Quantitative transmission electron microscopy at atomic resolution

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Abstract. In scanning transmission electron microscopy (STEM) it is possible to operate the microscope in bright-field mode under conditions which, by the quantum mechanical principle of reciprocity, are equivalent to those in conventional transmission electron microscopy (CTEM). The results of such an experiment will be presented which are in excellent quantitative agreement with theory for specimens up to 25 nm thick. This is at variance with the large contrast mismatch (typically between two and five) noted in equivalent CTEM experiments. The implications of this will be discussed.

1. Introduction
High-resolution conventional transmission electron microscopy (CTEM) and scanning transmission electron microscopy (STEM) are both powerful techniques capable of investigating local atomic structure. STEM uses a finely focused probe and CTEM uses plane wave illumination conditions. Interpretation of images in both techniques relies critically on image simulations [1]. For example, the intensity of columns in high-angle annular dark-field (HAADF or Z-contrast) imaging in STEM depends on the physics of thermal diffuse scattering (TDS) and Debye-Waller factors. Phase contrast and associated contrast reversals can dominate CTEM images. Quantitative agreement has recently been shown between experiments and theory in Z-contrast STEM [2,3] to within a few percent. Those results demonstrate that the current understanding of image formation is adequate in the modeling of TDS, which dominates Z-contrast images. However, quantitative comparisons in CTEM have often been plagued by a large contrast mismatch (typically between two and five) between theory and experiment, a phenomenon often referred to as the Stobbs factor [4]. Recently, it has been shown that careful consideration of the image recording process may account for the Stobbs factor in CTEM [5]. Bright-field STEM, in which lattice images are formed by interference of overlapping convergent beam discs, is related to CTEM via the principle of reciprocity [6]. As in CTEM, these images are dominated by phase contrast. Bright-field STEM images can be acquired simultaneously with Z-contrast images without the need to change the electron optical conditions of the microscope (except for slight adjustments in the defocus), providing near-simultaneous complementary information.
Similarly to CTEM, bright-field STEM images are more sensitive than Z-contrast images to light columns of atoms.

Here we report on quantitative comparisons between experiments and theory in bright-field STEM [7]. Experimental bright-field STEM images are placed on an absolute scale [8] and can thus be directly and quantitatively compared with image simulations. We show near perfect agreement between simulations and experiment and discuss the implications for CTEM.

2. Results and discussion
An FEI Titan 80–300 with a super-twin lens (Cs = 1.2 mm) was operated at 300 kV with a probe convergence semi-angle of 9.4 mrad. Experimental bright-field STEM images of a SrTiO$_3$ single crystal (2.8 mrad collection semi-angle) are shown in the left-hand column of each column pair in figure 1 for a range of thickness and defocus values. Contrast reversals and changes in the pattern occur for different defocus and thickness values, characteristic of coherent interference (phase

![Figure 1](image_url)

**Figure 1.** Top rows: experimental bright-field STEM images (left panel in each column, unfiltered) compared to multislice absorptive model calculations (right panel in each column). The upper labels in each image show their contrast values. The lower (black background) labels state the defocus, with underfocus being negative. Bottom row: experimental and simulated Z-contrast images (54 nm underfocus). Note that all images are on an absolute intensity scale relative to the incident probe and reported as a fraction of the incident probe intensity. The simulations have been convolved with a Gaussian of 0.11 nm FWHM to account for the effects of a finite source size.
contrast) images. The labels on each image state its measured contrast value, defined here as the ratio of the standard deviation of the image intensities to the mean image intensity.

Results from calculations using the multislice absorptive model [9] are shown in the right-hand column of the column pairs in figure 1. Spatial incoherence is the combined effect of a finite illumination source size, mechanical instabilities, sample drift, etc. and it is taken into account by convolving the simulated images with an effective source distribution function, a Gaussian with a full-width at half-maximum (FWHM) of 0.11 nm. Because spatial incoherence is difficult to measure directly in non-aberration-corrected STEM, several control experiments were performed to ensure the effective source size used here provides a realistic estimate for the spatial incoherence in this instrument. In particular, the bottom row in figure 1 shows Z-contrast images and comparisons with frozen phonon simulations [10] accounting for spatial incoherence. The Z-contrast images (detector inner angle of 65 mrad) were recorded at the same locations as the bright-field STEM images. A Gaussian effective source function with a FWHM of 0.11 nm yielded excellent agreement between simulation and experiment in both Z-contrast imaging (see bottom row in figure 1) and in bright-field STEM. The function is thus independent of the scattering processes, which are very different in bright-field STEM and Z-contrast imaging. The function is furthermore independent of the material and sample thickness, as has been shown previously using materials with a wide range of different scattering cross-sections, and thus it is not caused by a scattering mechanism not accounted for in the simulations. Furthermore, the mean image intensities, which are independent of spatial incoherence, agree quantitatively in both experiments and simulations in both bright-field STEM and Z-contrast imaging (not shown). Discrepancy in the mean intensities would indicate a redistribution of intensities by a scattering process not accounted for in the simulations, which is clearly not the case.

Figure 1 shows near perfect agreement between simulations and experiments across the entire range of thickness and defocus values. The average contrast mismatch factor between experimental and simulated bright-field STEM images is 1.15. This small residual mismatch is entirely within the experimental uncertainty caused by the noise of the detector, residual astigmatism, drift and, most importantly, the thickness determination (±1 nm).

3. Quantum mechanical model for TDS
The frozen phonon model has been used for calculating the Z-contrast images shown in Figure 1. That approach [10] models elastic scattering from atoms displaced from their equilibrium positions. It is motivated by the idea that the time taken for the fast electron to traverse the crystal is much smaller than the oscillation period of an atom. Within this semi-classical model the electron sees a snapshot of the atom frozen mid-vibration. Each electron “sees” a different configuration, and the contributions of different electrons are summed incoherently in the detector plane. In practice this is implemented by a Monte Carlo integration. The frozen phonon model approach has been shown to agree well with an absorptive model of thermal scattering and has produced simulations that compare well with experiment. However, the frozen phonon model does not contain within its conceptual framework the momentum or energy transfer one would normally associate with inelastic scattering, in this case phonon excitation. This shortcoming has contributed to the mistaken assumption

Figure 2. Diffraction pattern components for a 60-A-thick SrTiO$_3$ specimen formed with a convergent probe of aperture 0.112 Å$^{-1}$ positioned over a Sr column and displayed on a log scale; (a) the full intensity, (b) elastically electrons only, and (c) inelastically (thermally) scattered electrons only.
that scattering from a moving lattice and the excitation of inelastic transitions are distinct effects.

We have introduced a fully quantum mechanical model for TDS by solving the many body Schrödinger equation using a Born-Oppenheimer approximation [11]. The Born-Oppenheimer (BO) model predicts the scattered intensity for a single electron, including multiple elastic and inelastic phonon scattering to all orders. This is an advantage over other approaches based on the Yoshioka formalism [12] in which the single inelastic scattering approximation is usually made to allow for tractable calculations. In this model, we can explicitly calculate the elastic component and inelastic components of the scattered electron wave. This is illustrated in figure 2. Whilst the semi-classical conceptual underpinnings of the frozen phonon model are quite different from the quantum mechanical model it is important to note that the frozen phonon model yields results which are numerically equivalent to the BO model [11]. The bright-field and Z-contrast images in the previous section can be calculated in the BO model and agree with those simulations. This confirms that the match between theory and experiment demonstrated there is on a secure footing.

4. Summary
In summary, we have demonstrated excellent agreement between simulation and experiment in bright-field STEM. The excellent agreement in this and in prior studies of Z-contrast images [2,3] shows that current models of image formation adequately model both low and high-angle scattering, including thermal diffuse scattering. Other inelastic scattering processes do not play a significant role in contrast formation for samples that are sufficiently thin. Furthermore, inelastic scattering, which by reciprocity is present in bright-field STEM as much as it would be in CTEM, is not the origin of the contrast mismatch observed in the CTEM literature for similar specimen thicknesses.

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5. References
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