Fast Phase Retrieval from Reflection High Energy Electron Diffraction Intensities during Growth

Takaaki Kawamura
Surface and Vacuum Physics, Institute of Industrial Science, University of Tokyo, Komaba, Meguro-ku, Tokyo 153-8505, Japan
(Received 12 January 2018; Accepted 1 April 2018; Published 20 April 2018)

A fast phase retrieval approach from reflection high energy electron diffraction (RHEED) intensities is proposed for monitoring growing surfaces almost in real time. The approach is based on a hybrid input-output algorithm and the processing time is reduced in two ways by taking peculiarities to RHEED intensities during growth. One is to reduce the number of sampling points along the incident beam direction in which the number is much larger than that in the normal direction due to grazing incidence of the beam. The other is to reduce the number of iterations in the retrieving process. The resulting real space objects are well reproduced providing surface morphology during growth approximately in 0.1 s. [DOI: 10.1380/ejssnt.2018.97]

Keywords: RHEED; Phase retrieval; Surface morphology; MBE growth; Real time observation

I. INTRODUCTION

Reflection high energy electron diffraction (RHEED) has widely been used to characterize surface morphology during growth by MBE [1–6]. In most cases the specular beam intensity and its oscillations are used to determine quality of the surfaces and/or the number of growth layers. In order to fabricate materials on a nanometer scale, it is quite important to observe the real space image of surface morphology in real time. For this purpose we have developed a fast phase retrieval approach that makes it possible to obtain real-space morphology from a measured RHEED intensity distribution during growth.

For phase retrieval from RHEED intensities, there are three peculiar difficulties. One is that the 000 Fourier component is not measurable in the case of reflection. The second is that the surface normal component (k_z) of scattering vector k is not an independent variable as it is fixed once the surface parallel components are given. The specular beam intensity should be used as the 00 component results in better phase retrieval. Because it turns out that 00 is the Bragg peak above the shadow edge is used in the retrieving process. The resulting real space objects are well reproduced providing surface morphology during growth approximately in 0.1 s.

II. PHASE RETRIEVAL FROM RHEED INTENSITIES DURING GROWTH

The present approach is based on the hybrid input-output (HIO) algorithm for phase retrieval [7, 8]. In order to overcome the above three difficulties peculiar to RHEED the 008 Bragg peak is used as the 00-component of 2D Fourier transform and intensity distribution around the Bragg peak above the shadow edge is used in the retrieval. Because it turns out that 00l with l being an integer as the 00 component results in better phase retrieval in the present case and the 008 Bragg peak is the best among 004, 006, and 008. In order to minimize a multiple scattering effect the azimuth angle is taken at 90° off from the [110] direction satisfying the so-called one beam condition [10].

Figure 1 shows the basic process of the HIO algorithm. The j-th iteration of the algorithm consists of four steps.

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* This paper was presented at the 8th International Symposium on Surface Science, Tsukuba International Congress Center, Tsukuba, Japan, October 22-26, 2017.
† Corresponding author: tkawamur@iis.u-tokyo.ac.jp

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Let assume the j-th input surface atomic arrangement as \( f_j(r) \) in real space \( (r) \) and its Fourier transform as \( F_j(k) \) in Fourier space \( (k) \). As the absolute values of scattering amplitude \( |F_j(k)| \) are given by measured RHEED intensities, the first step is to generate the j-th Fourier pattern \( F_j^\prime(k) \) by replacing the absolute values with the given amplitude \( |F_j^\prime(k)| \). The second step is the inverse Fourier transform \( F_j^\prime(k) \) to get a tentative atomic arrangement \( f_{j+1}(r) \). The third step is to generate \((j+1)\)-th atomic arrangement \( f_{j+1}(r) \) by using the constraint in real space as

\[
f_{j+1}(r) = \begin{cases} f_j^\prime(r) & r, f_j^\prime(r) \in S \\ f_j(r) - \beta f_j^\prime(r) & r, f_j^\prime(r) \notin S \end{cases}.
\]

Here \( \beta \) is a constant taken between 0.6 and 0.8, and \( S \) stands for the beam irradiated area for \( r \) where given constraints are satisfied for \( f_j^\prime(r) \). We use the constraints that both the real and the imaginary parts of \( f_j^\prime(r) \) are positive. The fourth step is the Fourier transform \( f_{j+1}(r) \) to get \( F_{j+1}(k) \). For monitoring the convergence of the iteration process, an error function is used. The error function at the j-th iteration is defined according to Miao et al. [7]

\[
E_j = \left( \sum_{r \in S} |f_j^\prime(r)|^2 / \sum_{r \in S} |f_j^\prime(r)|^2 \right)^{1/2}.
\]

In Fig. 2 is shown the calculated RHEED scattering amplitude distribution at the 008 Bragg condition by using an atomic arrangement obtained by a kinetic Monte Carlo simulation of homoepitaxial growth on Si(001) [11]. The coverage of adsorbed Si atoms is 0.33 ML. The horizontal direction corresponds to \( k_y \) from the shadow edge \((-0.1\pi/a \) to \( 0.9\pi/a) \) and the vertical direction corresponds to \( k_x \) from \(-\pi/a \) to \( \pi/a \) with \( a = 0.384 \text{ nm} \) being the surface lattice constant on the Si(001) surface. The brightest contrast corresponds to the Bragg peak. The beam diameter is taken as 64\( a \). As the glancing angle is 3.873°, the beam irradiated area is elliptic with 14.8 times longer dimension along the beam azimuth. The number of sampling points along the beam azimuth \( (k_y) \) direction is taken as 16 times larger than that in the \( (k_x) \) direction normal to the beam. Then the number of basic sampling points in \( (k_x,k_y) \) space is taken as 128\( \times \)2048 which makes it possible to apply the current phase retrieval approach based on the oversampling method [7].

For fast phase retrieval, we reduce the number of sampling points, along \( k_y \) direction, i.e., the points at which RHEED intensities are sampled. The most time consuming part of the phase retrieval is repetition of Fourier transforms by fast Fourier transform (FFT) and its inverse transform. FFT requires \( N_k \log N_k \) operations when the system size is \( N_k \). If the system size is reduced from \( N_k = N_x \times N_y \) to \( N_k = N_x \times N_y / M_R \) with \( M_R \) the reduction rate, the processing time should be reduced by \( 1 - \log M_R / \log(N_xN_y)/M_R \). At the same time the spatial resolution in the real space \( f_j(r) \) should be lowered.
by a factor of $M_R$ along the incident beam direction.

Figure 3 shows three series of surface morphology variations during homoepitaxial growth on Si(001). Figure 3(a1–a5) in the first row shows surface atomic arrangements (morphologies) obtained by a kinetic Monte Carlo simulation. These morphologies are used as the input for calculating RHEED intensities. The coverage increases from 0.33 ML [Fig. 3(a1)] to 0.84 ML [Fig. 3(a5)]. The growth rate is 0.125 ML/s and the temperature is 700 K. The bright contrasts corresponding to growth islands slant upward to the right as the azimuth angle is 6° off from the [110]. Figure 3(b1–b5) in the second row shows retrieved morphologies from the calculated RHEED amplitudes after 8000 iterations using 128 × 256 sampling points. The number of sampling points is reduced by a factor of 8 ($M_R = 8$). The upper and lower bounds of sampling region in $k_y$ direction is indicated by two lines in Fig. 2. Surface morphologies are reproduced quite well, except for lowered resolution due to the reduced number of sampling points. The time needed in the above process is approximately 40 s on PC (Mac Pro, 3GHz 8-core Intel Xeon E5). When the sampling points of 128 × 2048 are used the processing time is approximately 400 s after 8000 iterations. The processing time is reduced by a factor of 10 by reducing the number of sampling points as expected.

For further reduction of processing time one constraint is added. At the first iteration process in Fig. 1, the values of $f_0(r)$ are given by the final values of $f_{8000}(r)$ at the previous time, i.e., the surface morphology at previous time, as those values have been obtained in a series of phase retrieval from RHEED intensities during growth except for the starting time of the growth. Figures 3(c1–c5) in the bottom row shows surface morphologies after 40 iterations with reference to the morphology at previous time as $f_0(r)$ (RMPT). Figure 3(c1) is obtained by referring the morphology $f_{8000}(r)$ at 0.20 ML (not shown). By comparing the corresponding morphologies between Fig. 3(b1–b5) and Fig. 3(c1–c5), it is clear that basic features of the surface morphology are well reproduced.

Figure 4 shows the variation of the error function in Eq. (2) as a function of the number of iterations. The inset shows the variations for the first 100 iterations. The red solid line shows the variation in the usual HIO process without RMPT. The blue dashed line shows the variation in the present approach with RMPT. The error function in the process with RMPT decreases faster than that in the process without RMPT. Moreover the values of the error function in the process with RMPT remain smaller up to 8000 iterations. From our experience values of the error function in the process with RMPT remain smaller in most cases, but they do not always remain smaller. The variations depend on the scattering amplitudes and the morphologies in question and those at the previous time. Especially when the difference in surface morphologies is small, the error function in the process with RMPT remains smaller.

It is noted that the coverage obtained with RMPT is slightly smaller than that obtained by the usual process. For example, the brighter area in Fig. 3(c2) looks smaller than that in Fig. 3(b2). This is because the effect of retrieved morphology remains in the retrieved morphology in the process with RMPT. It is confirmed that this memory effect becomes less as the number of iterations increases.

III. DISCUSSION

In order to understand the fast convergence of phase retrieval in the process with RMPT, Fig. 5 shows retrieved $f_j(r)$ at first several iterations ($j = 1, 2, 3, 4, 5, 20, 40$). Figures in the upper row show $f_j(r)$ in the usual process without RMPT and those in the lower row show $f_j(r)$ in the process with RMPT. The numbers in the middle show the number of iterations. For the first 5 iterations the shape of the irradiated area with elliptic form is gradually recovered in the usual process, whereas the bright and dark contrasts at previous time gradually changes to those corresponding to the given scattering amplitudes in the process with RMPT. After 20 iterations, some characteristic features are reproduced in both processes. After 40 iterations the characteristic features become clearer. The contrasts in surface morphologies in the lower row always look better than those in the upper row, which corresponds to the lower values of the error function for the morphologies in the lower row. The processing time for the retrieval with RMPT is approximately 0.1 s, which makes it possible to obtain surface morphology almost in real time.

Figure 6 shows the retrieved surface morphologies at 0.33 ML from three different scattering amplitudes, which are calculated from surface morphologies obtained by the kinetic Monte Carlo simulation. The morphology corre-
FIG. 5. Retrieved atomic arrangements in the first 40 iterations by putting 0 values as $f_0(r)$ (upper figures) and by putting values of final $f_{\text{final}}(r)$ in the previous time (lower figures). The numbers in the middle indicate the number of iterations.

FIG. 6. Retrieved atomic arrangements from different RHEED intensity distributions showing the same specular beam intensity (within the error of 0.01%).

Corresponding to Fig. 6(a) is the same as in Fig. 3(a1) and that corresponding to Fig. 6(b) is obtained by using a different sequence of random number generator. The morphology corresponding to Fig. 6(c) is obtained by changing the temperature from 700 K to 850 K. Although the scattering amplitudes are different, the specular beam intensities are almost the same for these three surface morphologies. It is difficult to say the difference in surface morphology from the specular beam intensity. In contrast the different surface morphologies are well reproduced by the present approach. It is clear that real space image obtained almost in real time by using phase retrieval approach is important and useful for observing and controlling the growth by MBE and other growth techniques. This study shows that the fast phase retrieval approach is potentially quite useful in near future once the beam size and parallelity of the electron beam for RHEED is well controlled.

ACKNOWLEDGMENTS

This work was supported by JSPS KAKENHI Grant Number JP25400321. The author would like to express his sincere thanks to Prof. K. Fukutani for useful discussions.

1 J. H. Neave, B. A. Joyce, P. J. Dobson, and N. Norton, Appl. Phys. A 31, 1 (1983).
2 C. S. Lent and P. I. Cohen, Surf. Sci. 139, 121 (1984).
3 T. Kawamura, P. A. Maksym, and T. Iijima, Surf. Sci. 148, L671 (1984).
4 T. Kawamura and P. A. Maksym, Surf. Sci. 161, 12 (1985).
5 T. Kawamura, Surf. Sci. 351, 129 (1996).
6 J. D. Fuhr and F. Müller, Phys. Rev. B 84, 195429 (2011).
7 J. Miao, D. Sayres, and H. N. Chapman, J. Opt. Soc. Am. A 15, 1662 (1998).
8 J. R. Fienup, Opt. Lett. 3, 27 (1978).
9 H. Watanabe and M. Ichikawa, Rev. Sci. Instrum. 67, 4185 (1996).
10 A. Ichimiya and P. I. Cohen, Reflection High-Energy Electron Diffraction (Cambridge University Press, Cambridge, 2004).
11 T. Kawamura, Prog. Surf. Sci. 44, 67 (1993).