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Study of the influence of higher-order Laue zones on the exit wavefunction*

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Based on an analytical expression of electron dynamical diffraction derived recently, the exit wavefunctions of non-periodic samples have been calculated. The results have shown that the amplitude and phase obtained by the analytical expression were in good agreement with those obtained using the multi-slice method. Besides, the influence of the higher-order Laue zone on the wavefunction for different sample thickness was studied in detail and the results showed that the exit wavefunction is dominated by the zero-order Laue zone. When the sample is very thin, the influence of the higher-order Laue zone should be considered.

Keywords: dynamical electron diffraction, electron crystallography, higher-order Laue zone
PACC: 6114D, 6116D

1. Introduction

In recent years, the field of electron crystallography has become increasingly active[1−8] because of the two important advantages over x-ray crystallography for the determination of atomic positions of specimens: phases of wavefunction can be determined directly from images and extremely small samples can be analysed. However, the strong interaction between electrons and matter[9] gives rise to complicated multiple-beam dynamical effects which make the observed diffraction intensities of a sample often significantly deviate from their kinematical values.[10,11] To solve the dynamical diffraction problem, a much more accurate analytical expression for electron dynamical diffraction has been derived in two papers[12,15], in which most of the electron propagation in the sample was considered. The results calculated by the new expression are much more accurate than those calculated by the phase object approximation (POA) formula. Moreover, the new formula is a general expression which can be used to calculate the scattering wavefunctions from zero-order to higher-order Laue zones. In this paper, we study the contribution of scattering wavefunctions from higher-order Laue zones.

2. Formula and calculation process

2.1. Introduction of the formula

An analytical expression of dynamical electron diffraction for crystal has been derived as follows[10]

$$\eta^{(l)} = \sum_h \sum_k \frac{1-\exp(-\mathrm{i}\lambda\pi g' z')}{4\pi^2 g'} F(hkl) \times \exp(-2\pi\mathrm{i}(h'x + k'y + l s_{13} z)), \qquad (1)$$

where $g' = (h'\boldsymbol{a}_1^* + k'\boldsymbol{b}_1^*)^2 + 2\boldsymbol{K}_{xy}(h'\boldsymbol{a}_1^* + k'\boldsymbol{b}_1^*) - \dfrac{2ls_{13}}{\lambda}$,

$$z' = z + \frac{p}{s}x + \frac{q}{s}y, \quad h' = h + s_{11}l, \quad k' = k + s_{12}l$$

and the scattering wavefunction can be written as

$$\phi = \exp\left(\sum_l \eta^{(l)}\right), \qquad (2)$$

where l can take number 0, 1, 2,..., denoting the lth order Laue zone, etc. Based on this expression, we can calculate the wavefunction of the sample under the following conditions: the zero-order or higher-order Laue zone was included; the incident beam can be along the normal of the surface of the sample or not; the incident beam can be along the crystal axis or not. More importantly, the wavefunction of a non-periodic sample can be calculated after some necessary modifications. In this paper, the wavefunction of C60

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(fullerene structure) is calculated for both zero-order and higher-order Laue zones.

2.2. Calculated examples

In order to calculate the wavefunction of a non-periodic entity according to expression (1), a necessary modification should be made as follows.

a) The summation $\sum_h \sum_k$ should be changed into an integral $\int_h \int_k$ in Eq.(1) for a non-periodic structure. Thus Eq.(1) is naturally changed into

$$\eta^{(l)} = \int_h \int_k \frac{1 - \exp(-\mathrm{i}\lambda\pi g' z')}{4\pi^2 g'} F(hkl) \times \exp(-2\pi\mathrm{i}(h'x + k'y + ls_{13}z)). \quad (3)$$

b) For computation, the integral in Eq.(3) can only be realized through summation. The difference between Eqs.(1) and (3) is as follows: in Eq.(3) h, k can only be integers, while in Eq.(1) they can be any real values. As is well known, the greater the number of sampling points, the closer to the integral the summation is, and therefore more computation will be needed. Thus, it is necessary to find a suitable number of sampling points to make sure that the error brought by the change from integral to summation can be neglected, and that the computation time is not too long.

c) In order to use the above expression, a unit cell should be defined for a non-periodic sample. For C60, the diameter is 0.7 nm[13] and we may define a bigger unit cell to decrease the influence from the non-periodic structure. In this paper, the parameter of C60 is defined to be 1.05 nm in three dimensions according to the atomic potential plots of C as a function of diameter r from the centre of the atom.[14]

d) We calculate $\eta^{(l)}$ according to Eq.(3) for different l.

e) We calculate the phase angle and the amplitude of the wavefunction ϕ after it is obtained.

2.2.1. Only zero-order Laue zone considered

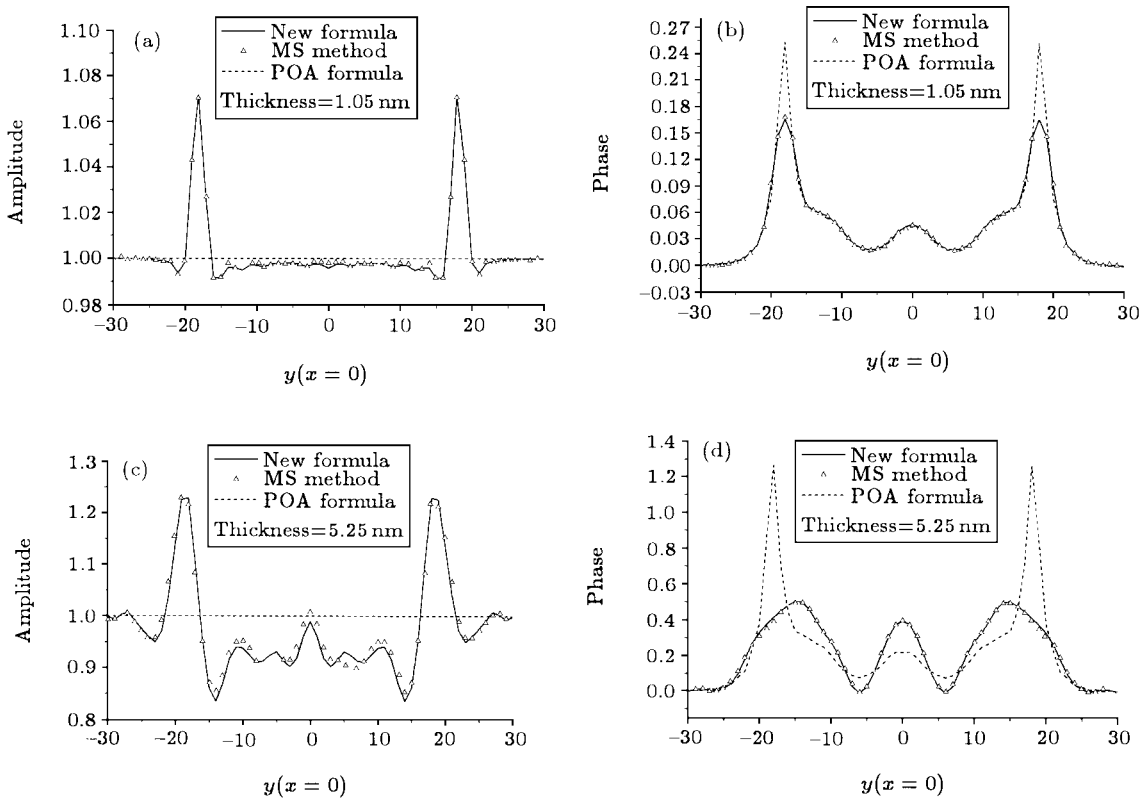


Fig.1. The wavefunctions of C60 molecules for the [001] Laue zone calculated using the new formula, the MS method and the POA formula with the wavelength of the incident beam λ=0.00251 nm and sample thickness=1.05 nm. The abscissa corresponds to the position (x, y) in a unit cell with x=0. The sampling points are 64×64, and the interval between sampling points is 0.0082 nm.

Firstly we just consider the zero-order Laue zone of C60 and assume that the incident beam is a monochromatic plane wave along the Laue zone axis and parallel to the normal of the sample surface. In this case $\boldsymbol{K}_{xy} = 0$, $p = 0$, $q = 0$, $s = 1$ and $l = 0$. Substituting these parameters into Eq.(3), we may calculate the scattering wavefunctions of samples, with different thicknesses. For comparison, the scattering wavefunctions calculated using the multi-slice (MS) method and the POA formula are also obtained and plotted in Fig.1.

From Fig.1, we see that the amplitude and phase of the wavefunction calculated using the new formula are in good agreement with those calculated using the MS method, even for thicker samples. However, those calculated using the POA formula deviate significantly from those calculated using the MS method, especially with no amplitude contrast shown.

2.2.2. Contribution of higher-order Laue zones to the wavefunction

According to Eq.(3), the scattering wavefunctions ϕ are calculated by considering a different order of Laue zone, as plotted in Fig.2. In order to obtain more accurate results, we take the number of sampling points to be 96×96, and hence the interval between neighbouring points is 0.0055 nm.

In Fig.2 we may find that the wavefunction is dominated by the zero-order Laue zone for amplitude and phase, and the thicker the sample is, the less the contribution of the higher-order Laue zone. When the sample is thin (see Figs.2(a) and 2(b)), the difference of wavefunction between $\sum_{l=0}^{2}\eta^{(l)}$ and $\sum_{l=0}^{3}\eta^{(l)}$ is very little, and thus the contribution from the third-order zone can be neglected. While if the sample is thick (see Figs.2(c) and 2(d)), the difference between $\sum_{l=0}^{1}\eta^{(l)}$ and $\sum_{l=0}^{2}\eta^{(l)}$ is very little, and the contribution from the second-order zone can also be neglected.

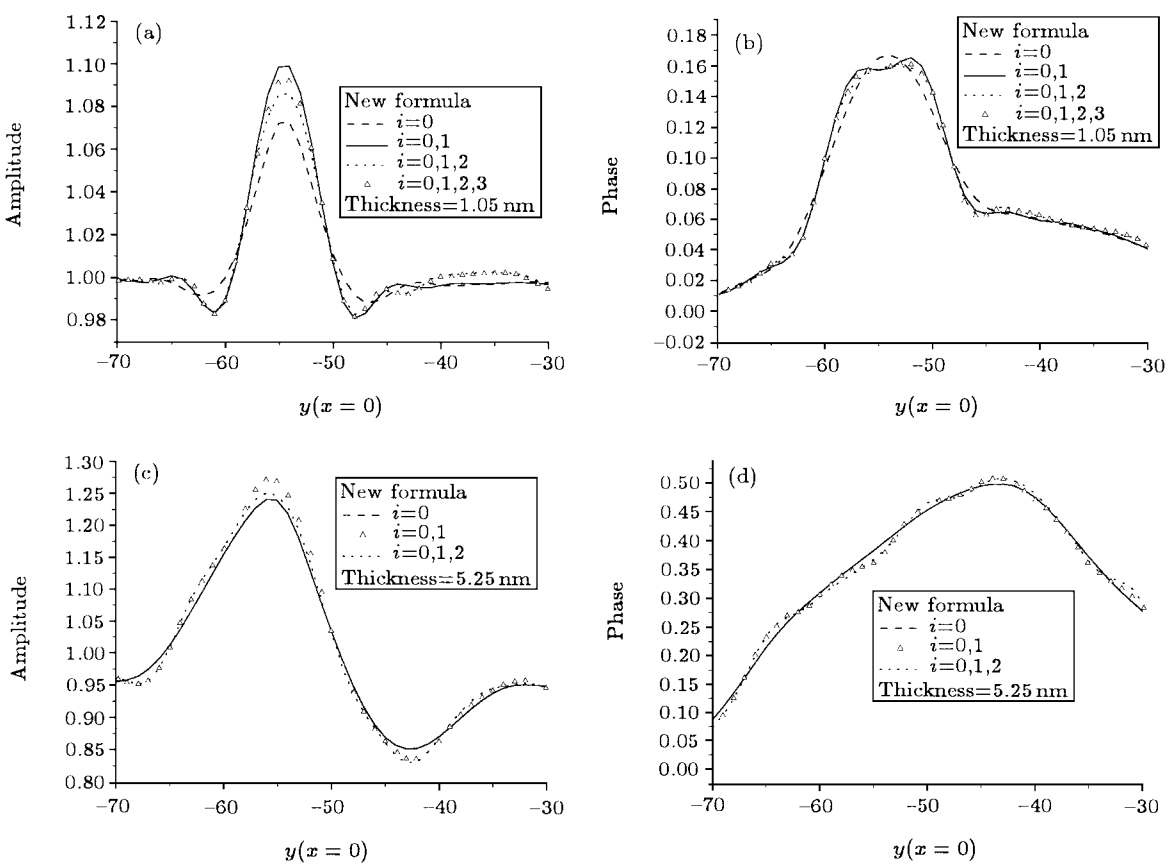


Fig.2. The scattering wavefunctions of C60 molecules calculated using the new formula for different order Laue zones with the wavelength of the incident beam λ=0.00251 nm. The abscissa corresponds to the position (x,y) in a unit cell with x=0.0 nm. (a) Thickness=1.05 nm; (b) thickness=5.25 nm (five periods along the z-axis). The number of sampling points is 96×96.

Letting $\phi^{(l)} = \exp(\eta^{(l)})$, $\phi = \phi^{(0)} \times \phi^{(1)} \times \phi^{(2)} \times \cdots \times$ and $\phi^{(l)} = |\phi^{(l)}|\exp(\mathrm{i}\alpha^{(l)})$, we have

$$|\phi^{(l)}| = \exp\Big(\int_h \int_k \frac{1-\cos(\lambda\pi g' z)}{4\pi^2 g'} F(hkl) \times \exp(-2\pi\mathrm{i}(h'x + k'y + l s_{13} z)) \Big) \quad (4)$$

and

$$\exp(\mathrm{i}\alpha^{(l)}) = \exp\Big(\mathrm{i} \int_h \int_k \frac{\sin(\lambda\pi g' z)}{4\pi^2 g'} F(hkl) \times \exp(-2\pi\mathrm{i}(h'x + k'y + l s_{13} z)) \Big). \quad (5)$$

We let $T_P^{(l)} = \dfrac{\sin(\lambda\pi g' z)}{4\pi^2 g'}$ and $T_A^{(l)} = \dfrac{1-\cos(\lambda\pi g' z)}{4\pi^2 g'}$, then $\phi^{(l)}$ can be expressed as

$$\phi^{(l)} = \exp\{\boldsymbol{F}[T_A^{(l)} F(hkl)]\}\exp\{\boldsymbol{F}[\mathrm{i} T_P^{(l)} F(hkl)]\}, \quad (6)$$

where $\boldsymbol{F}$ represents Fourier transformation. As above, we call $T_A^{(l)}$ and $T_P^{(l)}$ the intrinsic transfer functions for the lth order Laue zone. In order to study the contribution of the higher-order Laue zone to the wavefunction for different thicknesses, we plot the intrinsic transfer function versus the square of reciprocal vector in Fig.3.

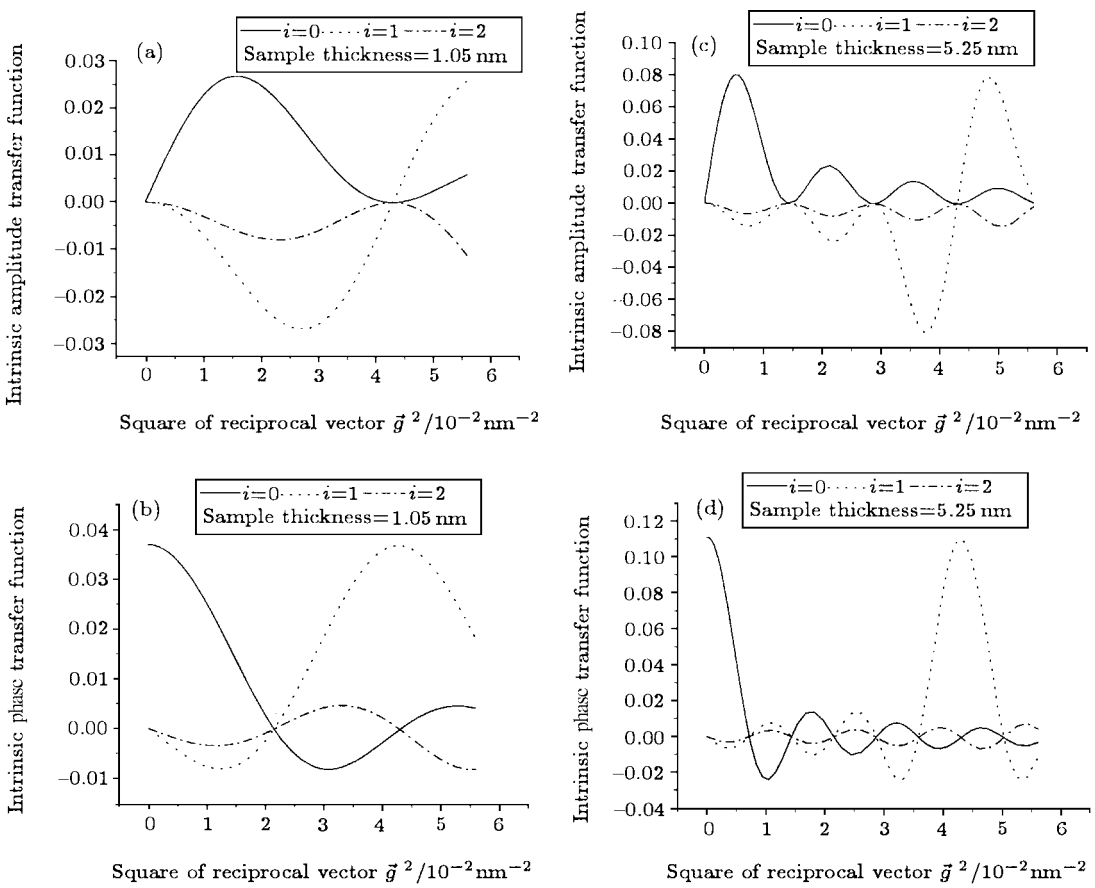


Fig.3. Intrinsic amplitude and phase of the transfer functions versus the square of reciprocal vector g^2 for different order Laue zones and different thicknesses calculated using the new formula.

From Eq.(3), we see that the contribution of amplitude from the lth order Laue zone can be expressed approximately by the product of $T_A^{(l)}$ and the structural factor $F(hkl)$, whereas the contribution of phase is expressed by the product of $T_P^{(l)}$ and $F(hkl)$. As is well known, the value of $F(hkl)$ decreases rapidly when the value of reciprocal vector $\boldsymbol{g}(hkl)$ increases. In Fig.3, we find that the transfer functions for different orders of Laue zone are oscillation functions. For the zero-order Laue zone, the location of peaks for amplitude and phase transfer functions correspond to small values of reciprocal vector $\boldsymbol{g}(hkl)$. For the higher-order Laue zone, the locations of peaks or valleys correspond to large values of reciprocal vector $\boldsymbol{g}(hkl)$, and the higher the order of Laue zone, the greater the reciprocal vector corresponding to the peak of transfer functions. In this way, we can conclude that the wavefunction is dominated by the zero-order Laue zone and the higher the order of Laue zone, the less the contribution to the wavefunction. Comparing Figs.3(a) and 3(b)

(thin sample) with Figs.3(c) and 3(d) (thick sample), we can see that the thicker the sample is, the less the contribution of higher-order Laue zones to the wavefunction. Thus it can be concluded that the influence of higher-order Laue zones can be neglected for thick samples.

In order to study the results obtained using the new formula, in which higher-order Laue zones are taken into account, and using the MS method, we have divided the sample, with a thickness of 1.05 nm, into sixteen slices and we calculate the wavefunctions of C60 using the MS method. We can find that the result calculated using the new formula, considering the second-order Laue zone, corresponds to that calculated using the MS method with the sample divided into sixteen slices (see Fig.4).

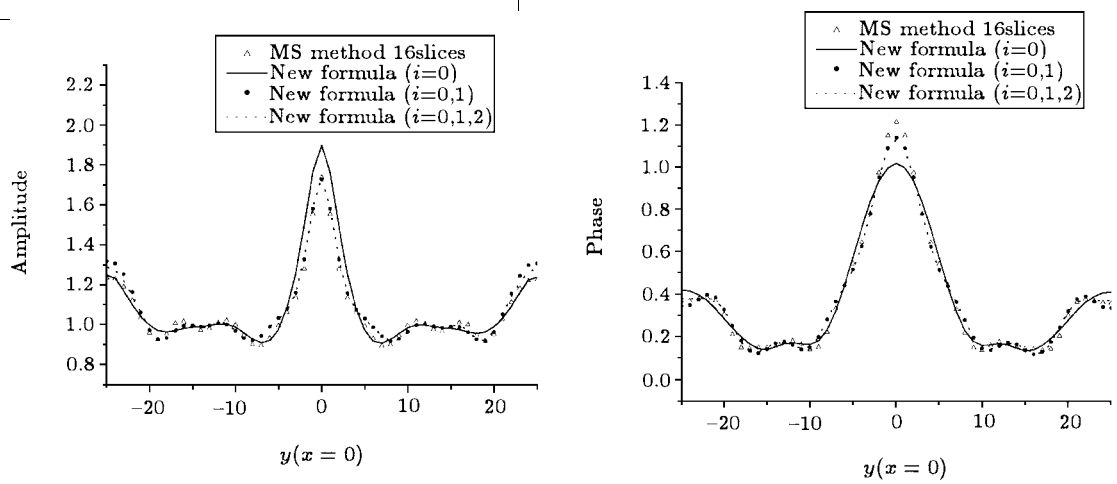


Fig.4. A comparison of the wavefunctions obtained using the new formula and using the MS method.

3. Discussion and conclusion

Based on the analytical expression, the exit wavefunctions of C60 have been calculated for zero-order and higher-order Laue zones. The following conclusions have been obtained. (a) The new formula can be used to calculate the wavefunctions of a non-periodic structure if a necessary modification is made. (b) The exit wavefunctions are dominated by the zero-order Laue zone for any sample thickness. (c) The thicker the sample is, the less the contribution from higher-order Laue zones to the wavefunction.

In principle, the difference among the three methods (the new formula, the MS method and the POA formula) can be described as follows. In the MS method, all the propagation of all electrons is considered, while in the new formula, most of the propagation of the electrons in the sample is considered. In the POA formula, none of the propagation is considered. So it is understandable that the results obtained using the new expression are much more accurate than those obtained using the POA formula.

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