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Are equilibrium fluctuations detectable in diffracted intensities?

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The capability of diffraction intensity fluctuations to probe dynamic phenomena at equilibrium is studied. The summation of fluctuations over different domains that scatter incoherently, the role of statistically ''white-noise'' generated fluctuations and the time necessary to collect a sufficiently long signal are investigated. The factors that determine the number of incoherent regions, and maximize the relative fluctuation without reducing the count rate to such low values that impractically long acquisition times are needed are discussed. Monte Carlo simulations modeling thermally generated step fluctuations are used to illustrate the results. © *1997 American Institute of Physics.* [S0021-8979(97)07615-9]

I. INTRODUCTION

The study of time-dependent phenomena on surfaces is of great interest in understanding the important microscopic processes that control mass transport and growth. In recent years the emphasis has been on studying surfaces under non-equilibrium conditions of practical interest during epitaxy at low temperature and high flux rates.[1] The motivation for such studies is to develop methods to grow epitaxially new materials with atomic scale control. However there is also a need to understand growth at higher temperatures when the system is closer to equilibrium, e.g., Si growth at step-flow conditions (i.e., $T>630\,^{\circ}\mathrm{C}$). In addition equilibrium studies are closely related to theoretical predictions about dynamic phenomena in the statistical mechanics of two-dimensional systems.[2,3]

Equilibrium studies are experimentally more difficult since they involve low amplitude concentration fluctuations generated thermally, which are difficult to measure. The response of the system back to equilibrium can be sufficiently slow depending on temperature to be measured within the acquisition rate of the experiment; however the fluctuations are in general only a few percent of the average intensity level. There are not many experimental methods available to follow the equilibrium response of a system.[4]

Equilibrium methods are usually based on following fluctuations: $\delta n(t)=n(t)-\overline{n(t)}$, where $n(t)$ is the relevant experimental quantity (e.g., concentration) within a probe area A positioned at the center of the surface. If the concentration is measured, $n(t)$ is the instantaneous number of atoms in the probe area and $\overline{n(t)}$ its average value. The relative fluctuations depend on $\delta n(t)/\overline{n(t)}\propto 1/\sqrt{A}$, so it is desirable to use small probe areas if the corresponding measured quantity used in practice amplifies sufficiently the concentration fluctuations. For example, the method has been implemented with the use of field emission microscopy (FEM)[4] or scanning tunneling microscopy (STM),[5] where the measured quantity is the tunnelling current which provides a high amplification factor (10^6) from concentration to tunnelling current fluctuations.

In this article we would like to address the question whether equilibrium fluctuations can be followed with diffraction techniques. If this is possible, one has a far broader range of applicability to practically all experimental systems; at the same time complementary information about the system's morphology and symmetry can be obtained from quantitative studies of the spot profile $S(q)$. By selecting the appropriate wave vector q_0 for the process of interest (i.e., superstructure spots for the formation of periodic overlayers and domains, split spots for step fluctuations on regularly stepped surfaces, wave vector of the edge of the Brillouin zone for island shape oscillations etc.) one can simply follow the intensity fluctuations

$$\delta S_{q_0}(t)=S_{q_0}(t)-\overline{S_{q_0}(t)}, \tag{1}$$

where $\overline{S_{q_0}(t)}$ is the average intensity. As usual, information about the system relaxation is found from the decay of the auto correlation function

$$C(t)=\langle \delta S_{q_0}(t)\,\delta S_{q_0}(0)\rangle=\frac{1}{M}\sum_{i=1}^{M}\delta S_{q_0}(\tau_i+t)\,\delta S_{q_0}(\tau_i), \tag{2}$$

with the configurational average $\langle\rangle$ replaced by the time average over the collected signal, where $\delta S_{q_0}=\sqrt{\langle[\delta S_{q_0}(0)]^2\rangle}$ is the root mean square (rms) value of the fluctuations.

In using diffraction to measure time-dependent fluctuations it is important to evaluate how summation over the different incoherent regions of the surface can degrade the fluctuations. The surface can be decomposed into $N=(L_b/L_c)^2$ regions (where L_b is the beam size and L_c the coherence length) with each region contributing incoherently to the time-dependent fluctuations. One expects that the choice $N=1$ (i.e., $L_b=L_c$) results in the maximum relative fluctuation $\delta S_{q_0}/S_{q_0}$, since it will eliminate the incoherent summation. However, the condition $N=1$ will reduce the average value $\overline{S_{q_0}}$ of the measured quantity and will in turn enhance the statistical fluctuations δc (i.e., $\delta c/\overline{S_{q_0}}\propto 1/\sqrt{\overline{S_{q_0}}}$). The central issue we would like to address is how to decide the optimal choice of N, depending on the particu-

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lar probe used for diffraction (electrons, x ray, etc.), to maximize the relative fluctuations but without increasing statistical fluctuations to such a level that prolonged collection times are necessary.

We consider the scattering process over the beam as the sum of N different stochastic processes (i.e., scattering over region i), each with its own average value $\overline{S^i_{q_0}}$ and fluctuation $\delta S^i_{q_0}(t)$. The total intensity is simply the sum over the average intensities $\overline{S_{q_0}}=\Sigma_{i=1}^{N}\overline{S^i_{q_0}}=N\overline{S^i_{q_0}}$ and under the assumption that each region generates uncorrelated fluctuations the fluctuations corresponding to the total intensity are simply given by $(\delta S_{q_0})^2=\Sigma_{i=1}^{N}(\delta S^i_{q_0})^2=N(\delta S^i_{q_0})^2$ i.e., the rms value of the fluctuations $\delta S_{q_0}=\sqrt{\langle(\delta S_{q_0})^2\rangle}$ grows like $\sqrt{N}$.[6] It is clear that the relative fluctuations for the total system $\delta S_{q_0}/\overline{S_{q_0}}=1/\sqrt{N}$ decreases as the number of incoherent domains increases.

The number of points M necessary to construct a correlation function with specified accuracy depends monotonically on the ratio between the statistical noise δc and the fluctuations δS_{q_0} (which are generated by the physical process under investigation). Since both of them have the same dependence on $\sqrt{N}$ it follows that their ratio is independent of N, i.e., the same sampling time is needed to construct the correlation functions of comparable accuracy. This is not the case if the average intensity $\overline{S_{q_0}}$ is kept constant as N is reduced (and therefore the relative statistical noise is also constant) by increasing the current density in such a way that the number of particles in the incident beam (i.e., electrons, photons, etc.) remains constant. For example with low-energy electron diffraction (LEED) this will require the increase of current density from 10^{-2} to 10^{6} A m^{-2}, when the beam size is reduced from 1 mm to 0.1 μm; otherwise there is an optimal choice of N that compromises the gain of the increase of the relative fluctuation with the disadvantage of reducing the counting rate to extremely low levels. The optimal choice of N can be found if the relation between the acquisition time (which is fully determined by the number of collected points M if measurements are made at the same average intensity level), and the ratio $\delta c/\delta S_{q_0}$ is known.

II. SIMULATION

We will illustrate the previous considerations with a specific example of thermally generated roughening of steps that we have modeled with Monte Carlo simulations. We can access the relative importance of the number of incoherent regions (N), statistical noise (δc), and acquisition data points (M). We have studied the fluctuations on a regularly stepped surface heated to a high temperature from an initial straight step configuration. The details of the simulation can be found elsewhere.[7] Briefly, we start from a stepped surface of size $L_x\times L_y$ ($L_x=512a$, $L_y=256a$, where a is the lattice constant) with 32 steps running in the x direction with periodic boundary conditions in the x direction and screw boundary conditions in the y direction. Initially the steps are straight. The model Hamiltonian involves two interaction energies E_n and E_b that describe the pairwise interaction between neighboring atoms. Hops are performed with the probability $\exp(-E/kT)$, where k and T are Boltzmann's constant and the temperature, respectively, and E is the activation energy for surface diffusion given by

$$E=n_iE_n+(n_i-n_f)E_b, \tag{3}$$

where n_i is the number of nearest neighbors in the initial site and n_f is the number of nearest neighbors in the final site. The surface can attain equilibrium faster, if it is first heated at a slightly higher temperature $E_n/kT=1.5$, $E_b/kT=0.5$, and then cooled down to a temperature $E_n/kT=2.1$, $E_b/kT=0.7$, where equilibrium is indicated by the constant average value of the split spot intensity and the constant terrace length distribution. By using this method we minimize the time required to reach thermodynamic equilibrium especially because of the effective step–step interaction controlling step meandering discussed elsewhere.[7]

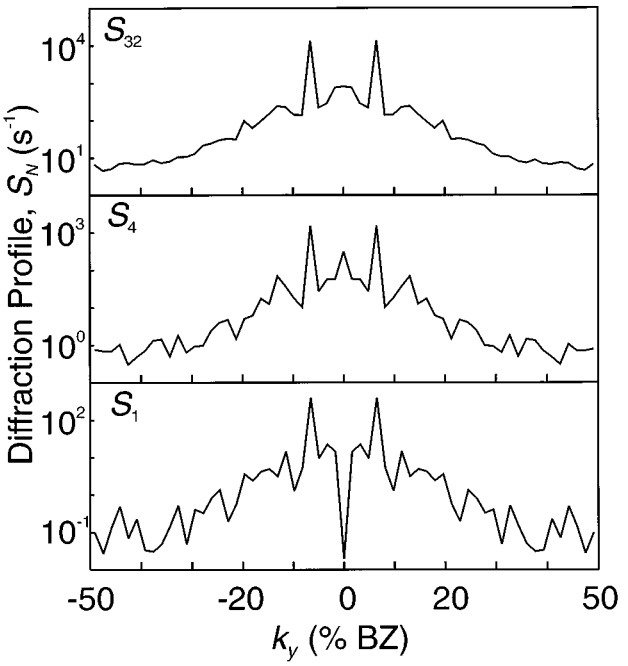

FIG. 1. Split diffraction intensity profiles of a regularly stepped surface annealed at $E_n/kT=2.1$, $E_b/kT=0.7$ of size $L_x=512a$ $L_y=256a$. The surface was decomposed into 32 regions, each consisting of eight terraces separated by eight initially parallel steps running in the x direction. The profiles were summed incoherently from the different surface regions $N=1$, 4, and 32 (bottom to top).

We have modeled incoherent scattering effects by dividing the surface into $N=32$ square regions of size 64×64. In calculating the diffracted intensity we have summed independently each square region and then added the intensity for different regions for $N=1$, 4, and 32. The regularly stepped surface at the out-of-phase condition $k_zd=(2n-1)\pi$ (where k_z is the wave vector normal to surface terraces, d is the step height, and n is an integer) produces the characteristic split spot in intensity versus k_y profiles (where k_y is the wave vector normal to the average step direction) with the inverse of the splitting proportional to the average terrace width.

Figure 1 shows the observed split profiles of the (00) spot for different values of N. Since the profiles were obtained by simple addition of N independent profiles the av-

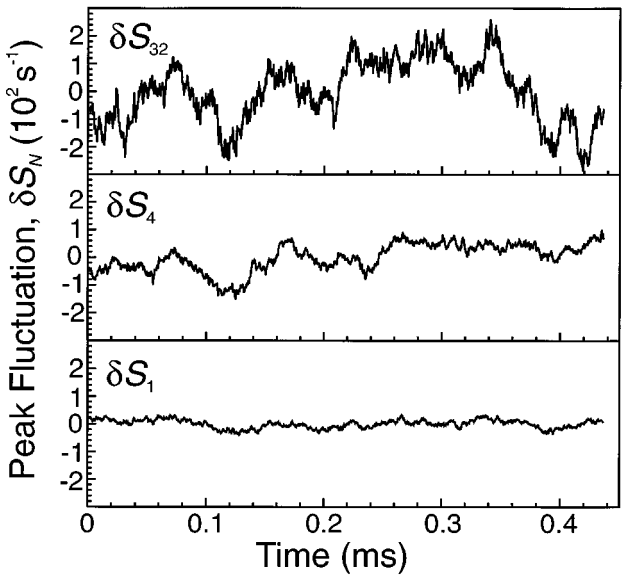

FIG. 2. Absolute level fluctuations incoherently averaged of the split spot intensity vs time for different number of regions. $N=1$, 4, and 32 (bottom to top).

erage intensity increases linearly with N as indicated by the change of scale on the y axis. At this high temperature ($E_n/kT=2.1$, $E_b/kT=0.7$) thermal fluctuations are generated easily and the step meanders around its average position which induces changes in the intensity of the split spots. The magnitude of the intensity fluctuations is monotonically related to the amplitude of the step fluctuations while the density of fluctuations in time (i.e., the power spectrum) is related to how fast the step moves and therefore the microscopic diffusion mechanism producing the fluctuations.

Figure 2 shows the fluctuating part of the signal (after the average level of the split spot intensity is subtracted out) for the different $N=1$, 4, and 32 incoherent region summations. A time scale was introduced in our simulations by choosing the normal frequency factor $v_0=10^{13}$ Hz in the transition probability $v=v_0\exp(-E/kT)$ as described before.[7] Figure 2 shows that the absolute level of the fluctuations increases with increasing N. We have used the rms value of the fluctuations δS_{q_0} as a measure of the effective level of the step meandering. For the chosen annealing temperature the step fluctuates by 20% of the average terrace length, which produces intensity fluctuations of the relative magnitude $\delta S/\overline{S}\sim 0.02$ at the wave vector of the splitting. More pronounced fluctuations should be present if higher temperatures and rougher steps are produced. Figure 3 shows that a linear dependence of δS on $\sqrt{N}$ is a good description of the results. Despite the expected correlation in meandering along the length of the step, it is interesting that the summation of the diffracted intensity shows no correlations, most likely because the domains are sufficiently large so that the atom exchange over the domain boundaries are of minor importance. This in turn will imply that the relative fluctuation $\delta S_{q_0}/\overline{S_{q_0}}$ is reduced as $1/\sqrt{N}$ since $\overline{S_{q_0}}$ increases linearly with N. The corresponding correlation functions for different values of N are shown in Fig. 4 normalized to the initial values. Similar time constants are extracted (defined by the time it takes the intensity to drop to half its value) within

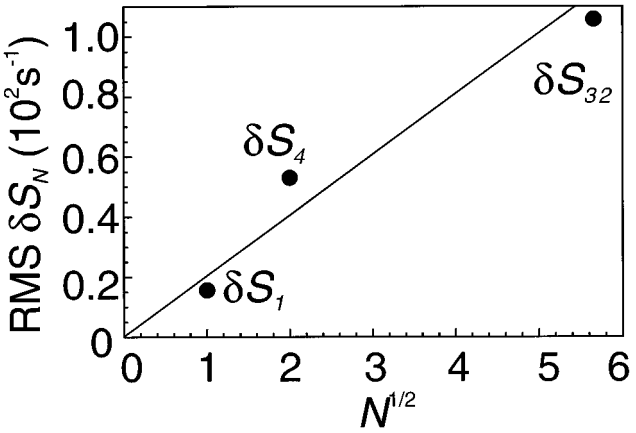

FIG. 3. The effective value $\delta S=\sqrt{\langle[\delta S(0)]^2\rangle}$ of the step fluctuations plotted vs $\sqrt{N}$, the number of incoherent regions. It grows as $\sqrt{N}$ similarly to the statistical noise increase with the number of incoherent domains.

some deviations, because of limited statistics ($M=1740$ was used to calculate the correlation functions).

III. DISCUSSION

The result of our simulations that the absolute fluctuation level grows like $\delta S_{q_0}\propto\sqrt{N}$ is expected to be valid in general for all types of uncorrelated fluctuations generated by other physical processes. In a given experimental measurement where the intensity is determined by measuring the count rate ($I=c/\tau$), as a ratio of the number of counts c over a fixed gate time τ, one should also consider the contribution of statistical fluctuations δc, which is proportional to the square root of the measured number of counts $\sqrt{c}$. The total fluctuation δS_t has contributions both from fluctuations produced by the relevant physical processes δS_p and statistical fluctuations δc; $\delta S_t=\delta S_p+\delta c$. The correlation function for δS_t can be composed into the sum of both correlation functions for the physical process and the statistical noise

$$\langle\delta S_t(0)\,\delta S_t(t)\rangle=\langle\delta S_p(0)\,\delta S_p(t)\rangle+\langle\delta c(0)\,\delta c(t)\rangle + \text{cross terms}. \tag{4}$$

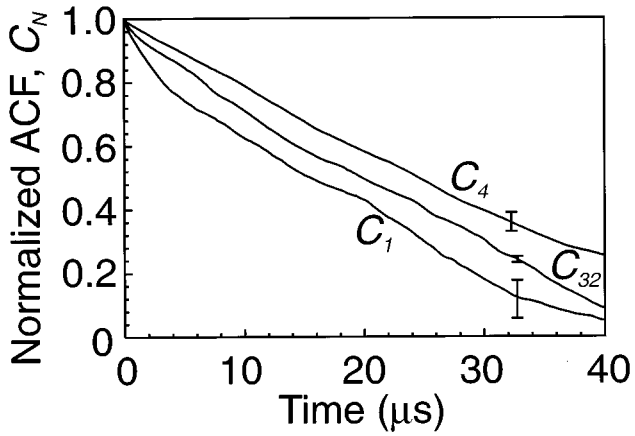

FIG. 4. Normalized correlation functions of the data shown in Fig. 2 for different numbers of incoherent domains N. They have similar shapes with approximately the same time constants (within some uncertainty because of limited statistics). The statistical errors in the data $(\delta S/\overline{S})^2$ are denoted by error bars.

The cross terms average to zero since the fluctuations of the physical process are uncorrelated to statistical fluctuations. In addition the correlation function of statistical noise averages out to zero over the time interval calculated, except for its initial value at $t=0$, i.e., $\delta c=\sqrt{\langle(\delta c(0))^2\rangle}$, i.e., the rms value of the statistical noise (i.e., a delta function). The δ function component of the correlation function can be easily separated out from the experimentally determined correlation functions.[8] However this requires sampling the fluctuations for sufficiently long time [i.e., a large value of M in Eq. (2)] to guarantee that the full ''white noise'' spectrum of statistical noise is measured. As discussed previously, this implies that the acquisition time needed to eliminate the contribution of statistical noise and to produce a correlation function of the same accuracy should be independent of N. In digital mode measurements it is also important to choose the correct gate time τ to sample the fluctuating process. If τ is chosen much larger than the characteristic decay constant of the process, then the fluctuations will be washed out.

These considerations are relevant to recent x-ray scattering experiments to measure the static ''speckle'' pattern that characterizes coherent scattering over a single region $N=1$ and the time-dependent fluctuations within this region.[2] While $N=1$ is a necessary condition to produce the ''speckle'' of an imperfect region, which in turn can be used to determine the exact location of the imperfections, measuring time-dependent fluctuations does not require the same strict condition $N=1$. As our previous discussion has indicated minimizing N (i.e., $N=1$) maximizes the relative fluctuation $\delta S_p/\overline{S_p}$ but at the same time increases by the same factor the statistical noise fluctuation $\delta c/\overline{c}$. In Ref. 2 the condition $N=1$ is obtained by the use of a pinhole in the path of the incident photon beam that reduces the number of scatterers in the diffraction process. The physical process under investigation in Ref. 2 involved the dynamics of critical fluctuations close to a second order phase transition and the expected critical slowing down. These are the optimal conditions to study time-dependent fluctuations since when the system undergoes a second order phase transition critical fluctuations can exceed 100% of the average intensity level. Because of the reduction of N the average fluctuation counting rate was as low as 6×10^{-2} counts/s. This required the use of large gate times ($\tau=100$ s) and only a limited number of data points was possible within the 2 h collection time of $M=60$ to construct the correlation function. In this case the optimal number of incoherent regions could be larger than one. This number is a compromise between maximizing the relative fluctuations but without reducing the average count rate and the number of data points M to an impractically low level.

The study of critical fluctuations was recently carried out with low energy electron microscopy (LEEM)[3] by analyzing fluctuations in real space images for the Si(113)-(3×1) surface deconstruction. As the transition temperature is approached and long range order is lost, critical fluctuations are observed and the disordered phase is formed separated by a high density of domain walls. The measured intensity over a square region (with linear dimension four times the instrument resolution) was used to monitor domain wall (i.e., energy) fluctuations and to deduce the specific heat critical exponent α. It is not clear to what extent the low spatial cut-off imposed by the resolution (i.e., no critical fluctuations over a length scale smaller than the resolution can be observed) implies a course grain average over critical fluctuations in time as well. It limits the range of measuring fluctuations to scales larger than the resolution, as can be seen from the restricted range of the correlation length (by a factor of 2). Possibly, it also filters out the fast frequency components of the dynamic relaxation of the system. It is possible to use LEEM in the diffraction mode, where the intensity versus time is recorded at fixed wave vectors, as suggested by our study with the clear benefit of reducing the number of incoherent regions to $N=(L_0/L_c)^2=10^4$ and only a 10^{-2} reduction in the relative level of fluctuations. This experiment allows a selection of any wave vector of interest, so other critical exponents corresponding to the order parameter can be measured.

IV. CONCLUSION

We have studied with the use of Monte Carlo simulations equilibrium time-dependent fluctuations that can be followed with diffraction. The major question is how diffraction over incoherent scattering regions (presently unavoidable because the beam size includes a large number of coherent regions) reduces the fluctuations. We have shown that the fluctuations originating from a physical process and the ones produced by statistical noise increase with $\sqrt{N}$, if the current density remains constant in the incident beam. This implies that the number of points M that can be collected to construct the correlation function of a given accuracy are independent of N. This is an important consideration in using different diffraction probes (i.e., electrons, x ray, He scattering); N should be determined as a compromise between maximizing the relative fluctuation at the minimal cost in acquisition time necessary to calculate the correlation function.

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[1] G. Rosenfeldt, B. Poelsema, and G. Comsa, in *The Chemical Physics of Solid Surfaces*, edited by D. A. King and D. P. Woodruff (Elsevier, New York, 1996), Vol. 8, Ch. 3.
[2] S. Brauer, G. B. Stephenson, M. Sutton, R. Brüning, E. Dufresne, S. G. J. Mochrie, G. Grübel, J. Als-Nielsen, and D. L. Abernathy, Phys. Rev. Lett. **74**, 2010 (1995).
[3] R. M. Tromp, W. Theis, and N. C. Bartelt, Phys. Rev. Lett. **77**, 2522 (1996).
[4] R. Gomer, Rep. Prog. Phys. **53**, 917 (1990).
[5] M. L. Lozano and M. C. Tringides, Europhys. Lett. **30**, 537 (1995).
[6] N. G. Van Kampen, *Stochastic Processes in Physics and Chemistry* (North Holland, Amsterdam, 1981).
[7] M. I. Larsson, H. Frischat, J. Wollschläger, and M. C. Tringides, Surf. Sci. (in press).
[8] K. Budde, M. I. Larsson, H. Pfnür, M. C. Tringides, and M. Henzler (unpublished).