2 \). Peak intensity increases linearly with atomic number (Figure S14). The trend is in contrast with the experimental observation where changes in peak intensity are observed in the first decimal place with the atomic number (table 1). The high peak value according to equation 3 is due to the convolution procedure and cannot be normalized individually as the image without PCTF is not known (Figure S9). Periodicity can be extended through the lattice vectors in the image plane for the images of MoS\(_2\) and BN lattice.

As already mentioned that the phase shift due to aberration cannot be added in the trigonometric function in the diffraction plane as this shifts the wave amplitude, rather it is used as a coherent envelope function or frequency filter in the diffraction plane and as a point spread function (psf) in the image plane. According to equation S29, the PCTF gives weight to the magnitude of psf and aperture function in the form of Bessel function equivalent to Abbe’s theory that sets the resolution in terms of full width at half maximum (FWHM) in the final image \([5]\). Scherzer phase transfer will have maximum weight for optimum values of spherical aberration and
The phase contrast image calculated using the above expression varies weakly with atomic number and the peak intensity is

\[
B = \int |f(x)|^2 \, dx
\]

Where, \(f(x)\) is the transformed wave function.

\[
\psi(x) = \exp(2\pi ikz) + f_e(q) \exp\left(\frac{2\pi i q r}{r}\right)
\]

Where, \(q = k - k_0\) and \(f_e(q)\) is the atom scattering factor and is defined by,

\[
f(q) = \frac{m}{2\pi^2} \int V(r') e^{-\frac{2\pi i q r'}{r'}} \, dr'
\]

Which is the FT of the scattering potential. The solution of wave function based on the differential form of Schrödinger equation has the form of a plane wave. On the other hand, the integral form gives a solution of spherical waves along with the amplitude factor as the atom scattering factor. The equivalence between the two solutions can be perceived in terms of envelope of all the spherical waves from many adjacent scattering centers will eventually result in a plane wavefront. The picture is akin to the Huygens’ construction that a plane wavefront is the envelope of many forward scattered spherical wavelets and equivalent to First Born approximation. This result is used along with the scattering factor as derived by Moliere to calculate the intensity of scattered wave that is equivalent to writing component of Gabor’s in-line holography instead of reading as given by equation 2 (equation S4). However, the above description based on WPOA does not have the information on the geometry of interference in terms of momentum vector directions. Instead, considering the change in momentum vector direction due to interaction with the object potential and ensuing interference effect draw a clear comparison between various pictures (section S2.1 and S2.2).

### Table 1. Peak intensity after considering aberration blurring for \(C_s = -35 \, \mu m\) and \(\Delta f = +1, +4, \) and \(+ 8 \, nm\) in comparison with experimental image recorded under similar condition.

| Defocus (nm) | Peak intensity Mo (exp, ±0.04) | Peak intensity B (exp, ±0.02) | Peak intensity Mo (sim) | Peak intensity B (sim) |
|-------------|---------------------------------|--------------------------------|------------------------|------------------------|
| +1          | 1.08                            | 1.04                           | 1.03                   | 1.02                   |
| +4          | 1.23                            | 1.05                           | 1.19                   | 1.09                   |
| +8          | 1.24                            | 1.05                           | 1.25                   | 1.11                   |

Table 1. Peak intensity after considering aberration blurring for \(C_s = -35 \, \mu m\) and \(\Delta f = +1, +4, \) and \(+ 8 \, nm\) in comparison with experimental image recorded under similar condition.

The phase contrast image calculated using the above expression varies weakly with atomic number and the peak phase shift \(\varphi_{\text{max}}(\text{rad})\) follows \(Z^{0.6} - Z^{0.7}\), where \(Z\) is the atomic number [9, 20]. Though the trend can be complicated depending on the valence electron filling, and for specific atoms with higher \(Z\) can have smaller contrast compared to atoms with lower \(Z\) next to each other in the periodic table [9, 17, 21]. The peak intensity is
almost the same irrespective of the atomic number and changes only in the second decimal place. Figure S14 and Table S1 summarized the peak intensity and FWHM values calculated using equation 3 (WPOA) and equation 6 (atom scattering factor) for Mo, S, B, N, Zn, and O atoms. One can notice that the difference in peak intensity and FWHM maximum calculated by two different methods are markedly different. The intensity values calculated based on equation 6 show frivolous dependence considering only PCTF irrespective of atom number. However, the peak values are much smaller, and FWHM are higher by a factor of two, respectively calculated by equation 6 and 3, and the difference will remain even after the flux balance.

2.3. Atom as an interferometer

In this section, the alternative method based on the concept of the atom as an electrostatic charge center and its action as an interferometer on the simulation of atom image is described. However, the present treatment is different than earlier considerations of contrast and resolution due to single atom focusers\[22–24\]. The size of the atomic nucleus is extremely small (~1.6–15 fm) compared to the overall size of the atom with electron clouds (~0.1–0.5 nm). Therefore, the nucleus can safely be considered as a source of a positive point charge (+Ze) with associated Coulomb potential that decays inversely away from the charge center (figure 5(a)). The electron clouds surrounding the nucleus only screens the radially symmetric positive Coulomb potential. Moreover, the surrounding electrons clouds scatter the probe electron inelastically\[25\]. The probability of inelastic events for the fast probe electron is negligible compared to the dominant elastic events for samples having a thickness less than the extinction length and at a short exposure time typically 1–2 seconds This screened positive potential is attractive to the negatively charged probe electron wave while passing by the nucleus. The force experienced by the traversing electron will depend on the distance away from the nucleus. The calculated screened projected potential of Mo atom following the Hartree-Fock model\[9\] and associated attractive force in terms of bending...
angle is shown in figure 5(b). Within this picture, the projected potential of an atom can be considered as an electrostatic circular prism similar to the experimental off-axis electron holography biprism except attractive force acting from all directions with varying strength as a function of radial distance around it. This implies that the atom bends electron trajectory along $\theta = 0 - 2\pi$ azimuthal direction around it where momentum vectors lying on a conical surface representing scattering angles within same order of magnitude whereas a cylindrical biprism does the same but along single pair of momentum direction across a mirror plane (figure 6). Therefore, for the atomic case, it is necessary to describe the interference pattern from a different geometrical perspective than typical unidirectional electrostatic biprism. The picture is founded on the classical concept of wave optics but differs from the concept of channeling of quantum mechanical Bloch waves of electron through the crystal lattice. The correspondence between the two different pictures and the evolution of far field diffraction pattern will be part of a separate discussion. In the following sub-sections, at first the method based on off-axis electron holography is presented briefly to introduce on the formation of image contrast and then extending the principle to ‘the atom as electrostatic interferometer’ to simulate the image of the atom.

2.3.1. Image contrast in off-axis electron holography with electron biprism

The formation of electron interference pattern and resulting contrast in off-axis electron holography is emphasized here. The basic principle is based on single electron wave interference and the expression corresponding to HRTEM is given in equation S4 (section S1). The intensity pattern of the hologram is given by equation 7. The details on the off-axis electron holography methods and practices can be found in [17, 18].

$$I(x, y) = I(x) = a_1^2 + a_2^2 + 2a_1a_2 \cos(2\pi q_c x + \Delta \phi)$$

Where, $q_c = 2k_c$, is carrier spatial frequency of the hologram. $a_1$ and $a_2$ are the amplitudes of waves undergoing interference at an inclination angle due to action of biprism. $x$ is the spatial coordinate variable of the interference pattern and $\Delta \phi$ is the difference in phase between the two interfering partial waves, typically acquired by one of the two waves due to object potential. $\Delta \phi = 0$ for the vacuum waves. Information on $\Delta \phi$ appears as a small deviation in a straight hologram fringe pattern. The maxima and minima of the intensity pattern can be determined from equation 7 and remains the same throughout the interference field. The carrier frequency $q_c$ can be considered as carrying the information about the field strength and associated potential of the biprism. Field strength, potential, and carrier frequency can be empirically put together in a functional form. Now, it is important to note that the total flux or energy of the interfering waves must be preserved on the resulting interference field. This will become essential while calculating the image intensity and distinguishing contrast between the various atoms upon extending the above principle for the atom as electrostatic interferometer.
The \( q \) of the hologram that represents the fringe wavelength depends on the angle of superposition for a given wavelength. The higher the angle of superposition larger will be the carrier frequency resulting in finer fringe spacing. This is due to the horizontal component of the wave vector is larger at a higher inclination angle. This can be thought of as equivalent to Abbe’s picture where wave vector is larger or higher frequency at higher scattering angle (section S2.3). Thus, the two pictures may be unified and said that the higher the inclination angle, the smaller will be the fringe spacing or inter feature distance resulting in better spatial resolution. This was Abbe’s hypothesis in describing the diffraction limited imaging, where restricting the higher frequency by numerical aperture coming at a higher scattering angle limits the spatial resolution. However, in the case of off-axis electron holography, the geometry of interference involves only pair of momentum vector directions across a mirror plane passing through the biprism compared to a set of momentum vector directions lying on the surface of cones with continuously varying slant length in case of Abbe’s geometry.

### 2.3.2. Image contrast due to atom charge center equivalent to biprism

Now, the image formation by the interference of waves due to atom charge center similar to 1D electrostatic biprism is described here. Considering the radially symmetric atomic potential the intensity pattern can be calculated following equation 7 after incorporating wave interference effect from a given radial zone of extent \( \Delta r \). The zones described here are similar to binary type Fresnel zone plates with multiple foci, which depends on the scattering angle with different order of magnitude. Calculating the interference pattern along all azimuthal inclination angles for the peripheral zone area requires two additional considerations compared to the unidirectional interference pattern. The first consideration is that the wave flux will depend linearly on the perimeter \( 2\pi r \) which is a function of radial distance \( r \) from the center of the atom (equation S8 & section S2.9). Larger the perimeter or the zone area \( \pi (r_2^2 - r_1^2) \) away from the atom center, higher will be the flux of the wave approaching for the interference. The relative intensity contribution at the center of the pattern from different rims belongs to the same spatial coherent zone is scaled with \( 2\pi r \), where \( r \) is the radial distance from the center of the atom (figure 6, equation 8).

\[
I_{\text{rad int}}(r) = a_1^2 + a_2^2 + 2a_1a_2 \cos (2\pi q, r) \times 2\pi (r_{\text{max}} - r)
\]  

(8)

The second consideration is that of flux balancing between the flux of wave at the plane of the atom as given by the coherent rim area \( \pi (r_2^2 - r_1^2) \times I \), where \( I \) is the intensity at a given pixel point within this zone, and resulting interference field over a circular area around the optic axis as given by \( \pi r^2 \times I_{\text{rad int}} (1\text{st law of thermodynamics}).

\[
\int_{-dr}^{dr} I(r) \, dr = \pi (r_2^2 - r_1^2) \times 1
\]  

(9)

Where, \( dr = (r_2 - r_1)/2 \) and \( r = \sqrt{x^2 + y^2} \). \( I = 1 \) is the minimum intensity count on a pixel of size of 1 pm considered for the present calculation.

The resulting interference pattern is different compared to the unidirectional interference geometry. The pattern is radially symmetric with peak intensity at the origin and falls in amplitude away from the center, unlike the unidirectional pattern where average intensity remains the same throughout the field of superposition (figure 6). There is another distinct difference between the two interference processes in terms of illumination geometry and the effective range of electrostatic field maintaining the spatial coherency, which pulls the waves from opposite directions to interfere. In the case of off-axis holography, it is well known that the contrast of the hologram improves significantly if an elliptical probe is used instead of a round probe (figure 7). This is due to superior spatial coherency associated with the elliptical shape compared to the round shape [17]. However, the spatial coherency of the wave can be correlated with the order of electric field magnitude for a given spatial extent and equivalent order of bending angle due to Coulomb attraction with respect to the charge center. This is in contrast with the earlier consideration of an effective decrease in the angular distribution of illumination aperture associated with the elliptical probe and high spatial coherency [26]. For the elliptical spread of beam, most of the spatial extent of a wave will approach from the electric field regions away from the biprism center and will have a bending angle almost within the same order of magnitude (figure 7). This is the region of high spatial coherency resulting in high contrast compared to regions close to the bi-prism charge center where field strength and associated inclination angle vary strongly with the spatial distance. The situation has the analogue with high energy electron diffraction at a small angle versus large angle scattering and their relationship with the spatial coherency. At a small angle, coherency in elastic scattering is well preserved and gives rise to crystal Bragg diffraction peaks whereas at larger angles, scattering becomes more and more incoherent i.e., Rutherford type scattering. However, for atom case, there is no advantage for shaping the probe elliptical as potential falls much more rapidly, presence of neighboring atoms in case of crystal and short range of potential compared to bi-prism case. In the case of an atom, the potential varies strongly close to the nucleus and slowly beyond a certain distance away. Therefore, there will be a different contribution from various radial zones with specific rim width defined.
by the angle of superposition within the same order of magnitude to the overall pattern that can be controlled by the lens focus (figure 8). The lens focus will effectively increase the width of the interference field for the coherent zone. Therefore, we have divided the entire spatial range surrounding the atom starting from 1 pm to suitable outer range in terms of various zones with respect to example 1, 4 and 8 nm focus step (experimental through focus condition) and calculated intensity pattern for different zones separately and then summing up the contributions (figures 8 & 9). The contributions to the overall intensity pattern from different zones are incoherent and can be thought of as either binary Fresnel zone plate already mentioned or annular Airy apertures giving rise to Airy pattern at the specific focal length. Each zone is acting as a filter to specific momentum vector components similar to the PCTF as a spatial frequency filter. The spatial extent of the zone increases away from the atom due to decreasing potential and associated scattering angle. For example, the outer spatial extent is 10.31 and 4.19 pm corresponding to 1 nm focal length on the optic axis for Mo and B atoms, respectively, and the extent of the outer zone contributing to the coherent interference field increases with increasing focus (figure 8, S15, Table S2). Now depending on the focus settings, various zones will contribute to intensity pattern (at focus) and background (away from focus) differently. For a given intermediate focus setting, the focus length smaller and larger than that will contribute to the background differently. The zones closer to the atom center would have formed an interference pattern and propagate with the information along the original direction and various diffracted direction. Diffracted interference patterns will appear as a displaced image with intensity contribution very small compared to direct pattern almost an order of magnitude smaller which is considered to be similar in terms of relative total intensity between CB and SBs in off-axis and between the direct beam and diffracted beams in HRTEM. Figure 9 is showing schematically various contributions from different zones, for example, the Mo atom case for 1 nm. Moreover, the flux contributions will be different from
different zones and will be proportional to the annular zone area. As already mentioned, that the area and corresponding flux will be higher for the zones away from the nucleus. The calculated image of the Mo atom at three different focus values are shown in figure 10(a). The image of lighter atom B for similar focus settings is given in Figure S16. Kindly note that, with increasing focus the zones with large radial distance will contribute and modify the peak value of the image. The peak values are smaller for the B compared to the Mo case as the extent of outer zone is reduced due to the smaller magnitude of potential field. Figure 10(a) is the graph of peak image intensity versus atomic number for three different focus values. One can see that with a larger focus value, the peak intensity decreases slightly. The peak intensity has a dependence of \( \sim AZ^\delta \), where \( A \) is a fitting constant.
and $Z$ is the atomic number. Exponent B changes from 0.5 to 0.26 for change in focal length from 1 to 8 nm. However, the peak intensity values decrease significantly after considering image aberration (section 2.3.3 and figure 10(b)).

The relationship between the phase of the OEW and the resulting magnitude of intensity can be understood as follows. Here the meaning of phase change is the change in momentum vector direction, which is similar to Fresnel, Fraunhofer, off-axis electron holography type of interference geometry where wave interference is involved at an angle and different than the WPOA type phase shift where no information on the geometry of interference is available. Radial interference geometry modifies the unidirectional straight electron interference fringe to a radially symmetric pattern with a peak intensity at the center of the pattern. Whereas the intensity pattern oscillates periodically with the same magnitude in the case of unidirectional interference geometry across a mirror plane in off-axis electron holography. Now this phase term appears as carrier frequency $Q$ in off-axis electron holography and in the present case alters the peak intensity of the radial pattern depending on field strength and extent of a given radial zone. The field strength and corresponding potential information is the object information that can be interpreted and used to identify atoms. In the case of WPOA, series approximation and PCTF contrast are required to observe intensity pattern due to object potential but in the present case only defocus is sufficient. Now, in practical HRTEM, the atom contrast never goes to zero even if PCTF is zero. Traditionally, this is explained based on amplitude contrast due to ACTF which is the higher order contribution from interaction constant $\sigma$ according to equation 3 and is attributed as a non-linear imaging condition. However, the presence of contrast, even if defocus and the corresponding PCTF is zero, can only be explained by the present method in terms of radial interference originating from sectors of potential very close to the nucleus. Therefore, in the present method as the meaning of phase is different and different incoherent zones will have different phase change in terms of $Q$, thus there will be more than one wave functions superimposed on top of each other incoherently and it will be appropriate to interpret intensity directly to the object information. An alternative proposal on image reconstruction based on intensity directly instead of wavefunctions is discussed elsewhere.

### 2.3.3. Effect of aberration and defocus on the image series

The effect of third order spherical aberration ($C_s$) and defocus ($\Delta f$) are now considered to modify the image contrast further. The image contrast calculated based on the alternative method without consideration of lens transfer does not match with the experimental images as a function of the atomic number recorded under the particular settings of imaging conditions. Generally, a suitable combination of $C_s$ and $\Delta f$ are used to transfer maximum phase contrast for a given band width of spatial frequency with the best possible point resolution as given by the first zero crossings of phase contrast transfer function (PCTF) (Figure S18). A point resolution of better than 1 Å can be achieved in an aberration corrected microscope for a combination of $C_s = -35 \ \mu m$ and $\Delta f = 8 \ \text{nm}$ [27]. As already mentioned that the effect of aberration phase shift can be considered either in the diffraction plane or image plane depending on the requirement. In the image plane, the aberration figure due to spherical aberration is given by a disk of radius $r_s$ at the Gaussian image plane (section 5.2.10). The effect of $C_s$ and $\Delta f$ act in opposite directions, which depends on the third power and linearly with the scattering angle, respectively. The resulting effect is to either increase or decrease the full width at half maxima (FWHM) of the point spread function (psf) and impairs or improves the resolution of the imaging process, respectively. A typical angular deviation from the ideal ray path and corresponding aberration figure can be correlated for a given combination of $C_s$ and $\Delta f$ (Figure S17 and Table S3). Now depending on the electrostatic zones and associated scattering angle, the diffraction limited information will be blurred further for a given setting of $C_s$ and $\Delta f$.

Contributions of peak intensity from various incoherent zones can be found in Table S2. The peak intensity will be reduced due to aberration blurring for zones which have scattering angle more than what can be adjusted through PCTF. The effect of Peak image intensity and contrast difference between Mo and B atoms after considering $C_s = -35 \ \mu m$ and $\Delta f = 1, 4, 8$ and 8 nm is shown in figure 10(b) and listed in table 1. The modified peak intensity has been calculated based on the flux balance approach considering the ratio between the original interference field width to the final blurred area. Kindly note that the simulated peak intensity values at focus close to zero remain a little over reference background 1 irrespective of atomic numbers. This corresponds well with the experimental observation (table 1).

Now, one can notice that after considering aberration blurring due to psf corresponding to each zone and scattering angle, the peak intensity falls significantly compared to the ideal lens case (table 1 and figure 10(b)). From the experimental image series recorded under similar conditions as simulation parameters, the peak intensity to reference vacuum ratio is $\sim 1.23 (\pm 0.04)$ and $1.05 (\pm 0.02)$ for Mo and B atoms, respectively after normalizing with respect to vacuum intensity (figure 11). Images at other focus settings are given in Figure S11. The peak intensity values remain almost the same with a small change in focus variation (table 1). The mono and few layers thin BN and MoS$_2$ 2D layers are prepared by first probe sonication of powder sample (Sigma Aldrich) and then drop casting on a holey C grid. Away from the edge, the thickness of the samples increases to two and
few layers. The HRTEM images were acquired in an aberration corrected FEI TITAN 80–300 kV transmission electron microscope at JNCASR. For example, we have obtained a peak value of 1.19 and 1.09 for Mo and B atoms from the calculation for a focus setting of 4 nm (table 1). The peak intensity does not vary significantly in the experimental image for a small variation of focus and a good matching is obtained with the simulated image after considering aberration blurring of intensity from different incoherent zones. Any deviation can be explained in terms of slight deviation in scattering angle and associated extent of zones, focus value, consideration of aberration figure while simulating the absolute intensity of atoms. The close agreement between experimental image and simulation results is in contrast with earlier prediction based on Stobbs factor where a difference in contrast by a factor of 4 was reported [28].

3. Conclusions

In conclusion, an alternative approach for simulating the atom image in HRTEM within few nanometers of focus variation is presented. The method is based on considering atoms as electrostatic charge centers inducing interference of waves along the radial direction and an extension of the conventional off-axis interference geometry with unidirectional electron biprism. The method put forth a physical picture based on the geometry of interference within classical wave optics. The method can predict the absolute intensity of atoms with atomic numbers in the correct order unlike the other two methods where relative intensity between atoms can be compared. The simulated results corresponding to the image intensity are in close agreement with the experimental images of Mo and B atoms recorded under the optimum combination of third order spherical aberration $C_3 = -35 \mu m$ and defocus $\Delta f = 1, 4, \text{and } 8 \text{ nm}$.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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