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COMPUTER SIMULATION OF REFLECTION HIGH ENERGY ELECTRON DIFFRACTION FROM ULTRATHIN IRON LAYERS

KOMPUTEROWA SYMULACJA ODBICIOWEJ DYFRAKCJI WYSOKOENERGETYCZNYCH ELEKTRONÓW DLA PRZYPADKU ULTRACIENKICH WARSTW ŻELAZA

The results of research work on development of new applications of electron diffraction in metallurgy and materials sience are presented. Namely, we investigate in detail the consequences of the use of reflection geometry which enables "in-situ" examination of a material surface during a selected technological process. We demonstrate that in theoretical interpretation of experimental data collected in such a geometry it is essential to take care about the refraction effects. It means that analyzing experimental data one must account not only the geometrical arrangement of atoms at the surface but also specific values of the scattering potential.

W pracy przedstawiono wyniki badań nad opracowaniem nowych zastosowań dyfrakcji elektronowej w metalurgii i inżynierii materiałowej. W szczególności badane są konsekwencje użycia geometrii odbiciowej, która umożliwia charakteryzację powierzchni materiału bezpośrednio poddanemu procesowi technologicznemu. Zademonstrowano, że teoretyczna interpretacja wyników musi w takim przypadku uwzględniać efekty refrakcji. Oznacza to, że każdy badacz analizujący dane doświadczalne powinien brać pod uwagę w swoich rozważaniach nie tylko ułożenie atomów przy powierzchni lecz także informację o wartościach potencjału rozpraszającego.

1. Introduction

Scientists working in the field of metallurgy and materials science are usually well familiar with basic concepts of electron diffraction. This is due to the fact that from the early 1950's they have applied transmission electron miscroscopy (which is based

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on the phenomenon of electron diffraction) to characterize structurally their samples [1]. However, it is worth pointing out that recently much research effort is devoted to investigations of possible extensions of well established standard routines in this area. Namely, detailed investigations of the consequences of the use of reflection geometry (see Fig. 1) are undertaken (most authors use in references to the work of this type the name of reflection high energy electron diffraction — RHEED). In fact, it is interesting to recall that some concepts of RHEED were developed already in the 1930's. However, RHEED received wide attention only in the 1980's and 1990's as a result of the considerable progress in surface science and engineering achieved during that time. Then it was demonstrated that RHEED may provide very detailed information on changes in structural parameters in a surface layer and it may be applied in-situ (although as with all techniques employing electron diffraction, it must be used under high vacuum conditions). Thus RHEED can be used in a number of preparation processes to monitor changes at the surface. For example, we may mention processes of modification of a surface layer by implantation, covering a material with a coating and preparation of nanoscale, two-dimensional structures. However, while nowadays there is no particular difficulty in conducting RHEED experimental work, a similar statement is not true for theoretical work. Currently, experimental results can be interpreted mostly qualitatively, employing single scattering (kinematical) approaches. If one is interested in the quantitative analysis of data, then one needs to take account of the multiple scattering of electrons. However, a theoretical treatment of this type covering all important experimental situations still does not exist. Thus a number of scientific workers are currently carrying out research work aimed at developing RHEED theoretical approaches which are both accurate and practically manageable.

Electron diffraction can be described in detail with the use of three-dimensional Schrödinger equation with a complex potential. However, if one deals with this

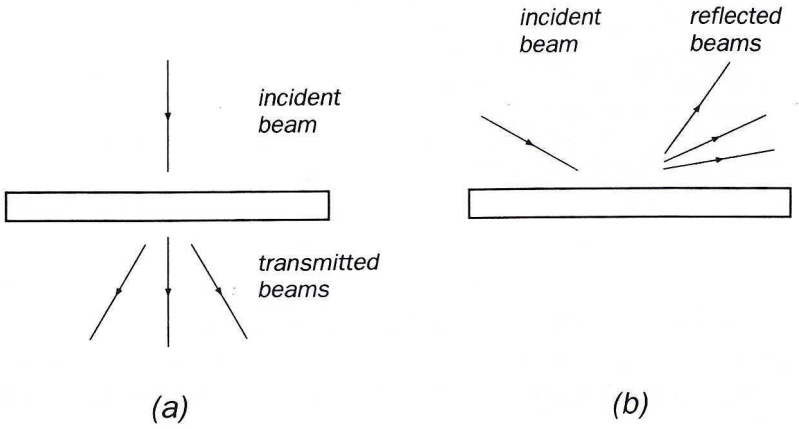


Fig. 1. The schematic representation of the difference between (a) transmission and (b) reflection geometry

fundamental equation without introducing any approximations one is able to solve it only for a very limited number of situations (even assuming the aid of computing machines). In any case, for realistic samples it is impossible to get solutions in such a way. To get close to experiment one must introduce some approximations. For the case of transmission geometry the situation seems to be easier. In theoretical analysis one may neglect backward scattering events. Due to this simplification many effective computing procedures have already been developed. However, for reflection geometry this approach is not valid and developing substantially new approaches turned out to be necessary. As a result of continuous research work during the last ten years a remarkable progress has been achieved in the description of RHEED using approaches assuming two-dimensional periodicity of crystals in planes parallel to the surface and allowing introduction of any changes in crystal parameters along the axis perpendicular to the surface. It seems that currently RHEED from flat surfaces is well understood. Consequently the present research work concentrates on analyzing diffraction from surfaces with steps. In particular, much attention is being paid to theoretical investigations of features of RHEED intensity oscillations. Answering the question of why such oscillations are observed seems to be the very important point in developing a complete theoretical treatment of RHEED.

The phenomenon of RHEED oscillations was discovered in the early 1980's by scientists at the Philips Research Laboratories in the U.K. [2]. It was experimentally found that during the deposition of a material on an intially flat substrate one may sometimes observe regular oscillating changes in the intensity of the specularly reflected electron beam and the period of the oscillations corresponds to the deposition of one atomic layer of the material (see Figure 2). Since then RHEED oscillations have been observed in many laboratories throughout the world engaged in the preparation of new materials. The reason of why researches have been interested in conducting that work is the fact that everybody who wants to modify in a designed way a material surface must consider two questions: (1) what experimental means should be used to carry out the modification and (2) how to control this process. Because in RHEED experiments the electron intensity may oscillate with the period corresponding to the deposition of one atomic layer, it can be concluded that RHEED might allow one to control surface modification processes with nanoscale precision. However, the question of why such oscillations appear is still open. In the past, in one approach it was proposed that intensity oscillations are the results of changes in roughness at the surface [3]. In an alternative approach, it was suggested that oscillations appear because of changes in interference conditions of electron waves reflected from different terraces at the surface [4]. However, both of these approaches (developed respectively by researchers at the Philips Research Laboratories and at the University of Minnesota) were based on single scattering considerations and this is why they could not be accepted as the complete explanation of the phenomenon of RHEED oscillations. Here, we report progress towards working out such an explanation. We use a more precise theoretical

approach, and on the basis of our findings we suggest that changes in the refraction conditions at the surface constitute one more important mechanism responsible for the appearance of RHEED oscillations.

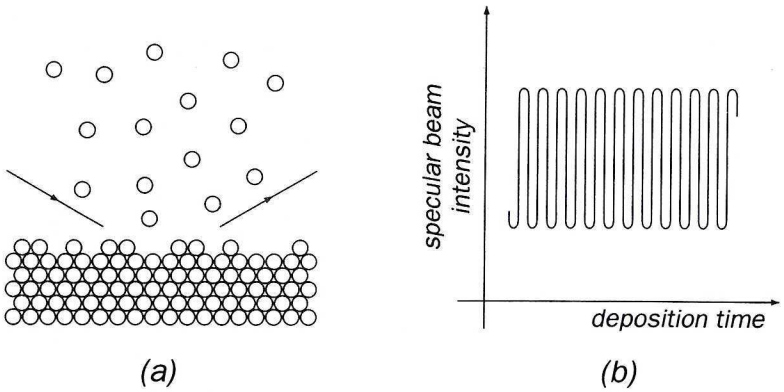


Fig. 2. The schematic illustration of the phenomenon of RHEED oscillations: (a) measurements of the intensity of the specularly reflected electron beam during the deposition of atoms at the surface, (b) the corresponding plot of the measured intensity

2. The description of a theoretical model

We start from the assumption that intensity of elastically scattered electrons can be calculated using the Schrödinger equation

$$\nabla^2 \Psi(\boldsymbol{r}) - v(\boldsymbol{r})\,\Psi(\boldsymbol{r}) + K^2\,\Psi(\boldsymbol{r}) = 0, \tag{1}$$

where

$$v(r) = \left(1 + \frac{|q_e|U}{m_0 c^2}\right)\frac{2m_0}{\hbar^2}\,V(\boldsymbol{r}), \tag{2}$$

$$K^2 = \left(1 + \frac{|q_e|U}{2m_0 c^2}\right)\frac{2m_0}{\hbar^2}\,|q_e|\,U. \tag{3}$$

In Eqs. (1)–(3), $V(\boldsymbol{r})$ means the three-dimensional scattering potential of a crystal and U means the absolute value of the accelerating voltage of the electron gun. m_0, q_e and $\hbar$ are the electron rest mass, the electron charge and Planck's constant. However, in computational studies presented in this paper we use a simplified approach. Namely, we replace the three-dimensional potential $V(\boldsymbol{r})$ (see Equation 2) by a one--dimensional potential $V(z)$ taking the mean in each plane parallel to the crystal. The introduction of the one-dimensional model of the potential simplifies all considerations. However it is still possible to find experimental conditions for which such a model is appropriate, namely, the azimuth of the incident beam should be fixed few degrees off a symmetry crystal direction.

Assuming the one-dimensional model of the potential, above the crystal ($z > z_T$), we can write the solution $\Psi(r)$ in the form

$$\Psi(\boldsymbol{r}) = \exp(i\boldsymbol{k}_{\parallel} \cdot \boldsymbol{\varrho})\left[\exp(-ik(z-z_T)) + R\exp(+ik(z-z_T))\right], \tag{4}$$

where $\boldsymbol{k}_{\parallel}$ and $\boldsymbol{\varrho}$ are parallel components of $\boldsymbol{K}$ and $\boldsymbol{r}$. Further, $k = |K| \sin \vartheta$, where ϑ is the glancing angle of the incident beam. R is an unknown amplitude o the reflected beam. It is assumed that the crystal extends below the plane $z = z_T$. The scattering potential $V(z)$ can be determined using electron scattering factors tabulated by D o y l e and T u r n e r [5]. The contribution to $V(z)$ coming from the j-th atomic layer can be expressed in the following way

$$V_j(z) = -\Theta_j(1+0.1\,i)\frac{4\hbar^2\pi^{3/2}}{m_0\Omega}\left[\sum_{n=1}^{4}\frac{a_n}{b_n^{1/2}}\exp\left(-\frac{4\pi^2}{b_n}(z-z_j)^2\right)\right], \tag{5}$$

where Θ_j is the layer coverage (the ratio of the number of places occupied by atoms to the number of all atom sites), Ω is the area of the two-dimensional surface unit cell and z_j is the layer position along the axis perpendicular to the surface. The coefficients a_n, b_n are given by D o y l e and T u r n e r [5]. In our model the potential from the partially completed layer is proportional to its coverage [see the multiplier in Equation (5)]. The potential of the whole crystal is determined by summing contributions from all layers, i.e. $V(z) = \sum_j V_j(z)$. In our model we assume that an electron beam reflected from the crystal bottom is neglectably small. In computational practice we realise this by considering a crystal which is composed of two thousand atomic layers. Additionally, we assume that our crystal is grown in a layer-by-layer mode. It means that for the crystal topmost layer its coverage may be equal to any value from the range 0 to 1 (inclusively). However, for all other atomic layers the coverages Θ_j are equal to 1. Finally, using numerical codes developed within the framework of the two-dimensional Bloch wave approach (for details see [6]), we are able to determine the amplitude R of the specular beam. Then the intensity I of this beam is given by $I = |R|^2$.

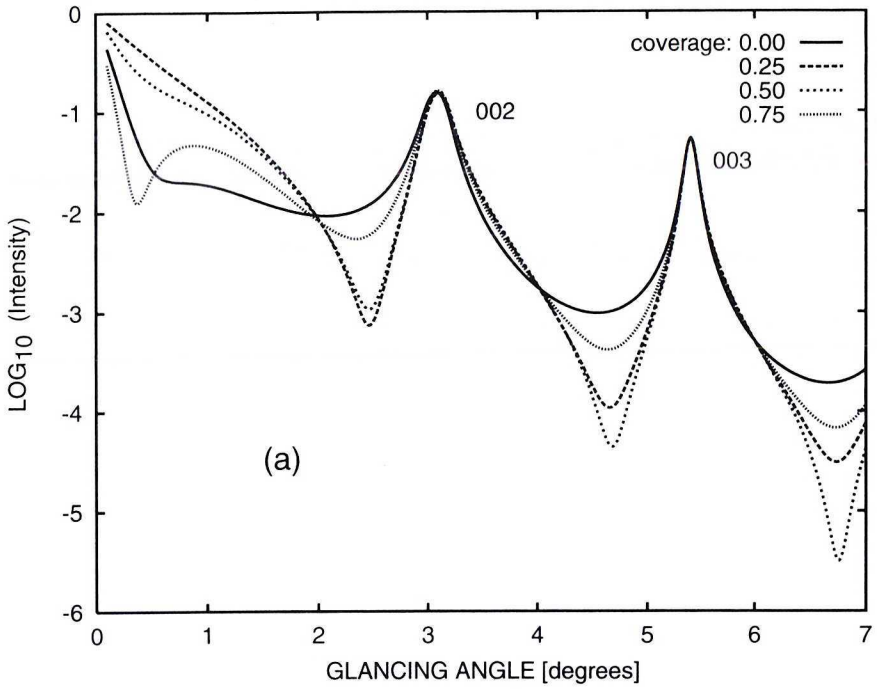
3. Calculations and results

We carried out a series of calculations corresponding to deposition of an iron layer on the flat substrate of the same material (see Fig. 3a). In order to get a deeper understanding of these results we also carried out computations for a geometrically identical situation, but for "artificial" Fe-like atoms, i.e. atoms with a scattering potential reduced by factor of 10 (see Fig. 3b). In such a way we are able to distinguish effects due to geometrical arrangements of atoms in a crystal from the effects caused only by the specific values of the crystal scattering potential. We

calculated the intensity of the specularly reflected electron beam as a function of the glancing angle determined for the incident beam (such a plot is called a rocking curve). Both for case (a) and (b) we calculated a series of four rocking curves for the following coverages of the topmost atomic layer: 0.00, 0.25, 0.50 and 0.75. It should be added that the coverages of 0.00 and 1.00 represent the situation when the surface of the crystal is flat, and due to the periodicity of crystal properties the corresponding rocking curves are identical.

It is useful to begin the description of our findings by discussing the results presented in Figure 3 b. The rocking curves of Figure 3 b show a high level of regularity; for completed and partially filled tompost layers one may easily recognize the presence of Bragg peaks. We may easily observe that for partial coverages of the topmost layer (i.e. for 0.25, 0.50 and 0.75) the intensity between Bragg peaks is smaller than for flat surfaces (i.e. for coverages of 0.00 and 1.00). However, the situation shown in Figure 3b represent the limiting case of what would happen if the scattering potential of atoms is "very small". The calculations for real atoms are presented in Figure 3a. The pattern for this case shows many similarities with the pattern of Figure 3b. Nevertheless, two important new effects appear: (1) Bragg peaks are shifted towards lower angles in comparison with position from Figure 3b (in fact the first peak even disappears), (2) Bragg peaks for partially filled tompost layers occur slightly shifted towards higher angles in relation to corresponding peaks for completed layers. These two effects are caused by refraction, i.e. one can say that they are consequences of the specific, finite values of the scattering potential rather than a consequences of the geometrical arrangement of atoms in a crystal. The second effect has very important implications for the appearance of RHEED intensity oscillations. Namely from Figure 3 a we can conclude that for angles lower than Bragg peaks determined for flat surfaces, the intensity should decrease after starting the deposition of a material, but for angles higher than Bragg peaks determined for flat surfaces, the intensity should increase after starting the deposition.

Fig. 3. (a) RHEED rocking corves for crystals composed of Fe atoms, (b) RHEED rocking curves for crystals composed of atoms for which the scattering potential is taken to be the potential of a Fe atom multiplied by the factor of 0.1, Both for case (a) and (b) the geometry of a Fe(001) crystal is assumed and the energy of the incident electron beam is taken to be 15 keV. RHEED rocking curves are computed for four different coverages of the tompost layer


coverage: 0.00
0.25
0.50
0.75
002
003
(a)
LOG10 (Intensity)
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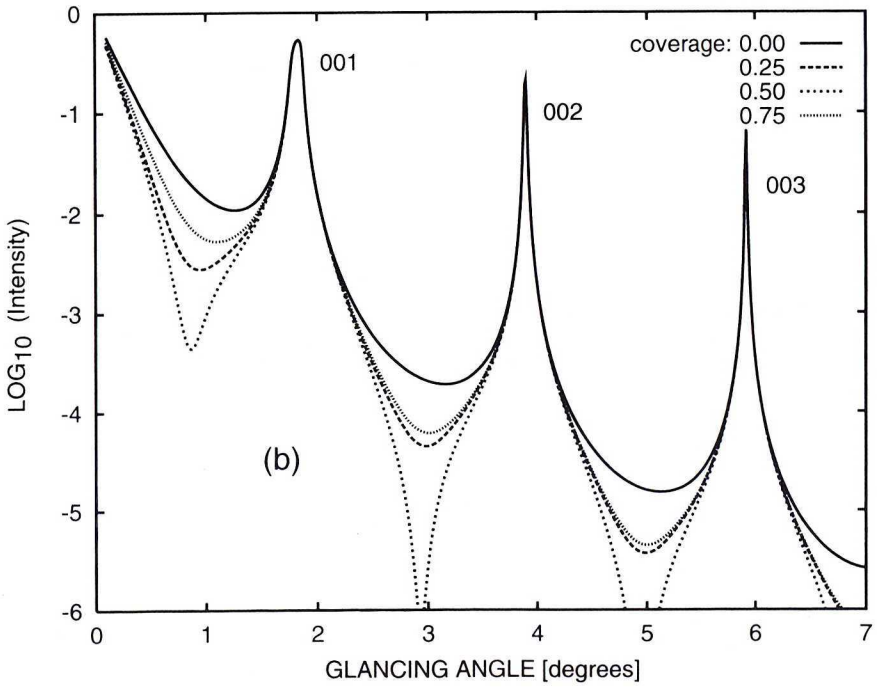
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(b)
LOG10 (Intensity)
GLANCING ANGLE [degrees]

4. Discussion and conclusions

An introductory discussion of the importance of the phenomenon of refraction for RHEED has been given in the past by Larsen et al. [7]. In the early 1990's theoretical investigations of the implications of the use of the one-dimensional model which we employ in this paper were started in a number of research groups [8–10]. Already then it was concluded that refraction might be of special importance for the appearance of RHEED oscillations. Next these initial findings have been confirmed by the interpretation of a few sets of experimental data [11–15]. It seems that the most important confirmation follows from the interpretation of the oscillation phase for GaAs(00) — 2×4 [14, 15]. The successful practical applications of the one-dimentional model motivated us to carry out additional theoretical studies on this model to elucidate the mechanism of the refraction. In this paper we demonstrate our most recent findings. We carried out computations for thin iron layers which are of technological importance both for traditional metallurgy and modern materials science [16, 17], whereas usually most papers in the literature refer to semiconductors. We believe that our theoretical results encourage researchers dealing with experiment to carry out a number of detailed measurements for iron layers actually grown.

In summary, the importance of accounting of the refraction in the analysis of RHEED experiments from stepped surfaces is demonstrated.

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