Origin of Electron Diffraction Oscillations during Crystal Growth

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Measurements of the intensity oscillation phase of reflection high-energy electron diffraction during molecular beam epitaxy growth of GaAs and AlAs indicate that the oscillations are due to an interference effect within the surface reconstruction layer forming on the growing layer. The experimental results along a low-symmetry azimuth are explained by a basic theoretical model using only the layer thickness as a fitting parameter. Our conclusions are supported by energy loss measurements showing the absence of diffuse inelastic contributions with a different phase. [S0031-9007(98)06277-2]

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Reflection high-energy electron diffraction (RHEED) is widely used as an *in situ* characterization technique during molecular beam epitaxy (MBE) since the electron beam impinges on the sample at grazing incidence and does not interfere with the molecular beam geometry. It can therefore probe the surface in real time during crystal growth. When intensity oscillations of the diffraction pattern during crystal growth were discovered [1–3], it was immediately recognized that the period of the oscillations corresponded to the deposition of one lattice plane, usually consisting of two atomic layers for a binary compound like GaAs. This has led to a widespread use of RHEED oscillations for surface characterization and growth rate calibration in MBE.

The oscillating intensity can be explained by the changing surface morphology, alternating between completed layers and a rougher intermediate state. However, a more detailed understanding of the diffraction process generating the oscillations is complicated by strong multiple scattering effects. Numerical treatments of realistically large surface unit cells are still out of reach for current computers. Meanwhile, an important remaining problem is the explanation of the RHEED oscillation phase dispersion measured at the specularly reflected position as a function of electron beam incidence angle [4]. The positions of the oscillation minima strongly vary, assuming all possible values and showing no obvious periodicity. Furthermore, the phase of the oscillations depends on the As_4 overpressure during the rate-limiting deposition of Ga with different slopes for different surface reconstructions [5].

The cyclic variation of the oscillation amplitude with incidence angle points towards a kinematical model [6]. This model assumes interference of beams reflected from the top of the growing layer with electrons reflected from the uncovered areas exposing the lower level the layer is deposited on. It predicts oscillations for incidence angles different from the bulk Bragg condition, where the two beams interfere destructively. The minimum of the oscillations always occurs at half-layer coverage, and the oscillation phase is constant. This is not observed experimentally. To overcome this discrepancy, diffuse scattering processes have been proposed that would give rise to an intensity variation proportional to the surface step density [7,8]. In a simplified picture, the intensity variation at the Bragg condition would then be proportional to the step density, whereas for other incidence angles, the combination of diffuse and kinematical scattering could explain the observed phase dispersion as a function of incidence angle. The step density model has been extensively used to interpret RHEED data [9]. At the same time, theoretical treatments still disagree on whether the step density should contribute proportionally [10] or antiproportionally [11] to the RHEED intensity.

In this Letter, we explain the variation of the oscillation phase as a function of the beam incidence by an elastic multiple scattering process. Experimental investigations of this mechanism lead us to a new model that links the phase of the oscillation to the surface reconstruction during growth. Our model does not include the step density, suggesting that it is not an important quantity in the explanation of RHEED oscillations.

Because of the large scattering cross sections of electrons, RHEED in most cases involves strong multiple scattering. A simple model involving multiple or dynamical scattering is shown in Fig. 1(a). It is based on more complicated models [12–14] inspired by multislice dynamical theory. The reflectance of the surface is calculated from the interference of the beam reflected from the top of the growing layer with the one refracted by the top and reflected from the bottom interface. Growth of one layer is simulated by linearly increasing the layer potential from zero to the substrate value, at which the bottom interface vanishes. Since the surface parallel component of the wave vector remains unchanged, the mathematics of the problem reduces to the textbook example of a one-dimensional quantum mechanical particle incident on a twofold downward potential step. This layer interference model can be regarded as the simplest dynamical (multiple scattering) treatment, since it takes into account only the zeroth order Fourier component of the electronic potential inside the crystal. It has two free parameters, the potential V and the layer thickness d. Absorption is ignored since it does not significantly affect the results [15].

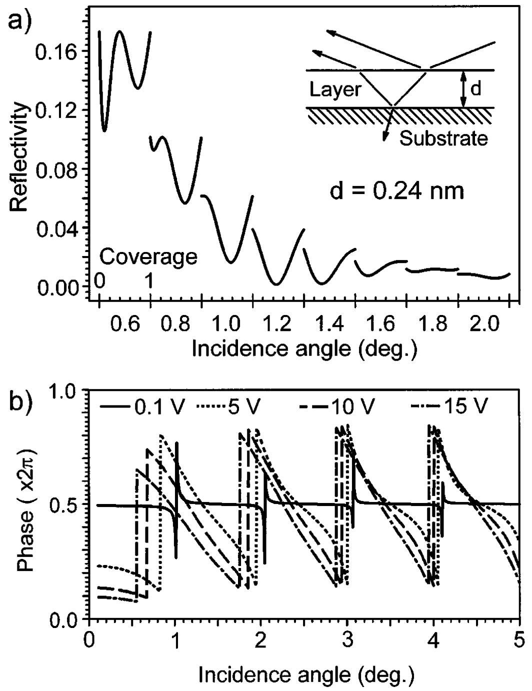

FIG. 1. Layer interference model. The reflectance change during the deposition of one layer is shown in (a) for different incidence angles together with the geometry used for the calculations. Two amplitude minima at $\approx 0.7°$ and $\approx 1.8°$ are shown. Phase dispersion plots (position of the oscillation minimum as a function of incidence angle) for different potential values are given in (b).

Simulations using $V = 10.5$ V, $d = 0.24$ nm, and 20 keV electrons are shown in Fig. 1(a) for a range of incidence angles. The reflectivity always assumes a maximum for completed layers, which leads to sharp peaks at these positions. We therefore define the phase of the oscillations as the relative position of the minimum, being π for a minimum at half coverage. The sharp maxima at integer coverages, although occasionally observed on GaAs for very smooth surfaces [16], are usually absent due to interference with other levels if the growth front is distributed among several layers [13]. The reflectivity decreases with increasing incidence angle, and the amplitude of the oscillations shows periodic local minima. Both of these features are observed in experiments [6]. In addition, the layer interference model predicts strong oscillations with double minima at low angles that have been observed in metal epitaxy [17,18], as well as a strong phase dispersion.

The oscillation phase is plotted as a function of incidence angle and potential V for $d = 0.24$ nm in Fig. 1(b). The positions of the phase jumps coincide with the amplitude minima and can be regarded as generalized Bragg conditions. In the limit of very small V we obtain the kinematical case with no phase dispersion and evenly spaced Bragg conditions at the positions an x-ray diffraction experiment would produce. For larger V, the phase shows a sawtooth behavior with strong dispersion and a displacement of the Bragg conditions towards the shadow edge. Variation of d (not shown) stretches or compresses the diagram along the horizontal axis.

The potential V can be obtained experimentally from a fit to the Kikuchi line pattern [15,19–21]. The results are shown in Fig. 2(a) for a GaAs $\beta(2 \times 4)$ and in Fig. 2(b) for an AlAs $c(4 \times 4)$ reconstructed surface. The points indicate the kinematical bulk diffraction spot positions used for the construction of the line pattern. For both surfaces, a value of $V = 10.5 \pm 0.5$ V is obtained. We therefore use $V = 10.5$ V for the calculations throughout this work.

Since the layer interference model does not take into account lateral potential modulations, a corresponding experiment has to be performed away from high-symmetry azimuths to avoid influences from lateral diffraction. This so-called one-beam condition corresponds to a diffraction pattern with a minimum number of Kikuchi lines. In our measurements, we have therefore chosen a position close to the $[\bar{2}10]$ azimuth and measured the RHEED oscillation phase as a function of incidence angle for both GaAs and AlAs homoepitaxy.

The results are shown in Fig. 3. In 3(a) and 3(b), the circles denote the experimental data, whereas the thicker solid lines are fits using the layer interference model. Except for some discrepancies at very low angles, the theory is in excellent agreement with the data, indicating that the model is valid. Since the lattice mismatch between

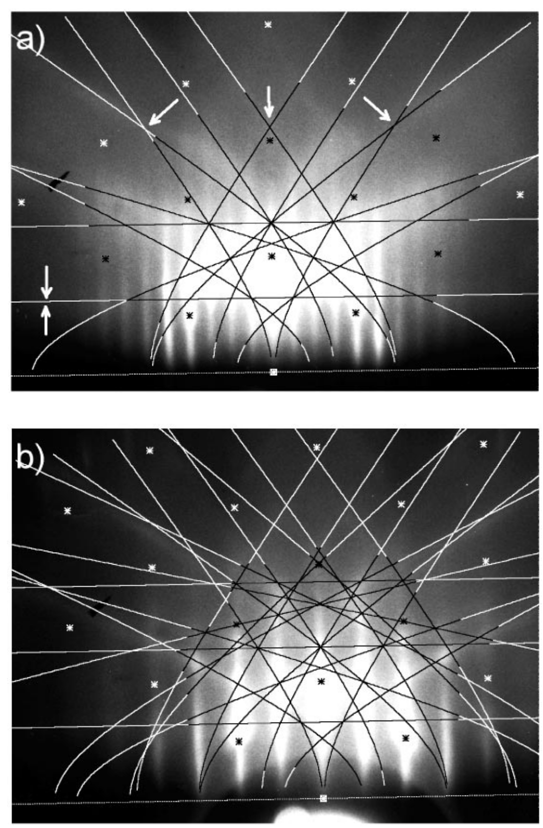

FIG. 2. Kikuchi line fits to diffraction patterns of (a) GaAs (001) $\beta(2 \times 4)$ and (b) AlAs (001) $c(4 \times 4)$ reconstructed surfaces. Arrows indicate the most reliable points for fitting.

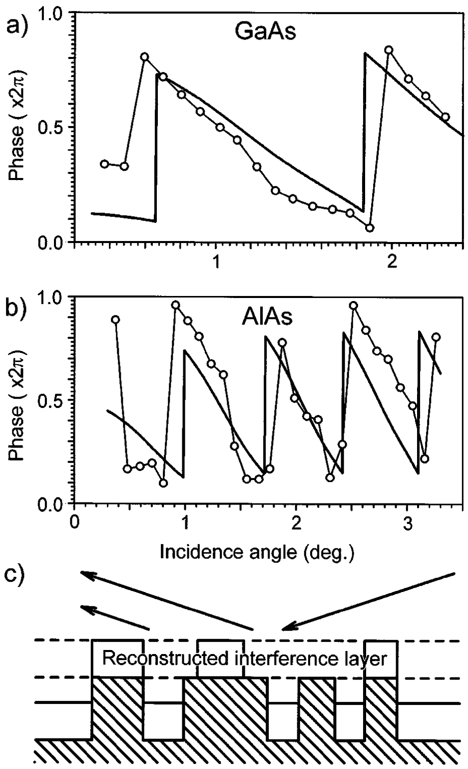

FIG. 3. Comparison of experimental data recorded in the one-beam condition with layer thickness fits obtained from the layer interference model. In (a), a value of $d = 0.24$ nm is obtained; the result in (b) is $d = 0.38$ nm. The different thicknesses obtained for materials with practically identical bulk lattice constants leads to the model shown in (c), where the interference takes place in the surface reconstruction on top of the growing layer.

GaAs and AlAs is well below 0.1%, their layer thicknesses can be regarded as identical for the purpose of this study. However, a fit using the bulk lattice constant for both does not agree with the experiment. Instead, we obtain $d = 0.24 \pm 0.02$ nm for GaAs and 0.38 ± 0.02 nm for AlAs. This means that the interference does not take place in the growing bulk structure layer.

GaAs and AlAs differ strongly in their surface reconstructions. In Fig. 3(a), the surface reconstruction was $\beta(2 \times 4)$, whereas in Fig. 3(b), the $c(4 \times 4)$ reconstruction was observed. We are therefore led to the conclusion that the interference takes place between the top surface and the reconstruction–bulk structure interface. This configuration is shown in Fig. 3(c). Shaded areas represent bulk structure material, and open areas indicate the surface reconstruction. During growth, the reconstruction forms on the new layer as it increases in area. Since the energy gained in surface reconstruction is significant, the delay of surface reconstruction on the growing layer is presumably small, and the reconstructed surface area closely follows the layer coverage. The values found for the layer thickness d agree remarkably well with the structural data of the $\beta(2 \times 4)$ and $c(4 \times 4)$ surface reconstructions [22,23], which consist of one and one and a half atomic bilayers, respectively. In both cases, the reconstructed layers are incomplete and relax towards the bulk.

While in general it is difficult to experimentally separate diffuse scattering components from elastic scattering, this is possible for diffuse scattering involving plasmon and band-to-band losses. We performed RHEED oscillation measurements on GaAs (001) and AlAs (001) as a function of energy loss and incidence angle. The energy filter consisted of two fine pitch gold nets in front of the RHEED screen and is described elswhere [24]. At 20 keV electron energy, the energy resolution was better than 2 eV. A typical spectrum obtained from (2×4) reconstructed GaAs (001) is shown in Fig. 4(a). Apart from the elastic peak, single and twofold surface plasmon losses (11 and 22 eV) dominate the spectrum. For the chosen material and diffraction conditions, the elastic contribution (≤ 2 eV energy loss) increases monotonically from zero at small incidence angles and constitutes approximately 20% of the total intensity around 1° incidence angle. Figures 4(b) and 4(c) show RHEED intensity oscillations from the same surface chosen so that the two sets of curves are out of phase to each other. The measurements were performed on the specularly reflected spot. The energy filter was adjusted so that either only the elastic peak or additionally single and double plasmon losses were included. Within each panel, the lower three curves represent the measured data on the same intensity scale, whereas the top three curves are normalized to their pregrowth intensity and shifted for clarity.

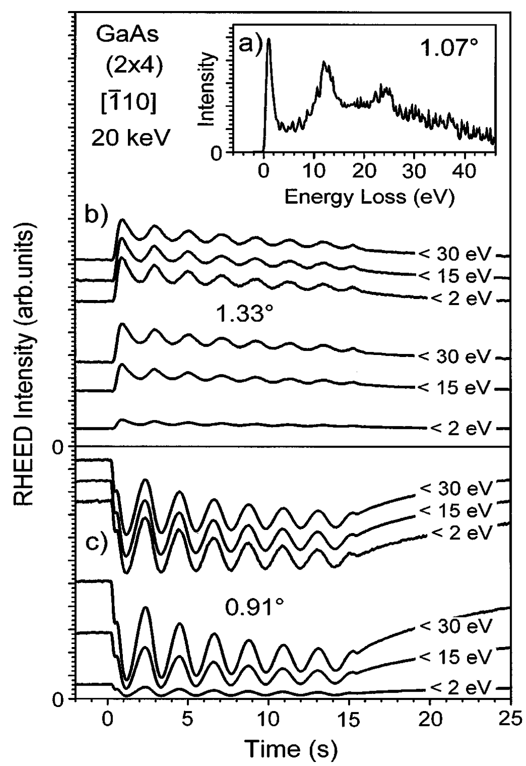

FIG. 4. RHEED intensity oscillations for GaAs (001) growth recorded on the specular spot along [$\bar{2}$10] with different inelastic components included. The inset (a) shows a typical energy loss spectrum.

The normalized curves are identical within measurement accuracy in both Figs. 4(b) and 4(c).

This means that the dominant peaks in the energy loss spectrum do not contribute different oscillation phases to the total intensity. The process responsible for RHEED intensity oscillations is therefore independent of diffuse scattering involving plasmon or band-to-band losses. Processes involving larger energy losses such as core shell excitations contribute too little to the total intensity to shift the phase measurably. The remaining processes that could accommodate a diffuse scattering mechanism are band-to-band transitions below 2 eV, phonon scattering, and diffuse scattering without energy loss.

The phase dispersion along high-symmetry azimuths assumes a much more complicated form [4], since it contains contributions from higher-order beams and is more kinematical in character. For very well ordered surfaces, the RHEED pattern can be described by a purely kinematical treatment [25]. RHEED oscillations in this case are very weak and detectable only at the kinematically forbidden reflections, again emphasizing the dynamical nature of the oscillation process [15,26]. For less ordered surface reconstructions, the assumption of kinematical components with the periodicity of the bulk and unshifted Bragg conditions combined with layer interference contributions of different periodicity may explain the observed complexity. The transition between many-beam and one-beam conditions can account for the observed azimuthal dependence of the RHEED oscillation phase [27]. The layer interference model also directly explains the dependence of the GaAs oscillation phase on As_4 overpressure [5], since variation of the As_4 pressure changes the type of surface reconstruction as well as the relative As coverage within one type [22,28].

In conclusion, we propose a basic model that explains the occurrence of RHEED intensity oscillations based on elastic multiple scattering in the reconstructed layer on top of the growing layer. The model agrees remarkably well with measurements in the one-beam condition and describes several experimental phenomena in a unified approach. Among these are the phase dispersion as a function of incidence angle [4], the absence of inelastic components with different phase in the energy loss spectrum of the oscillations, the occurrence of sharp additional maxima [16], large amplitude oscillations with double minima at low angles [17], and the dependence of the GaAs oscillation phase on As_4 pressure [5]. Since for the one-beam condition a link between experiment and theory is now established, the layer interference model can serve as a basis for more complicated descriptions of the many-beam case in the future.

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