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Multiple-ellipse fitting method to precisely measure the positions of atomic columns in a transmission electron microscope image

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ABSTRACT

In this paper, we propose a multiple-ellipse fitting method to accurately determine the atomic column positions in transmission electron microscopy (TEM) images. The column is enclosed by a series of ellipses fitted from contour lines at equidistant intensity levels, and each atomic column is shaped by an averaged elliptical shape to obtain its positions. In particular, the intensity profile of the atomic column can be obtained by an elliptically rotational average based on its shape; therefore, the intensities of the neighbouring atomic column can be subtracted for each atomic column during subsequent position refinement. This method can achieve precision in the picometre range, and we quantitatively measure this precision by analysing an image containing two Gaussian-shaped atoms and some simulated high-resolution transmission electron microscopy (HRTEM) images of $SrTiO_3$.

1. Introduction

Currently, sub-angstrom resolution is achievable due to the widespread use of spherical aberration (Cs) correctors in transmission electron microscopy (TEM) (Haider et al., 2008), and quantitatively analysing TEM images is crucial. Because an individual atomic column can be directly distinguished, information on the position and intensity of the atomic column has attracted the attention of researchers. In high-resolution transmission electron microscopy (HRTEM) images and scanning transmission electron microscopy (STEM) images, the quantitative position can help researchers measure the lattice structure and distortions, e.g., elemental mapping of a monolayer crystal or for atomic number counting in STEM images (Feng et al., 2014; Lebeau et al., 2010), in interface research on superlattices containing a highly confined 2D dopant layer (Wang et al., 2016a,b) and atomic-scale polarization mapping on the domain walls of a ferroelectric film in HRTEM image (Catalan et al., 2011; Jia et al., 2008; Nelson et al., 2011). Notably, non-rotationally symmetric aberrations may cause a non-negligible intensity-centre shift of the atomic column on a HRTEM image (Lin et al., 2015). The centre of the atomic column is better measured on the phase image of an aberration-corrected exit-wave function for HRTEM images, and more information can be ascertained regarding the atomic column on the phase image. For example, argand plots obtained from the exit-wave function reconstructed from focal-series HRTEM images have been used to determine atomic thickness (Wang et al., 2010, 2012). The intensity distribution of an atom along its radial direction has been used to measure the focal distances of atoms to determine the three-dimensional structure of graphene (Van Dyck and Chen, 2012).

Several methods have been developed to quantitatively determine the atomic column position. (i) The moment method calculates the mass centre within the atomic column, and its precision can reach 0.1 pixels (Feng et al., 2007). (ii) The model-based method (MB method) utilizes a two-dimensional (2D) Gaussian function to fit the intensity distribution of the atomic column with a precision of a few picometres (Van Aert et al., 2009; Bals et al., 2006). In particular, the empirical parametric model can estimate the intensities of atomic columns to determine a mixed composition (Martinez et al., 2014). Therefore, the intensity of the atomic columns of heavier elements can be subtracted to highlight the intensity of lighter elements (Nord et al., 2017). (iii) The correlation peaks of the lattices, obtained by correlating the template of an atomic column or a group of atomic columns with the images, helps determine the position of the atomic columns (Zuo et al., 2014). (iv) A computer-vision-based approach was recently applied to an experimental electron micrograph to automatically detect atomic columns and classify local structural states, e.g., strained interfaces and

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crystal surfaces, without supervision (Laanait et al., 2016). However, most methods neglect the intensity distribution from neighbouring atomic columns; therefore, their calculation precision is limited.

Here, we propose a new method called the multiple-ellipse fitting (MEF) method, which can provide the intensity distribution of each atomic column and can accurately show the positions of atomic columns by subtracting the intensity contributions from neighbouring columns. The intensity distribution is calculated by elliptically-rotationally averaging the intensities of an atomic column, and the averaged intensity profile may be in any functional form, including a Gaussian-like function. The Gaussian-like function is denoted by a sum of several Gaussians and is more meaningful to describe the intensity distributions of atomic columns because the scattering factors for each atom are fitted from a sum of five Gaussians to estimate the projected potential (Peng et al., 1996). On the other hand, two-fold astigmatism A1 drifting on a timescale of a few minutes and the average motion of atoms may systematically elongate the atomic column image along one certain direction in an HR(S)TEM image (Biskupek et al., 2012). Therefore, fitting the intensity distribution of an atomic column may be help identify residual aberration and even the average motion of the atoms. In contrast, the general peak fitting scheme, e.g., the MB method, requires that the intensity profile must only be a Gaussian function (Wang et al., 2016a,b). Regarding computational efficiency, the MEF method is not complex because only a few ellipses must be fitted for each atomic column, and subtracting the contributions of neighbouring atomic columns is highly efficient compared to simultaneously fitting the intensities of nearby atomic columns using several Gaussian functions in the MB method (De Backer et al., 2016).

In this paper, we introduce the algorithm of the MEF method. Next, by using a special image containing two Gaussian-shaped atoms and a simulated HRTEM image of $SrTiO_3$, we prove its high precision.

2. Algorithm of the MEF method

2.1. Basic MEF method

In a TEM image, if the surface height in the z-axis represents the image intensity, then the atomic columns appear as “mountains”, as shown in a representative example in Fig. 1(a), which is intercepted from a STEM experimental image of a single hexagonal lattice (MoS_2 monolayer). On an intercepted image including only one atomic column shown in Fig. 1(b), given that the size of the intercepted image is $(2r + 1)^*(2r + 1)$ pixels^2 (r is one-half of the average distance between any two neighbour columns or slightly larger), a series of enclosed contour lines at different intensity levels I_k ($k = 1, ..., K$) outline the shape of the atomic column and I_K is the intensity level of the outermost contour line. Generally, the levels $\Delta = (I_{k+1} - I_k)$ are equidistant and only a few contour lines are required because the centres and shapes obtained from the contour lines at very close levels I_k are very similar to each other. For the k^{th} contour line, its centre $(\bar{x}_k, \bar{y}_k)$ is averaged from the coordinates of the line. The centre of the contour line is generally very local because the contour line must be enclosed and restricted within a small intercepted image. If a total of N_k points are sampled on the k^{th} line (N_k is determined by the length of the line), then the mass centre can be averaged from the centres of these contour lines, given by

$$\begin{cases} x_{mc} = \sum_k \bar{x}_k N_k (I_k - t) / \sum_k N_k (I_k - t) \\ y_{mc} = \sum_k \bar{y}_k N_k (I_k - t) / \sum_k N_k (I_k - t) \end{cases} \quad (1)$$

where $N_k(I_k - t)$ is the intensity summation of points on the k^{th} contour line, and $\sum_k N_k (I_k - t)$ gives the total intensity of all points on all contour lines. Moreover, the constant $t = I_K - \Delta$ can improve the weight of the intensity level of the contour line on the mass centre, and its effect is the same as that of the image-background-intensity subtraction applied in the moment method (Feng et al., 2007).

Although the centres and shapes of all contour lines are theoretically identical, they are slightly different due to noise and imaging conditions. To obtain a regularly shaped atomic column, the contour lines are fitted by ellipses, which can be resolved by using a non-iterative algorithm based on least-squares minimization (Halir and Flusser, 1998). In Fig. 1(c), the black dotted lines represent the ellipses fitting the contour lines, and each ellipse has five parameters: the centre coordinates (x_k, y_k), semi-major axis A_k, semi-minor axis B_k, and inclination angle θ_k. Then, we use one parameter $\xi_k = (x_k, y_k, A_k, B_k, \theta_k)^T$ to refer to all the parameters of each ellipse. Because the shape and centre of the atomic column are described by the average ellipse, the method is called the multiple-ellipse fitting method. Similar to equation (1), the parameter ξ of the averaged ellipse is given by

$$\xi = \sum_{k=1}^{K} \xi_k N_k (I_k - t) / \sum_{k=1}^{K} N_k (I_k - t) \quad (2)$$

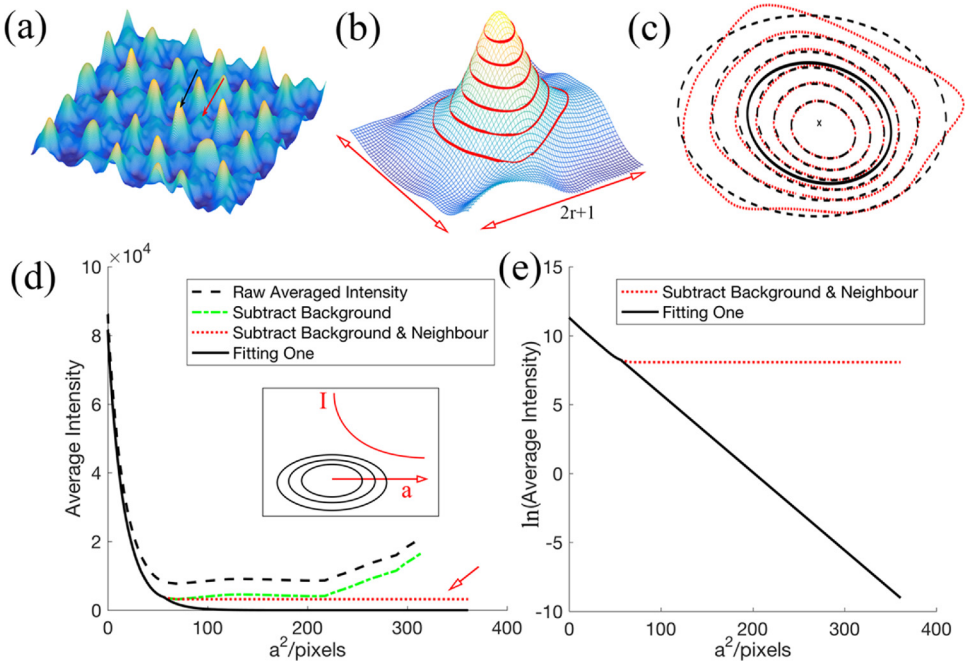

Fig. 1. (a) Schematic STEM image, in which the atomic columns on a hexagonal lattice indicated by the black and red arrows represent heavy and light atomic columns, respectively (the image is intercepted from an experimental STEM image of an MoS_2 monolayer, and the black and red arrows indicate the molybdenum atom and two superimposed sulphur atoms, respectively). (b) The intercepted image of the atomic column is indicated by the black arrow in (a). A series of contour lines outlines the shape of the atomic column. (c) Ellipses fitting the contour lines. The average ellipse is the black solid line, and its centre is marked by a cross. (d, e) The IDP of the atomic column of (b) shown in (d) the normal coordinate system and (e) the natural logarithmic coordinate system. The illustration in (d) shows the intensity of the atomic column attenuated along the semi-major axis. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).