Role of the Step Density in Reflection High-Energy Electron Diffraction: Questioning the Step Density Model

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The step density model of reflection high-energy electron diffraction oscillations is investigated. Within this model, the temporal evolution of the specular beam intensity during growth by molecular beam epitaxy represents the evolution of the step density during deposition. This is found to be inconsistent with diffraction theory. In particular, when the concentration of atoms in the deposited layer is fixed, an increase of the step density causes an increase of the specular beam reflectivity, contrary to the prediction of the step density model. [S0031-9007(97)02678-1]

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The standard experimental technique for monitoring the growth of ultrathin films and advanced materials by means of molecular beam epitaxy (MBE) is reflection high-energy electron diffraction (RHEED). The reason for this is the high sensitivity of RHEED to surface structure and morphology combined with its excellent compatibility with MBE. The molecular (or atomic) beam is incident almost normal to the surface, whereas the RHEED electrons hit the surface at grazing incidence so the diffraction pattern can easily be observed *during* deposition. The most commonly exploited feature of the diffraction pattern is the temporal RHEED oscillation, i.e., the periodic variation of the intensity of the specularly reflected beam. This corresponds directly to the period of monolayer incorporation [1] and allows control of growth with monolayer precision.

The occurrence of RHEED oscillations is qualitatively understandable because the state of the surface passes from monatomically flat to disordered to flat during layer by layer growth. However, strong multiple scattering complicates the interpretation of RHEED intensities and an exact general theory of RHEED oscillations is not yet available, although it has been shown rigorously that oscillations occur in a number of systems and models [2–8]. An alternative to diffraction theory is the very general proposal that the temporal evolution of the specular beam intensity directly reflects the evolution of the step density during growth. This is called the step density model and was deduced empirically by comparison of experimentally measured RHEED oscillations with step densities obtained from Monte Carlo simulations of growth by means of SOS (solid-on-solid) models [9,10]. The physical argument given in support of the step density model is that each step acts as a localized source of diffuse scattering and causes a reduction of the specularly reflected intensity.

Although this approach appears intuitively reasonable ("a flat surface reflects better than a rough one") the evidence for the step density interpretation is highly empirical and supporting arguments relying on scattering theory are lacking. Nevertheless, the step density model has been widely applied to interpret the temporal behavior of specular RHEED intensities [11–18] and it seems that this interpretation has developed into an established procedure, particularly after steps in STM images were found to be correlated with RHEED intensities [19]. Recent work has pushed the step density model even further, and it has been used to derive surface diffusion parameters such as energy barriers [11,12] and preexponential factors [18] from RHEED measurements.

In this work we investigate the influence of the step density ρ on the specular intensity from the viewpoint of scattering theory. The step density model is found to be highly questionable because both multiple scattering calculations and arguments based on the dynamical (multiple scattering) theory of RHEED consistently show that an increasing step density alone tends to produce an *increase* of the specularly reflected intensity rather than a decrease. We show that this behavior is linked to the grazing incidence geometry of RHEED, the high electron energy, and the fact that the step density does not affect the averaged periodic part of the surface scattering potential.

During growth the surface is in a more or less disordered state where, to a good approximation, the atoms occupy regular lattice sites. If one divides the scattering potential $V = V_p + \delta V$ of each disordered layer into a periodic part V_p and into a nonperiodic part δV, V_p will have the same symmetry as if the layer were perfectly ordered. Therefore if δV is taken to be a perturbation, the term of zeroth order in δV only contributes to the sharp diffracted beams, the first order term contributes to the diffuse background but does not affect the sharp diffracted beams and the second and higher order terms both contribute to the background and influence the intensity of the beams. V_p is simply given by the potential of the perfectly ordered layer reduced by the factor θ which denotes the coverage (i.e., the concentration of occupied sites) of the disordered layer. V_p is not affected by the step density and it is clear that the variation of V_p (via θ) during growth potentially leads to a temporal change of the diffracted intensities [6]. The δV term is affected by the step density and for fixed coverage ($0 < \theta < 1$) various magnitudes of step densities can be realized, depending on the typical terrace size at the surface (θ and ρ are not "*a priori*" correlated). This means that θ and ρ both affect the diffracted intensities, *but in different orders of the scattering series corresponding to the different potential parts they contribute to.*

In order to elucidate the role of the step density in RHEED, it is therefore important to distinguish the effects of changes in the step density from the effects of changes of the coverage. During MBE growth it is experimentally very difficult to satisfy this requirement. The simulation of RHEED intensities from model structures, however, offers a way out of this problem. We have calculated the diffracted intensities from structures where the step density was varied but the coverage was kept constant. As the higher order (diffuse) scattering due to δV is essentially determined by the disorder along the incident beam azimuth [20], it is sufficient to concentrate on one-dimensional disorder where the incidence azimuth of the electron beam is perpendicular to the step edges.

For the simulations we used the dynamical theory of RHEED in conjunction with a supercell technique. Here, the disorder is modeled within a large unit cell (supercell) which is repeated periodically. Thanks to recent advances in computational capacity and program optimization, the use of supercells with several hundred lattice units extension is now possible [20,21]. Such calculations give the RHEED intensities to infinite order in δV and correctly take into account the influence of the diffusely scattered electron wave function on the part of the wave function belonging to the sharp diffracted beams. This is a very important requirement regarding the particular problem treated in this work.

The disorder was assumed to follow a one-dimensional geometrical terrace size distribution [22,23], restricted to two levels. As will become evident below, the exact choice of model is unimportant for our arguments. The key point is that with various step densities various degrees of short range correlations are produced. The probability of encountering a downward step from the top level of the surface on going from one lattice site to an adjacent one is p_d. Analogously, p_u is the probability for an upward step from the second to the top level. This model produces the coverage $\theta = p_u/(p_d + p_u)$ and the step density $\rho = \theta p_d + (1 - \theta)p_u$. These equations uniquely relate each combination of ρ and θ to a combination of p_d and p_u.

The calculations were carried out for an unreconstructed, stepped Si(100) surface with bilayer terraces and step edges along the $[011]$ direction. Si is one of the most commonly used materials in thin film growth. Bilayer steps frequently appear at the Si(100) surface and simultaneously offer the possibility of using a defect feature that produces within the scope of our model disorder in more than just one layer. Finally, extensive computational experience for this system, particularly with regard to disorder, is available [20].

The disorder configurations were generated within supercell 400 surface lattice units (1 LU $= 3.84$ Å) long by means of random numbers and the jump probabilities defined above. This allowed us to produce configurations with different step densities at constant coverage. The extension of the supercell is of the same order of magnitude as the distance within which electrons under typical RHEED conditions are scattered coherently. The calculations were carried out for an electron energy of 15 keV, the incident beam azimuth was $[01\bar{1}]$, and 400 beams (diffuse and sharp) were included. For computational details we refer to Ref. [20].

Figure 1 shows the calculated specular beam rocking curve (reflectivity versus incident angle) for the angular range $0°$–$3°$. This range is typically used to control growth. The three curves plotted in each panel correspond to different step densities but the same coverage. For constant coverage, the general shape of the plots is quite similar for all step densities (see also Fig. 2). This similarity is due to the common periodic potential which mainly determines the shape of rocking curves [20,24]. However, the absolute reflectivity depends significantly on ρ. The most striking fact is that the reflectivity systematically *increases with increasing step density*. This behavior is exactly opposite to the prediction of the step density model and is found for all the three coverages. It is particularly significant that this finding also holds for the "in-phase conditions" at 1.05° and 2.1°, where different terrace levels would kinematically interfere constructively and the step density interpretation is supposed to be particularly suitable [11].

What is the physics driving this, at first glance peculiar, behavior? Corresponding to the potential parts V_p and δV there are two types of scattered wave. First, the set of strongly excited waves ψ_p which corresponds to the sharp diffracted beams generated by the periodic potential V_p, and second, the diffusely scattered waves ψ_d generated by the nonperiodic potential δV. Both wave sets can interact by multiple scattering via δV, i.e., an electron belonging to ψ_p can be scattered into states corresponding to ψ_d and vice versa. In terms of this interaction the above finding becomes understandable.

As explained above, to zeroth order in δV, the intensity diffracted into the sharply defined beams (e.g., the specular beam) *does not explicitly depend on the step density*. A dependence on the step density appears only if one accounts for higher orders through the multiple scattering interaction between the wave fields ψ_p and ψ_d. The strength of this interaction not only determines the strength of ψ_d, but also the *feedback* of ψ_d into the wave field ψ_p. Physically, this feedback is responsible for the loss of diffracted intensity due the diffuse scattering [25]. Hence, it is essential to understand how the strength of this feedback depends on δV.

Let $k_{\mathbf{s}}$ denote the perpendicular (to the surface) component of the vacuum wave vector of a diffuse wave that corresponds to the two-dimensional (parallel to the

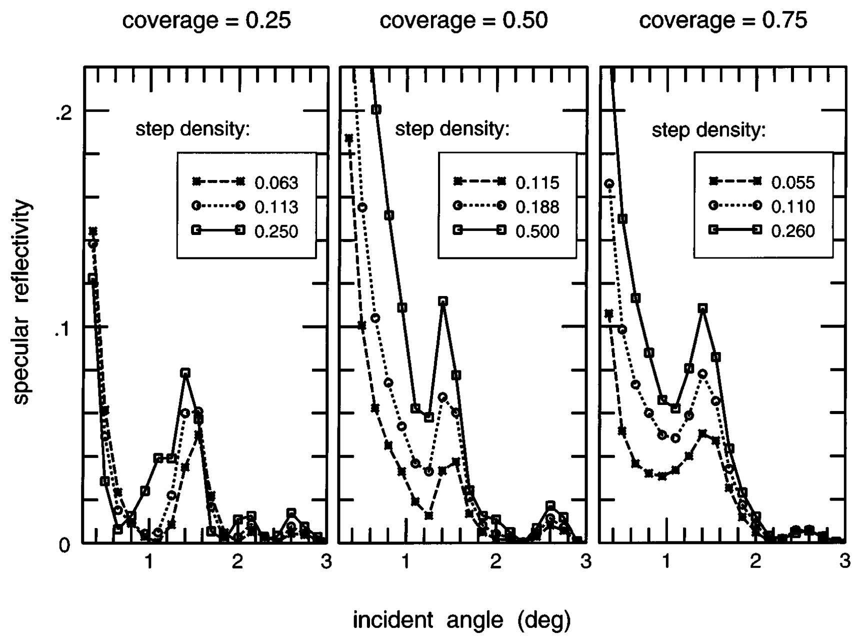

FIG. 1. Specular beam rocking curves from Si(100) for various step densities and coverages. The electron energy is 15 keV, the incident beam azimuth is $[01\bar{1}]$.

surface) reciprocal vector $\mathbf{s}$. For evanescent waves $k_{\mathbf{s}}$ is imaginary. It is now essential to recall the following three properties of the surface potential and the electron wave field in RHEED. Each of these properties alone is well known in RHEED and diffraction physics. It is their combination that explains the computational findings.

(i) The probability that a wave $\mathbf{s}$ of the diffuse set ψ_d couples to the set of strong waves ψ_p generally increases with decreasing $|k_{\mathbf{s}}|$. The physical reason for this is that in the RHEED geometry waves with low $|k_{\mathbf{s}}|$ tend to be strongly excited [20,26,27] and move nearly parallel to the disordered surface layer. Thus, the probability of being scattered by the nonperiodic potential δV is very high. Waves with high $|k_{\mathbf{s}}|$ are usually only weakly excited. Furthermore, if these waves are propagating ones, they will tend to propagate out the surface and be scattered in the ordered bulk where $\delta V = 0$ and a coupling with ψ_p cannot occur.

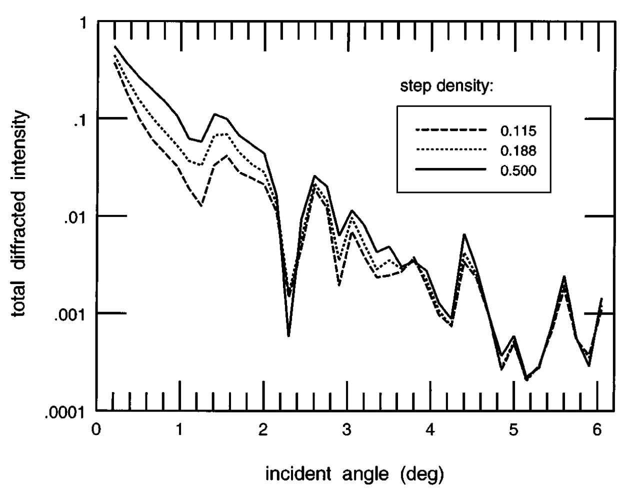

FIG. 2. Total diffracted intensity versus incident angle from Si(100) for various step densities at half coverage. The electron energy is 15 keV, the incident beam azimuth is $[01\bar{1}]$.

(ii) Because the parallel component of the incident wave vector is very large, energy conservation requires that a small momentum transfer in the direction of the incident beam azimuth is connected with a large change of $|k_{\mathbf{s}}|$.

(iii) In reciprocal space, δV has strong values only for those $\mathbf{s}$ which are situated within an "intensity region" around each reciprocal surface lattice vector. The extension Δs of the region is about $2\pi/L$ where L denotes the typical length of short range correlations in the system. For a stepped surface L is related to the mean terrace width. It is now physically evident that L monotonically decreases with the step density ρ and Δs increases with ρ. For the geometrical terrace size distribution in one dimension and fixed coverage, $\Delta s \propto \rho$ holds to a very good approximation.

The latter behavior in conjunction with property (ii) means that with increasing step density the majority of the $|k_{\mathbf{s}}|$ components of the relevant diffuse waves becomes large. This is nothing but the fact that a RHEED pattern becomes more streaky if the lateral short range order parallel to the incident beam azimuth is reduced. Waves with large $|k_{\mathbf{s}}|$ components, in turn, tend to couple only weakly with the strong waves [property (i)]. Consequently, *the multiple scattering interaction between the strong (diffracted) waves and the diffuse waves decreases with increasing step density and for this reason the loss of diffracted intensity due to the diffuse scattering is expected to behave in the same manner*.

In Fig. 2 we show the loss of the diffracted intensity, calculated for half coverage and high step densities up to $\rho = 0.5$. It has been claimed that especially for $\theta = 0.5$

and high ρ the step density model applies particularly well [10]. In the figure the *total diffracted intensity* (i.e., the sum of the intensity of all integer beams) is plotted versus the incident angle. This time, the intensities are plotted on a logarithmic scale and the calculations were carried out for incident angles up to 6°. It is evident that the total diffracted intensity systematically increases if the step density increases. It is noteworthy that this holds for an intensity range that covers about three orders of magnitude.

Our arguments are also supported by the results of a recent investigation [20] concerning the strength of this interaction, expressed in terms of the magnitude Δc of the corresponding coupling terms in the system of coupled differential equations describing the wave field in the crystal. The dependence of Δc on the coverage and the correlation length approximately follows the law

$$\Delta c \propto \theta(1 - \theta)L\,, \tag{1}$$

provided that the correlation length L is not too large (i.e., in the regime of high step densities, for details we refer to Ref. [20]). Because L increases with decreasing ρ, the dependence clearly points to the above discussed influence of the step density on the diffracted intensities. We also emphasize that the dependence on ρ (or L) has to be considered if phenomenological absorption potentials for RHEED calculations are constructed in order to model the loss out of the diffraction channels. Inclusion of the coverage dependence as the only structural quantity [28] is obviously not sufficient.

Equation (1) also shows that the coverage θ contributes to the intensity loss. Therefore the effect of θ, is more complicated than its influence on the diffracted intensities via V_p. This might explain why empirical inverse correlations between step density and specular reflectivity have been found in STM data [19]. In the STM studies changes of ρ are accompanied by changes of θ and, interestingly, STM images [19] with low/high ρ, systematically correspond to cases where, within the typical coherence length of RHEED, the factor $\theta(1 - \theta)$ is low/high. According to the results of the present work, however, it is difficult to justify the assumption that the physical reason for any decrease (increase) of the specular RHEED intensity is the increase (decrease) of the step density.

In summary, we have determined the influence of the step density on the diffracted intensities in RHEED and shown that RHEED oscillations during crystal growth cannot simply be explained in terms of an oscillating step density. Our arguments indicate that the temporal evolution of the specular beam intensity is determined both by the short range correlations (expressed, for instance, in terms of the step density) and by the coverage. Both are potentially oscillating quantities and both can affect the RHEED intensities. Our calculations and investigation of higher order diffuse scattering consistently show that in the high step density regime an increase of the step density at constant coverage is correlated with an increase of the specular beam intensity instead of a decrease, and this is contrary to the commonly assumed model.

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