Influence of beam coherence on measurements of roughness in film growth

M. G. Lagally and R. Kariotis
Department of Materials Science and Engineering, University of Wisconsin, Madison, Wisconsin 53706

(Received 17 April 1989; accepted for publication 23 June 1989)

Diffracted intensity oscillations in epitaxial growth are discussed in terms of the existing roughness scale on the surface and the coherence length of the radiation used to measure them. It is shown that for systems with unbounded interface width growth, the greater the coherence length, the more rapidly the oscillations damp out. On the other hand, oscillations can occur and be persistent if the coherence length of the beam is much shorter than the characteristic horizontal length scale for roughness, even in the case where unbounded growth is present. This behavior is governed by the magnitude of the coherence length of the beam, relative to the lateral length scale that characterizes the surface structure.

Oscillations in the diffracted intensity during experiments of epitaxial deposition have been shown to provide a measure of the growth mechanism and an indication of how the growth front roughens.[1,2] When the substrate is infinitely large and the diffracting beam has perfect coherence, the evolution of the roughness (the "interface width") is characterized by two length scales, one horizontal to the substrate (determined by the diffusive properties of the adsorbate) and one vertical to the substrate (determined largely by the deposition rate), as described in earlier articles.[2] The interface width can be defined as the mean-square height difference[3] $\langle [h(r_1) - h(r_2)]^2 \rangle$, where $h(r)$ is the column height at position r on the substrate. Epitaxial deposition can be monitored by means of diffracted intensity oscillations.[1] A simple interpretation of the observed results requires that the oscillations damp out as the surface grows increasingly rough. The diffracted intensity at the out-of-phase condition is given in the kinematic approximation by

$$I(s_z) = \int d^2 r_1 d^2 r_2 \langle \exp\{is_z [h(r_1) - h(r_2)]\} \rangle, \qquad (1)$$

where s_z is evaluated at the conditions for maximum destructive interference between amplitudes scattered from adjacent layers.

The interface width is either finite or infinite in the long-time limit depending on whether surface diffusion is more or less rapid, respectively, than the deposition rate. However, the physical structure of the adlayer does not alone determine the qualitative behavior observed in the laboratory. In the following paragraphs we will present arguments, including reference to Monte Carlo simulation and recent experimental data, that attempt to delineate those situations for which intensity oscillations are characteristic of bounded interface growth, and those for which the effect is largely due to experimental limitations, rather than to bounded growth. The major premise of this letter is that the coherence length (hereafter referred to as ξ) of the diffracted beam relative to the lateral length scale of the surface configurations must also be considered when interpreting the results of diffraction experiments. Previous literature[4] has considered the problem of the relative size of the beam with respect to the coherence length. We will not take up this question, but consider only the case of a beam made of a single coherent wave.

First consider a physical system observed with a perfect instrument. For very small deposition rate, there will exist an intermediate temperature range in which the interface width remains finite even for very long times.[2] Below the equilibrium roughening temperature any surface fluctuation must have a finite lifetime, and so if the deposition rate is lower than this decay rate, it should be possible to deposit without creating a rough surface. Thus, in Fig. 1, an adatom must be able to diffuse a distance k in less time than it takes for another adatom to be deposited.

During a diffraction experiment, the physical structure of the deposit is probed with a beam of finite coherence length. In Fig. 1 we show two possible coherence lengths ξ_1 and ξ_2 of the diffracted beam. The larger of the two will be used to describe the situation in which the diffracted beam samples a sufficiently large horizontal length of the substrate so that the full vertical range is adequately represented. The shorter of the two is meant to describe the case where the true physical scale of lateral fluctuations is much larger than the coherence length, so that only a portion of the actual vertical displacement is seen within a single zone of coherence of the radiation used to probe the surface. The first case (ξ_1) is one in which the experiment should see accurately the real character of the interface. If the physical interface is rough, the experiment will observe the intensity oscillations

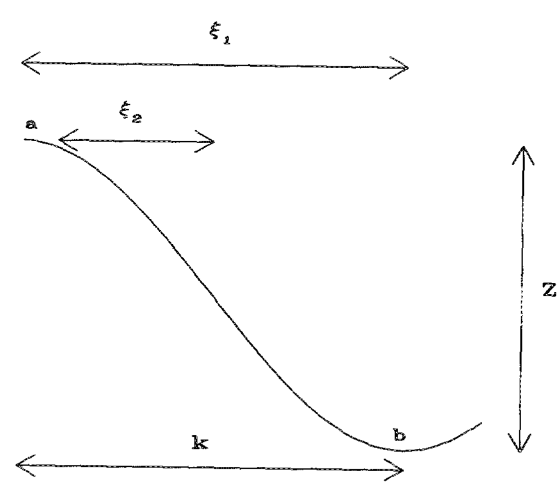

FIG. 1. Typical configuration labeling the scales of a surface fluctuation. The vertical and horizontal length scales are shown relative to two possible diffracted-beam coherence lengths ξ_1 and ξ_2.

to decay monotonically as a function of dose. If the interface is bounded the experiment will observe oscillations that damp in peak intensity to a lower value, at which they become persistent with an amplitude characteristic of the interface width. The second case (ξ_2) is more interesting, and also more common. In this situation the beam does not sample the entire range of vertical values of the surface, and hence it is possible that intensity oscillations persist even though the interface width is growing without bound. The different combinations can be summarized as follows:

(a) Bounded growth, $\xi_{1,2} > k$. This is the ideal case, oscillations will persist, indicative of bounded growth. (In the limit of low deposition rate, the lateral motion of the atoms will be rapid enough to keep vertical fluctuations from growing without bound.)

(b) Bounded growth, $\xi_2 < \xi_1 < k$. Both short and long ξ may yield persistent oscillations, with the shorter one being the least attenuated. Figures 2(a) and 2(b) show Monte Carlo calculations that demonstrate this effect. The smaller ξ produces longer lived oscillations.

(c) Unbounded growth, $\xi_{1,2} > k$. This is the ideal case, oscillations decay, indicative of unbounded growth.

(d) Unbounded growth, $\xi_2 < \xi_1 < k$. The smaller ξ will show longer lived oscillations. In Figs. 2(a) and 2(b), we present Monte Carlo calculations performed at two different temperatures for the lattice gas model above a substrate. The diagrams show the intensity oscillations produced with radiation of two different coherence lengths. The data demonstrate the influence of a probe that samples only a portion of the full lateral scale, that is, the damping of the intensity for the smaller lattice is less. The difference is less pronounced at lower temperatures, for which the lateral scale to achieve the same roughness is shorter. The conclusion of this study is that when the diffracted beam coherence length ξ is shorter than the characteristic lateral length scale of the surface, intensity oscillations may be observed even if the interface width has grown without bound, since the beam is unable to sample the entire range of vertical values described by the interface.

This result is of practical relevance in the analysis of diffracted intensity oscillations. As noted, a number of intensity oscillation measurements have been made, showing eventual complete damping, and are therefore correctly interpreted in terms of an interface width that grows unbounded. However, the rate at which this occurs cannot be inferred from the data unless the coherence of the radiation used is known. The instrument resolving power, i.e., the lateral dimension over which the instrument acts as a correlation detector, provides a lower limit to this coherence. The smaller this distance is, the more slowly the oscillations will damp; in other words a worse instrument may give more persistent oscillations because it looks at less of the roughness. Measurements with a worse instrument will also exhibit greater dephasing if the oscillations persist.

Differences in intensity oscillations measured in different laboratories or using different probe radiation may not be due to different growth conditions, but possibly to differences in the coherence of the radiation. In particular, persistent oscillations must be viewed with caution.[5] For example,

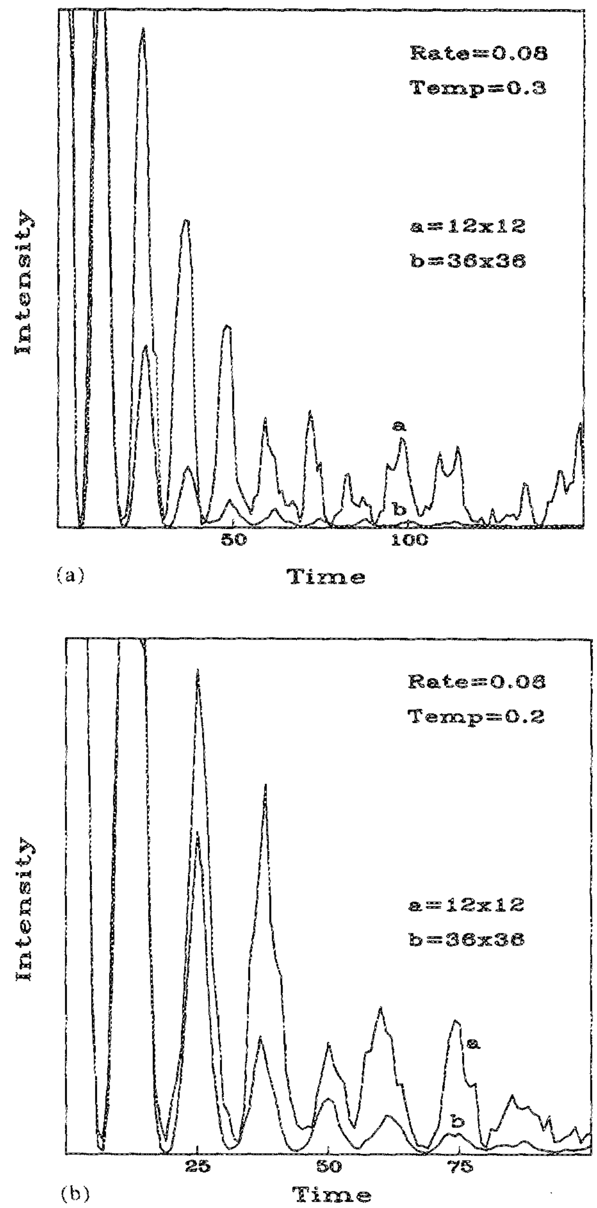

FIG. 2. (a),(b) Monte Carlo runs at different temperatures showing the effect to two different coherence lengths: (a) higher temperature, (b) lower temperature. In each, curve a is for a smaller coherence length and b for one three times as large.

a very highly focused small beam that has a high convergence may have a small coherence and therefore lead to "artificial" persistent oscillations. The effect of different coherence of the probe radiation can be demonstrated, e.g., by comparing reflection high-energy electron diffraction (RHEED) and x-ray diffraction measurements. Evidence is provided by the data for Ge on Ge (111) performed using both x-ray diffraction and RHEED.[6] The x-ray diffraction oscillations damp out more quickly, consistent with the higher coherence of the photons.

A practical application of this effect is that if the coherence length of the instrument is variable it gives one the opportunity to determine not only the vertical roughness scale for the ξ value corresponding to a particular instrument, but also a means of determining the lateral scale of the roughness. In those cases where the instrument resolving power can be externally controlled and for those physical circumstances in which it has already been determined that the interface width is unbounded, a measure of the lateral scale may be obtained by determining the value of ξ for which oscillations finally die out. Measurements to determine this effect are under way.

This work was supported by National Science Foundation Solid State Chemistry Program grant No. DMR-86-15089. We thank J. F. van der Veen for the opportunity to view the Ge RHEED data prior to publication.

[1]J. H. Neave, B. A. Joyce, P. J. Dobson, and N. Norton, Appl. Phys. A **31**, 1 (1983); J. M. van Hove, C. S. Lent, P. R. Pukite, and P. I. Cohen, J. Vac. Sci. Technol. B **3**, 741 (1983).

[2]R. Kariotis and M. G. Lagally, Surf. Sci. **216**, 557 (1989); R. Kariotis, J. Phys. A (in press).

[3]P. Meakin, P. Ramanlal, L. M. Sander, and R. C. Ball, Phys. Rev. A **34**, 5091 (1986).

[4]G. Comsa, Surf. Sci. **81**, 57 (1979); T.-M. Lu and M. G. Lagally, *ibid.* **99**, 695 (1980).

[5]T. Sakamoto, N. J. Kawai, T. Nakagawa, K. Ohta, and T. Kojima, Appl. Phys. Lett. **47**, 617 (1985); T. Sakamoto, T. Kawamura, and G. Hashiguchi, Appl. Phys Lett. **48**, 1612 (1986).

[6]J. F. van der Veen (private communication); E. Vlieg, A. W. Denier van der Gon, J. F. van der Veen, J. E. Macdonald, and C. Norris, Phys. Rev. Lett. **61**, 2241 (1988).