Efficient kinematical simulation of reflection high-energy electron diffraction streak patterns for crystal surfaces

Kangkang Wang*, Arthur R. Smith

Department of Physics and Astronomy, Nanoscale and Quantum Phenomena Institute, Ohio University, Athens, OH 45701, USA

Abstract

An efficient program with a user-friendly graphical interface has been developed for simulating reflection high-energy electron diffraction patterns obtained on crystal surfaces. The calculations are based on a kinematical approach which considers single electron scattering events at the surface. This time-efficient approach is in most cases sensitive enough for distinguishing different structural models, even if they differ very subtly. The results are presented in realistic two-dimensional density plots which can be directly compared to experimental observations. This program provides a useful tool in studying different structural models for crystal surfaces.

Keywords:
Reflection high-energy electron diffraction (RHEED); kinematic; electron diffraction; surface reconstruction; computer simulations.

PROGRAM SUMMARY

Manuscript Title: Efficient kinematical simulation of reflection high-energy electron diffraction streak patterns for crystal surfaces
Authors: Kangkang Wang and Arthur R. Smith
Program Title: RHEEDsim
Journal Reference:
Catalogue identifier:
Licensing provisions: none
Programming language: Matlab
Computer: Personal Computers
Operating system: Windows with Matlab environment
RAM: Greater than 1 MB
Number of processors used: one
Supplementary material:
Keywords: Reflection high-energy electron diffraction (RHEED), kinematic, electron diffraction, surface reconstruction, computer simulations.
Classification: 7.4 Experimental analysis of crystal surfaces
External routines/libraries: Standard Matlab library with graphical user interface support (version>7.6.0)
Subprograms used: local-distfinder, local-gbroaden, local-IM2D, local-IM, local-LEEDgen, local-recip, RHEEDcmap
Nature of problem:

*Corresponding Author. *Email Address:* kw136705@ohio.edu

Reflection high-energy electron diffraction (RHEED) has been widely used in studying crystal surface structures all over the world, especially in combination with ultra-high vacuum molecular beam epitaxy systems. In addition to determining the surface smoothness, RHEED is also a very useful tool in studying surface reconstructions, which are often encountered at the growth surfaces of semiconductors and alloys. While the positions of the fractional streaks can be used to determine the basic information of the surface supercells, the intensity modulations on the fractional streaks provide further insights on the details within these unit cells. Kinematic approach is an efficient method for simulating the RHEED patterns based on various surface structural models, which can help unraveling the surface atomic structure.
Solution method:
The kinematic approach utilized here assumes single scattering events. Furthermore, the Ewald sphere is approximated into a planar surface for computing the streak intensities (which are most relevant to real experiments). Structure factors are calculated based on a given input of atomic species and their coordinates, with user modifiable form factors. In addition to the intensity modulation within the surface plane, additional modulations extending into the z direction is also taken into consideration, resulting in realistic density maps of the RHEED streaks, which can be directly compared to experimental observations.
Unusual features: The main program RHEEDsim.m calls several local subprograms for certain computational tasks. As a result, all programs should be extracted into a single folder, and that folder should be set as the main directory in Matlab.
Running time: The computing time is computer and user parameter dependent, but typically ranges from few seconds to few minutes.

1. Introduction

Reflection high-energy electron diffraction (RHEED) is a widely used surface-sensitive technique in crystal growth processes. Especially in molecular beam epitaxy (MBE) growth of semiconductors and other crystalline thin films, RHEED is commonly used as a tool for probing the surface morphology, surface reconstruction, and to help determine surface structural models.[1] While intensity oscillations of specular beams have been exploited to determine growth rates, and many literatures and programs have been developed to explain and simulate these oscillations,[2, 3, 4] there are other very important information contained in the intensity distribution of the RHEED streaks.

Reconstructions at the surface, often encountered at the growth fronts of semiconductors and alloys, manifest themselves as fractional streaks/spots in RHEED patterns. While the positions of these fractional streaks/spots can be used to determine the surface supercell size and direction, additional intensity modulations provide insights into the detailed atomic structure within the supercell. The most common method for analyzing experimentally obtained RHEED patterns is to first examine the pattern itself and obtain basic information such as reciprocal lattice spacing, super-periodicity, etc. Then several (sometimes many) trial structural models would be considered, and for each of the structural models, corresponding RHEED pattern would be calculated theoretically and compared to the experimental observation. Because of the nature of RHEED (finite probing depth, multiple scattering centers, form factors, etc.), such theoretical calculations are usually quite lengthy. A repeatable, time-efficient algorithm performing such tasks would be extremely beneficial, for quick determination of the best structural model.

Kinematic approach which considers single scattering events has been shown to be a time-efficient method for computing the intensity modulations.[5] In this communication we demonstrate how this method can be applied in the creation of a practical program which simulates

realistic RHEED patterns for a wide variety of crystal surfaces. The program (RHEEDsim) is written in Matlab with a graphical user interface for easy interaction. All data computed can be either displayed in the program itself or exported in ASCII format for plotting with other software. Using time-efficient structure factor calculation routines, an intensity modulated quasi-two-dimensional RHEED pattern is created for any given RHEED beam direction within the surface plane. These patterns can be compared directly to the experimental observations and help distinguish different candidate surface structural models.

2. Geometrical setup

The geometrical fundamentals of RHEED can be found in many books and review articles, such as by W. Braun [1] and by J. E. Mahan *et al.*[6]. In this section only the geometrical setup relevant to the program will be described.

Because RHEED is a surface-sensitive tool sampling only a few atomic layers beneath the surface, here we treat the crystal surface as a thin sheet of atoms, repeated infinitely in the $x-y$ plane, while neglecting the periodic part extending into z direction (normal to surface). The infinite dimension in the $x-y$ plane results in reciprocal points within the plane having zero dimension, while the small depth of the slab results in near-continuous reciprocal rods extending into the z direction. In the program, these surface reciprocal rods with zero thickness are artificially broadened for practical purposes, a point we will be elaborate later.

The basic unit vectors in the reciprocal lattice mesh are calculated as:

$$\overrightarrow{\mathbf{b}}_1 = \frac{2\pi \overrightarrow{\mathbf{a}}_2 \times \hat{\mathbf{n}}}{\overrightarrow{\mathbf{a}}_1 \cdot \overrightarrow{\mathbf{a}}_2 \times \hat{\mathbf{n}}} \tag{1}$$

$$\overrightarrow{\mathbf{b}}_2 = \frac{2\pi \overrightarrow{\mathbf{a}}_1 \times \hat{\mathbf{n}}}{\overrightarrow{\mathbf{a}}_1 \cdot \overrightarrow{\mathbf{a}}_2 \times \hat{\mathbf{n}}} \tag{2}$$

where $\overrightarrow{\mathbf{a}}_1$ and $\overrightarrow{\mathbf{a}}_2$ are the unit vectors in the real-space lattice, $\hat{\mathbf{n}}$ is the unit surface normal vector. The reciprocal vector $\overrightarrow{\mathbf{G}}_{\parallel}$ in the $x-y$ plane can only take combinations of integer multiples of the reciprocal unit vectors:

$$\overrightarrow{\mathbf{G}}_{\parallel} = h * \overrightarrow{\mathbf{b}}_1 + k * \overrightarrow{\mathbf{b}}_2 \tag{3}$$

while the reciprocal vector $\mathbf{G}_{\perp}$ in the z direction is essentially a continuous vector.

Under the assumption of elastic scattering events, the diffraction condition is satisfied where the Ewald sphere cuts the reciprocal lattice points.[7] In our construction, the tip of the incident beam wave vector $\overrightarrow{\mathbf{k}}_0$ is tied to a reciprocal lattice point, and the center of the Ewald sphere lies on the origin of the $\overrightarrow{\mathbf{k}}_0$ vector. The radius of the Ewald sphere can be estimated using $k_0 = \frac{1}{\hbar}\sqrt{2m_0E + \frac{E^2}{c^2}}$, where E is the relativistic kinetic energy. For typical 20 keV electrons used in RHEED, k_0 is much larger than typical reciprocal lattice spacings. Ideally this Ewald sphere cuts into the reciprocal rod mesh of the surface, resulting into an RHEED pattern consisting of arrays of sharp points corresponding to the reciprocal lattice points lying on the Ewald sphere surface. In reality however, the reciprocal rods are broadened due to finite coherent surface area, and the Ewald sphere will also have a finite thickness due to energy losses and incoherency. As a consequence, the sharp points are often broadened into streaks or spots for growth surfaces of crystals. At a small glancing angle (mostly <3°) and a large radius, the Ewald sphere can be

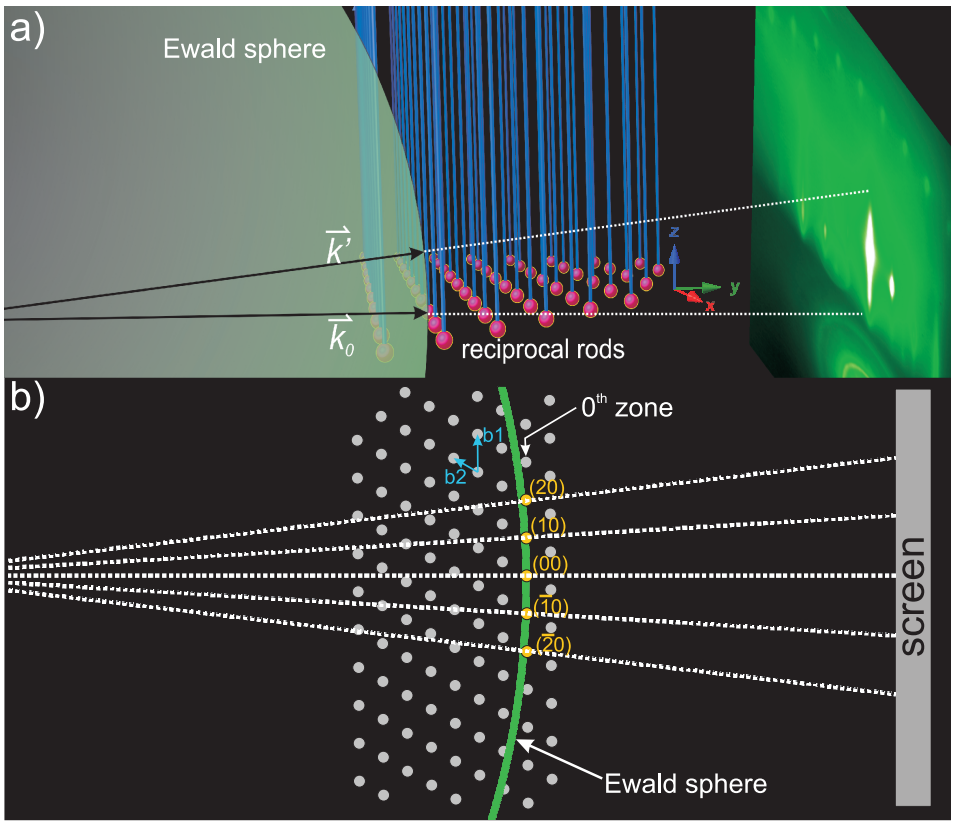

Figure 1: a) Schematic showing an Ewald sphere with radius k_0 cutting into the surface reciprocal rods, leading to a diffraction pattern. b) top view of the geometrical setup showing the reciprocal points along perpendicular-to-beam direction are lying on the Ewald sphere, forming the zeroth zone streaks on the RHEED screen. Both the Ewald sphere and the reciprocal mesh are for illustration purposes only, and not to scale.

approximated into a two-dimensional plane parallel with the reciprocal rods (z direction) near the crystal surface. We will implement this approximation in the program and show that it works well in simulating the zeroth zone streak patterns, which are most relevant in the studies of surface reconstructions.

A schematic of the geometrical setup is shown in Fig. 1. As we can clearly see that whenever the Ewald sphere cuts into a reciprocal lattice point, the diffraction condition is satisfied and that $\vec{\mathbf{k}'}$ vector leads to a diffraction spot on the RHEED screen. Fig. 1(b) shows the top view of the surface geometry setup, we can see that the RHEED spots in the same order zone correspond directly to the reciprocal vectors $\vec{\mathbf{G}}_{\parallel}$ that are perpendicular to the incident beam direction. As a result the streak spacings in the RHEED pattern are directly proportional to the distances between these reciprocal points lying along the perpendicular-to-beam direction.

In the program, once the reciprocal mesh is calculated and the incident beam direction is entered, an algorithm (implemented in the subprogram local-distfinder) is then applied searching for all matching reciprocal points along the perpendicular-to-beam direction in the $x-y$ plane. The search starts from the origin $(h,k) = (0,0)$ and radiates out based on distance from the

origin, with a maximum distance cutoff. The resulting discrete set of matching reciprocal points, together with their distances to the origin, are stored in a matrix, providing the basis for all subsequent intensity calculations.

3. The structure factor calculations

3.1. Computing one-dimensional line intensity profiles

As can be seen from the geometrical setup, when the Ewald sphere is approximated into a planar surface, all reciprocal points lying in that surface (perpendicular to the electron beam direction) will lead to diffraction spots in the RHEED pattern. In the case where the unit cell contains more than one atom, the intensity of these diffraction spots will be modified based to the structure factor equation:[5]

$$F(\overrightarrow{\mathbf{G}}) = A \sum_i f_i(s) e^{-B_i s^2} \times e^{i\overrightarrow{\mathbf{G}}\cdot\overrightarrow{r}_i} \tag{4}$$

where $\overrightarrow{\mathbf{G}}$ is the reciprocal vector corresponding to the diffraction spot, $\overrightarrow{r}_i$ is the real-space position vector for the i^{th} atom in the multi-atom basis. The parameter s is defined as $s = \frac{sin\theta}{\lambda}$; θ is the angle between the incident and the scattered wave vector, which is very small in the case of RHEED. λ is the electron's wavelength. The atomic form factor f_i depends on a number of parameters including atomic species, s, and electron beam energy. Atomic form factors for isolated atoms can be obtained using the Doyle-Turner formula,[8] and from the database maintained by the National Institute of Standards and Technology (NIST).[9]

In the program, several approximations are made to the structure factor equation (4) in order to reduce computation time dramatically. For one-dimensional line intensity profile where z and $\mathbf{G}_\perp$ are zero, θ remains nearly the same for different peaks, also the electron energy is the usually a constant for a given RHEED pattern. Therefore, the s and E dependent parameters in the structure factor equation will become effectively a constant for the line profile, which can be normalized away by being absorbed into the normalization constant A. In real surfaces, the atomic form factors will depend on the atomic coordination: whether it is being shadowed by other atoms; as well they differ from the form factors for isolated atoms because in a crystal the electronic density of an atom usually depends on its bonding environment. Taking these into considerations, it is often desirable to enter various form factors for a given type of scatter and determine the best values by comparing the resulting pattern to experiments. Therefore in the program the atomic form factors are read in from an input file created by the users.

With these approximations, the structure factor for one-dimensional line intensity profiles becomes effectively:

$$F(\overrightarrow{\mathbf{G}}_\parallel) = A \sum_i f_i \times e^{i\overrightarrow{\mathbf{G}}_\parallel\cdot\overrightarrow{r}_{i\parallel}} = A \sum_i f_i \times e^{i[(hb_{1x}+kb_{2x})x_i+(hb_{1y}+kb_{2y})y_i]} \tag{5}$$

Once all the allowed $\overrightarrow{\mathbf{G}}_\parallel$ values are calculated from the geometrical calculations, and all the atomic coordinates are read in from the input file, the intensities $I = |F|^2$ for each $\overrightarrow{\mathbf{G}}_\parallel$ will be computed based on equation (5). This step generates a list of discrete intensity values corresponding to the discrete diffraction spot positions. To compare to experimentally obtained line profiles, these discrete peaks are being artificially Gaussian broadened with user-definable

broadening parameters. A symmetry with respect to the origin (namely, $I_{(h,k)} = I_{(-h,-k)}$) is utilized throughout for speeding up the calculations. Note that this symmetry is independent of the surface structural symmetry.

3.2. Computing quasi-two-dimensional streak intensity maps

While the one-dimensional line intensity profile at $z = 0$ (reciprocal space) is very powerful in distinguishing different atomic configurations within the surface unit cell, it does not truly reflect the surface roughness. For example, a three-dimensional surface (quantum dot) usually results in a spotty pattern, while a flat surface resulting from a layer by layer growth usually results in a streaky pattern. To simulate these streak intensities, the z component of the atomic coordinates will be important.

Since we are neglecting the periodic repeatability in the z direction (described in section I), the perpendicular-to-surface reciprocal vector becomes a continuous vector as opposed to the discrete vectors in the $x - y$ plane. Adding this vector into equation (5), the structure factor for the quasi-two-dimensional streak are then given by

$$F(\vec{\mathbf{G}}) = A \sum_i f_i \times e^{i[(hb_{1x}+kb_{2x})x_i+(hb_{1y}+kb_{2y})y_i]+iG_\perp z_i} \tag{6}$$

where z_i are the z coordinates of scatters in the multi-atom basis, and $\mathbf{G}_\perp$ is the continuous vector perpendicular to the crystal surface plane. In practice of course, $\mathbf{G}_\perp$ is incremented by a finite size depending on the desired pixel density of the quasi-two-dimensional density map. The program makes use of the already computed one-dimensional line intensity data to speed up the computation. Similar to the one dimensional case, here the streaks are also artificially Gaussian broadened for better visibility and direct comparison to experimentally obtained RHEED patterns. The resulting data is in the form of an intensity matrix and plotted as a density plot (i.e. the graylevel/color represents the intensity value of that diffraction spot).

4. Program structure

The program flow chart is shown in Fig. 2. The main program interacts with users via a graphical user interface, and calls several local subprograms to perform certain computational tasks such as Gaussian broadening of calculated peaks.

4.1. Inputs

The input to the program takes on a form of a text file created by the user. In this input text file, the user provides some essential information related to the surface structural model. Since the surface is considered to be a repeated lattice in the $x - y$ plane, the input file will include the real-space unit cell vectors (in the unit of Å) in the surface plane. Then a series of atomic coordinates (three-dimensional, x, y, and z) together with the atomic type for each scatter in the multi-atom basis will be entered. A series of trial atomic form factors for each different type of scatter will also be entered by the user.

This information will be sufficient for the program to set up the geometry and initiate the calculation. Other information such as the electron beam direction corresponding to the desired RHEED pattern, Gaussian broadening parameters, etc, will be required as one moves along with the calculation. These additional values can be conveniently entered into several input boxes in the graphical user interface.

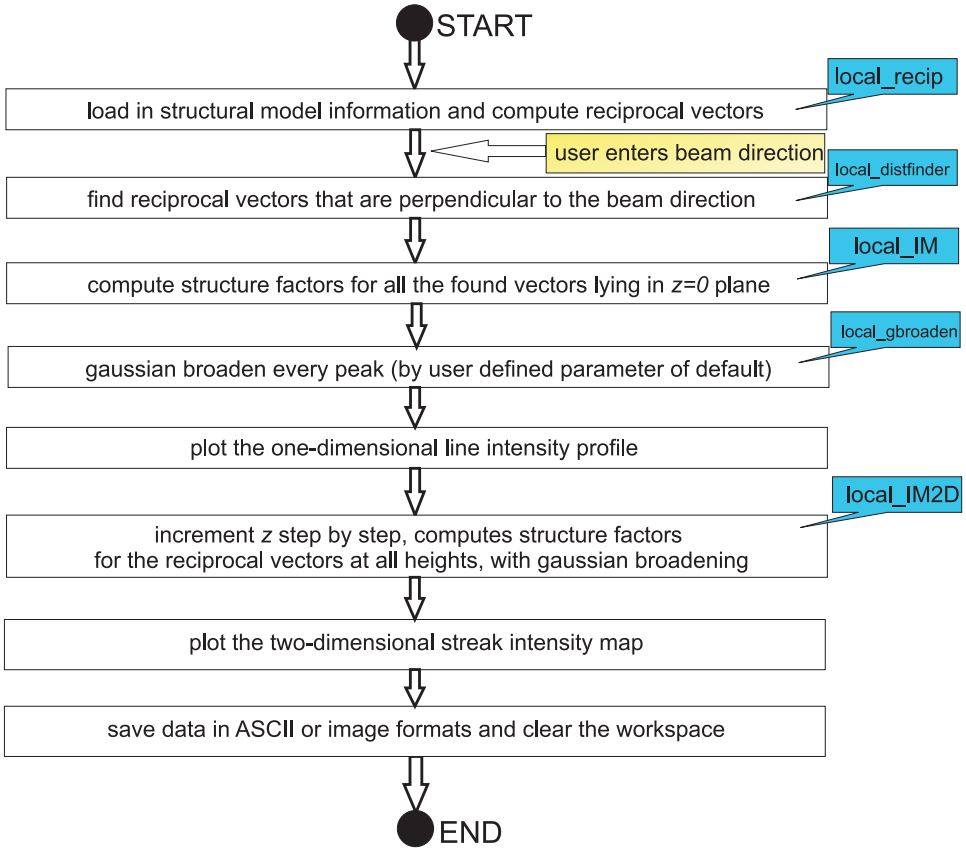

Figure 2: Example program flow chart for a typical RHEED streak pattern simulation

4.2. *Outputs*

The line intensity profile is computed and stored as a two column data array. The first column is the reciprocal spacings, and the second is the intensities. On the other hand, for the quasi-two-dimensional streak pattern, the data are stored in a matrix form, with the number of rows and columns corresponding to the number of pixels horizontally and vertically. Each element in the matrix records the calculated intensity for that particular diffraction spot. Both of these data are plotted in real-time in the graphical user interface, and can be opened up in new Matlab windows with figure toolbar for further editing and manipulation. Alternatively, data can be saved in standard ASCII format, to be used by other software.

5. Tests of the program

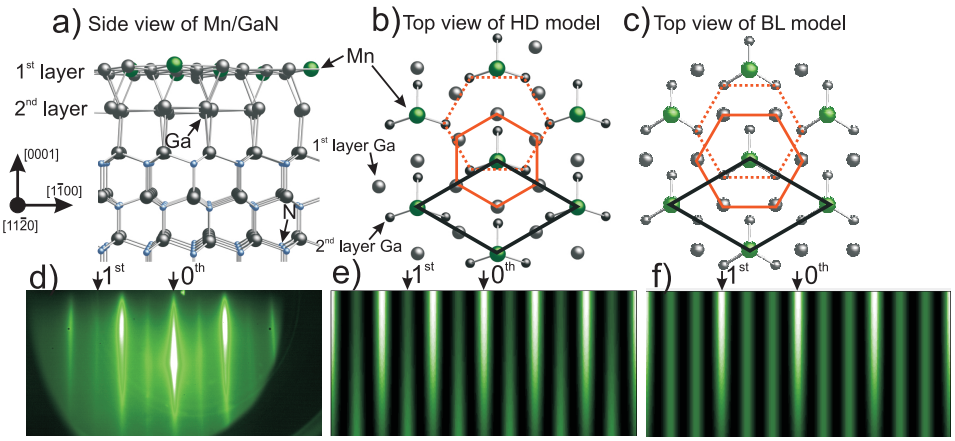

Figure 3: a) side view and b) and c) top view of HD and BL models. Black rhombus labels the surface unit cell. Orange hexagons (solid and dashed) labels the Ga configurations (1^{st} and 2^{nd} layer, respectively) d) experimentally obtained RHEED pattern along [$1\bar{1}00$] azimuth. e) and f) are simulated patterns for HD and BL models, respectively.

Recently we discovered that when Mn atoms are deposited onto the growth surface of GaN(0001), a $\sqrt{3}\times\sqrt{3}$-R30° reconstruction is induced.[10] Different structural models have been developed for this reconstruction, which provide an ideal testing opportunity for the RHEED simulation program. Under Ga rich growth conditions, there exist two extra Ga layers on the bulk-terminated GaN surface. Fig. 3(c) shows the first model (BL) developed which consists of Mn atoms substituting Ga atoms in the top layer of the GaN-bulk-like Ga bilayer. Fig. 3(b) shows the second model (HD) which consists of Mn atoms incorporating into the top Ga layer but in this case the Ga atoms rearrange themselves laterally into a 30°-rotated and contracted configuration. In both cases, the Mn atoms are in a $\sqrt{3}\times\sqrt{3}$-R30° arrangement, which produces $3\times$ along [$1\bar{1}00$] and $1\times$ along [$11\bar{2}0$]. The difference in the atomic arrangements, however, results into different intensity modulations. Fig. 3(e) and 3(f) show the simulated RHEED patterns for the two models computed using the RHEEDsim program, which can be directly compared to the experimentally obtained RHEED pattern shown in Fig. 3(d). As we can clearly see, the HD model resulted in a RHEED pattern where the $\frac{2}{3}^{rd}$ order streaks have now higher intensity than the $\frac{1}{3}^{rd}$ streaks, in much better agreement with the experimental findings than the BL model which produces

equal-intensity 3×. Furthermore we notice that in Fig. 3(e) the $\frac{2}{3}^{rd}$ order streak gradually reduces its width and intensity as z is increased (from top to bottom in the pattern). This effect is resulting from a slight z corrugation in the HD model, namely the Mn atoms are situated ~0.3 Å higher than the Ga atoms. This comparison further proves the validity of the HD model against other models, in addition to the total energy first-principles calculations carried out for these models.[10]

6. Summary

To summarize, we have developed a repeatable, time-efficient program with easy to use graphical interface for simulating RHEED patterns obtained on crystalline surfaces. Kinematical approach is implemented in the program for fast computing speed which is very important for quickly distinguishing different structural models. Both one-dimensional line intensity profiles and quasi-two-dimensional streak intensity maps are calculated within the program and displayed in real-time. The results can be then directly compared to experimental observations or exported for further processing. This program provides an effective tool for studying the RHEED patterns obtained during growth of crystalline semiconductors and alloys, and it is particularly helpful in the development and determination of atomic structural models for surface reconstructions.

Acknowledgements

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