Quantitative approach to temporal diffraction from stepped surfaces

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We demonstrate how quantitative information on surface kinetics can be obtained from the intensity fluctuations in a diffraction experiment. As an example we have calculated the diffraction intensity time autocorrelation function from a surface consisting of fluctuating steps. Two different rate limiting atomic processes are considered as the source of the fluctuations. We show that the mechanism for equilibrium of a step can be distinguished by the characteristic time decay of the diffraction intensity autocorrelation function. In addition we discuss limits on experimental parameters necessary to observe the diffraction fluctuations.

I. INTRODUCTION

Measurements of dynamic properties have recently been demonstrated using either x-ray photon correlation spectroscopy[1,2] (XPCS) or temporal low-energy electron diffraction (LEED) spectroscopy (TLS).[3] The dynamics of opaque materials can be measured using XPCS, while TLS offers the ability to measure the dynamics of surfaces down to atomic length scales. With the experimental realization that time correlations can be resolved in a diffraction probe, it is necessary to connect the observed time structure with the physical surface phenomena that generate them. As a specific example, we will discuss how the dynamics of steps can be measured using TLS.

Step dynamics and their relationship to crystal growth[4,5] and equilibration of crystal structures are an important area of research.[6] A large volume of experimental work, using both scanning tunneling microscopy and reflection electron microscopy (REM), exists for both stepped metal surfaces[7–9] and stepped Si surfaces, most notably on Si(111) (Ref. 10) and Si(001) (Ref. 11). A number of models for step dynamics have been proposed and studied.[12–14] Bartelt *et al.* have used a model for isolated steps to show that step fluctuations on Si(111) seen in REM images are rate limited by atom evaporation/condensation kinetics.[15] In that work dynamic variables like the step correlation function, $\langle x(t)x(0)\rangle$ (where x is the position of a step edge), were calculated and compared with REM measurements to infer the step kinetics. In temporal diffraction experiments, however, different dynamic variables are measured. Specifically, while the step correlation function $\langle x(t)x(0)\rangle$ can be measured in a real-space probe, a diffraction probe measures correlations proportional to $\langle\cos[x(t)]\cos[x(0)]\rangle$ (see below). Therefore, a comparison of temporal diffraction fluctuation measurements to pertinent surface dynamics variables requires some development.

In a TLS measurement (or any other time resolved diffraction experiment) a time series of the intensity is acquired, i.e., $I(\vec{q},t)$. From this series the autocorrelation function, $G(\tau)$ can be generated:

$$G(\tau)=\langle \delta I(\vec{q},t)\,\delta I(\vec{q},t+\tau)\rangle_t, \tag{1}$$

where $\delta I(\vec{q},t)=I(\vec{q},t)-\langle I(\vec{q},t)\rangle$. Note that the intensities are integrated over the resolution width, Δq, of the diffractionmeter. If Δq is large, the coherent diffraction intensity only comes from a small coherent region of size $\zeta=2\pi/\Delta q$. Since the actual diameter, D, of the incident beam is much larger than ζ, the diffraction intensity is an incoherent sum over the M incoherent regions in the beam given by $M=(D/\zeta)^2$. In the best high-resolution LEED diffractionmeter $(D/\zeta)^2\sim 10^5$. The question is: to what extent does the averaging over these incoherent regions degrade the measurement of $G(\tau)$? To answer this question we must compare the contributions to $G(\tau)$ from both the physical signal we are interested in measuring, $G_{\rm p}(\tau)$, and from statistical noise in the measurement.

The measured autocorrelation function contains contributions from both statistical noise and from the correlated motion of the structural property measured by the diffraction (e.g., step motion, region size, adsorbate density, etc.). As mentioned, the incident electron beam has a finite correlation length, and we must concern ourselves with the number of incoherent scattering regions contained within the area illuminated by the beam. The total collected current is given by an incoherent sum over the M regions

$$\delta I(t)=\sum_{i=1}^{M}\{\langle I_{\rm p}\rangle-[I_{{\rm p},i}(t)+\eta_i(t)]\}, \tag{2}$$

where $I_{{\rm p},i}(t)$ is the diffraction intensity from the ith incoherent region and $\langle I_{\rm p}\rangle$ is the average signal from each region (i.e., $\langle I\rangle=M\langle I_{\rm p}\rangle$). $\eta(t)$ is a noise source presumed to be purely statistical so that its autocorrelation function is $\langle \eta_i(t)\,\eta_j(t+\tau)\rangle=\langle I_{\rm p}\rangle\delta_{i,j}\delta(\tau)$. Combining Eqs. (1) and (2) and using the statistical properties of $\eta(t)$ gives[3]

$$\frac{G(\tau)}{\langle I\rangle}=\delta(\tau)+\frac{\langle I\rangle}{M}\frac{1}{\langle I_{\rm p}\rangle^2}\{\langle \delta I_{{\rm p},i}(t)\,\delta I_{{\rm p},i}(t+\tau)\rangle + (M-1)\langle \delta I_{{\rm p},i}(t)\,\delta I_{{\rm p},j\neq i}(t+\tau)\rangle\}. \tag{3}$$

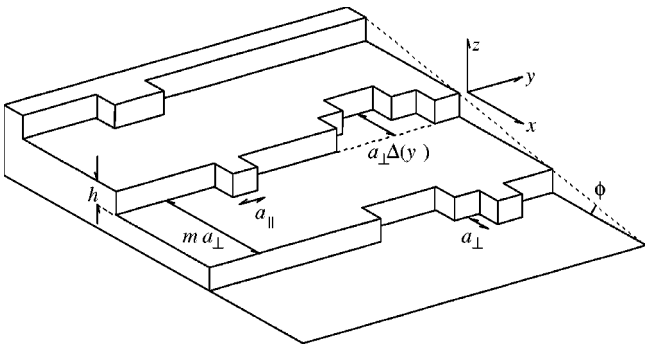

FIG. 1. Schematic drawing of a stepped surface with average terrace length $ma_\perp$.

The first term in Eq. (3) is the statistical noise contribution to the signal. If there are no correlations between the M regions, the last term in brackets is zero. The second term in Eq. (3) is the autocorrelation function from a single region, defined as $G_p(\tau)=\langle\langle \delta I_{p,i}(t)\,\delta I_{p,i}(t+\tau)\rangle_t\rangle_i$. Since M is proportional to the area of the incident beam, $\langle I\rangle/M$ is proportional to the incident current density. Therefore, as long as the incident current density is large enough so that the second term in Eq. (3) is comparable to 1, $G_p(\tau)$ should be measurable compared to the statistical noise.[3]

As a prototypical system we will explore the intensity fluctuations from a surface with atomic steps that are fluctuating in time due to a number of possible atomic processes. To illustrate how kinetics information can be derived from a measured TLS autocorrelation function we seek an expression for $G_p(\tau)/\langle I_p\rangle^2$ for step motion. Once $G_p(\tau)$ is known, we can use it along with surface energetics and kinetics parameters derived from pervious work to place limits on the sensitivity of a diffraction fluctuation measurement.

II. TEMPORAL DIFFRACTION FROM STEPS

We begin by considering a model system of steps. Figure 1 shows a vicinal surface consisting of an ordered staircase structure. At $T=0$ K the equilibrium surface consists of straight mono-atomic steps of height h and a terrace width $L=ma_\perp$, where $a_\perp$ is the distance between atomic rows on the terrace and m is an integer that selects the angle ϕ between the average stepped surface normal and the low-index terrace normal. At elevated temperatures kinks form on the step edges with unit length $a_\parallel$ causing the steps to meander with an amplitude that depends on the kink energy ε and the step-step interaction.[16]

The instantaneous displacement of the jth step at a position y along the step relative to the $T=0$ K position is given by $a_\perp\Delta_j(y,t)$. Step meandering is limited by interactions $V(\Delta,m)$ between adjacent steps [here $V(\Delta,m)$ is the potential per unit length of the step]. We will ignore the case where the only interaction is to exclude overlapping of neighboring steps. While this model has been discussed in the literature,[17] it leads to the unphysical result that the intensity fluctuations decrease with increasing temperature.[18] The form of the potential for small fluctuations (i.e., $\Delta \ll m$) is given by

$$V(\Delta,m)=U(m+\Delta)+U(m-\Delta)\approx 2U(m)+c(m)a_\perp^2\Delta^2. \tag{4}$$

In this harmonic approximation, $c(m)=U''(m)$.

The time-dependent kinematic scattering amplitude from the stepped surface in Fig. 1 is given by

$$A(\vec{q},t)=A_0\sum_{\vec{r}} f e^{i\vec{q}\cdot\vec{r}(t)}. \tag{5}$$

A_0 is the incident amplitude, f is the atomic scattering factor and $\vec{q}=\vec{k}_f-\vec{k}_i$ is the momentum transfer vector. The position of each atom can be written conveniently as: $\vec{r}_{j,n}=x_j(y,t)\hat{x}+\vec{\rho}_{j,n}(y,t)-jh\hat{z}$, where $x_j(y,t)$ is the position of the jth step and $\vec{\rho}_{j,n}(y,t)$ is the position of the nth atom on the jth terrace (relative to the jth step edge).[19] The position of each step edge can further be rewritten in terms of the $T=0$ K position and a displacement, $\Delta_j(y,t)$, as: $x_j(y)=jma_\perp+\Delta_j(y,t)a_\perp$. Similarly $\vec{\rho}_{j,n}(y,t)=n_j(t)a_\perp\hat{x}+\vec{y}$, where $n_j(t)=1,2,3,\ldots,[m+\Delta_{j+1}(y,t)-\Delta_j(y,t)]$. For convenience we break $\vec{q}$ into components perpendicular $q_\perp$ and parallel $q_\parallel$ to the low index terrace surface.

Since the intention is to calculate the intensity fluctuations from a stepped surface, we can choose $\vec{q}$ in such a way as to maximize the sensitivity to step motion. It is easy to show that the diffraction will be maximally sensitive to steps when $q_\parallel$ has only an x component and obeys the equality: $ma_\perp q_\parallel - hq_\perp = n\pi$.[19] For generality we choose $q_\parallel=\pi(1+\beta)/ma_\perp$ which requires that $hq_\perp=[1+\beta-n]\pi$. For $\beta=0$, $q_\perp$ will be at an out-of-phase condition so that adjacent terraces scatter 180° out of phase. The amplitude in Eq. (5) for this value of $\vec{q}$ becomes

$$A(\vec{q},t)=g(\beta,m)\times\sum_j^{N_x}\left\{\sum_{y=-\zeta/2}^{\zeta/2}\cos\left[\pi(1+\beta)\frac{\Delta_j(y,t)}{m}\right]+i\sum_{y=-\zeta/2}^{\zeta/2}\sin\left[\pi(1+\beta)\frac{\Delta_j(y,t)}{m}\right]\right\}, \tag{6a}$$

$$g(\beta,m)=\frac{A_0 f\cos(\pi\beta/2)e^{i\pi(1+\beta)(1+m/2)/2m}}{\sin(\pi(1+\beta)/2m)}, \tag{6b}$$

where the sums are over the coherent size of the beam: $N_x=\zeta/a_\perp$.

In order to calculate the diffraction intensity from Eqs. (6) we must make some assumptions about the relationship between the $\Delta_j(y,t)$'s. To do this we will use a quasi-independent step model. That is, we assume that an isolated step is subject to the potential in Eq. (4) caused by neighboring steps. Using this potential in the Langevin formalism (described in the next section), we calculate the time-dependent fluctuations of a single step. For the purpose of calculating the diffraction intensity from a surface with many steps, we assume that the steps are far enough apart or that the step fluctuations are sufficiently small so that fluctuations on one step do not influence the fluctuations on neighboring steps. In other words fluctuations on neighboring steps do not alter the potential an isolated step is subject to. In this model, the transverse step-step correlation function $\langle[\Delta_j(y)$

$-\Delta_i(y)]^2\rangle$ becomes a constant $2\langle\Delta^2\rangle$; twice the mean squared amplitude of an isolated step.

However, even for relatively large vicinalities, stepped surfaces are generally above their roughening temperature as low as room temperature. Above the roughening temperature $\langle[\Delta_j(y)-\Delta_i(y)]^2\rangle$ has some type of logarithmic divergence in $|i-j|a_\perp$.[20] The quasi-independent approximation used here will therefore fail to reproduce the correct diffraction line shapes even at relatively large $q_\parallel = q_x$. For the purpose of calculating diffraction intensities, however, our assumption of quasi-independent steps leads to the very physical result of a Debye-Waller form of $I(\vec{q},T)$ that depends on the mean squared fluctuation of a step as is shown in Sec. III. Therefore, in the interest of beginning to understand TLS signals, we shall embrace the idea of weakly interacting steps and leave the case of coupled step motion or the case of surfaces above their roughening temperature to be explored at a later date.

For independent steps $A(\vec{q},t)$ [Eq. (6a)] becomes a sum of j independent vectors in the complex plane. In other words, this problem is equivalent to a $2D$ random walker in the real-imaginary plane where each step of the walk has components given by

$$\tilde{x}_j(t) = g\sum_y \cos[q_\parallel a_\perp \Delta_j(y,t)], \tag{7a}$$

$$\tilde{y}_j(t) = g\sum_y \sin[q_\parallel a_\perp \Delta_j(y,t)]. \tag{7b}$$

The total scattered amplitude is then a vector sum of $A(\vec{q},t)$'s from each step after a total of N_x steps. This problem has been thoroughly explored in the literature.[21] The magnitude and phase of $A(\vec{q},t)$ are determined solely by the statistics of $\Delta_j(y,t)$ for a single step.

To calculate the time-dependent correlation function we must take the average $\langle I_p(t) I_p(t+\tau)\rangle$. The average will contain terms like $\langle\tilde{x}_i(t)\tilde{x}_j(t)\tilde{x}_k(t+\tau)\tilde{x}_l(t+\tau)\rangle$ as well as similar terms for $\tilde{y}$. These terms take on different values depending on i, j, k, and l and can be determined exactly within an independent step model. For instance, when $i=j=k=l$ the terms become $\langle\tilde{x}(t)^4\rangle$, for i, j, k, and l not equal to each other the terms become $\langle\tilde{x}(t)\rangle^4$, and so on. By careful counting and only retaining terms proportional to N_x^3 or higher, the time dependent normalized correlation function and diffraction intensity become

$$\frac{G_p(\tau)}{\langle I_p\rangle^2} = \frac{4}{N_x\langle\tilde{x}\rangle^2}\{\langle\tilde{x}(t)\tilde{x}(t+\tau)\rangle - \langle\tilde{x}(t)\rangle^2\}, \tag{8}$$

where

$$\langle I_p\rangle = N_x^2\langle\tilde{x}\rangle^2. \tag{9}$$

Note that the fluctuations are proportional to $1/N_x$, the number of statistically independent steps sampled by the coherent part of the beam. Also note that the averages in Eq. (8) represent an ensemble average over a single step so that the subscript j in Eqs. (7) and on $\Delta_j(y,t)$ can be dropped in subsequent discussions. All that remains in order to evaluate Eqs. (8) and (9) is to calculate the appropriate averages. We outline the calculations for two specific dynamic models describing fluctuations of a step about its equilibrium position.

III. TIME CORRELATIONS

To illustrate how an atomic mechanism gives rise to intensity fluctuations we will look at two separate models for step motion. In the first model diffusion of atoms or vacancies on the terraces is fast enough that the step kinetics are limited by exchange of step atoms with either vacancies or adatoms on the terrace. This is the evaporation/condensation (EC) model. It has been shown that the position of a step in this limit is governed by a Langevin equation[12,15]

$$\frac{\partial\Delta}{\partial t} = \frac{\Gamma_e\tilde{\beta}}{kT}\frac{\partial^2\Delta}{\partial y^2} - \frac{2\Gamma_e c}{kT}\Delta + \frac{1}{a_\perp}\eta(y,t), \tag{10}$$

where Γ_e is the step mobility and $\tilde{\beta}$ is the step edge stiffness given by[12]

$$\Gamma_e = \frac{a_\parallel a_\perp^2}{\tau_e}, \tag{11}$$

$$\tilde{\beta}(T) = \frac{a_\parallel kT}{b^2(T)}. \tag{12}$$

Here, τ_e is roughly the time for an atom to attach/detach from the step edge.[12] The step diffusivity, b^2, is the local mean square length of a kink perpendicular to the step. The attachment/detachment of atoms from the steps is described by an uncorrelated thermal noise term given by

$$\langle\eta(y,t)\,\eta(y',t')\rangle = 2\Gamma_e\delta(t-t')\,\delta(y-y'). \tag{13}$$

In the second model, referred to as the step-edge diffusion model (SD), the atom density on the terraces is assumed to be so low that mass transport is primarily along the step edges. The kinetics are limited by an atom hopping from one step edge site to an adjacent one. The analogous equation to Eq. (10) for the step position is[12]

$$\frac{\partial\Delta}{\partial t} = -\frac{\Gamma_s\tilde{\beta}}{kT}\frac{\partial^4\Delta}{\partial y^4} - \frac{2\Gamma_s c}{kT}\frac{\partial^2\Delta}{\partial y^2} + \frac{1}{a_\perp}\eta(y,t). \tag{14}$$

The noise term is now correlated because of hopping through adjacent sites

$$\langle\eta(y,t)\,\eta(y',t')\rangle = 2\Gamma_s\delta(t-t')\,\delta''(y-y'), \tag{15}$$

$$\Gamma_s(T) = \frac{a_\parallel^3 a_\perp^2}{\tau_s}. \tag{16}$$

Here, τ_s is roughly the time for an atom to hop between two adjacent edge sites.[12] Again we caution that both models fail to accurately represent long wavelength fluctuations of the steps. As mentioned in the last section, stepped surfaces even at moderate temperatures are rough. When many step effects are included, the step edge correlation function $\langle[\Delta_j(y)-\Delta_i(y')]^2\rangle$ diverges as $\ln(|y-y'|)$ whereas the models used here give rise to an exponentially decaying function (see below).[20,22] However, since our intent is to estimate the TLS

signal and identify relevant physical parameters that affect it, these models should be adequate. We also note that equilibration times will be drastically underestimated because larger fluctuations above the roughening temperature will require mass transport at levels much higher than predicted by the models used in this discussion.

Equations (10) and (14) can be solved to give the average intensity and normalized correlation function (see appendix). In the continuum limit ($N_y \to \infty$ and $N_y a_\parallel = \zeta$) the average intensity is found by substituting the appropriate γ_k [Eqs. (A4)] and κ_k [Eqs. (A6)] for either model into Eq. (A17) in order to evaluate $\langle \tilde{x} \rangle^2$ in Eq. (9). This gives

$$\langle I_p \rangle = N_x^2 (\zeta/a_\parallel)^2 |g(m,\beta)|^2 \exp[-q_\parallel^2 \omega^2]. \tag{17}$$

Here $\omega^2(T,m)$ is the mean squared width of a step fluctuation in equilibrium.[23]

$$\omega(T,m) \equiv \left[\frac{b^2 kT}{8c(m)a_\parallel} \right]^{1/4}. \tag{18}$$

Similarly, from Eq. (A18) the normalized autocorrelation function becomes

$$\frac{G_p(\tau)}{\langle I_p \rangle^2} = \frac{4}{N_x(\zeta/a_\parallel)} \sum_{l=-\frac{\zeta}{2a_\parallel}}^{\frac{\zeta}{2a_\parallel}} \{\cosh([q_\parallel \omega]^2 F(\tau,l)) - 1\}. \tag{19}$$

The function $F(\tau,l)$ is the relative correlation of two points on a step separated by $l a_\parallel$

$$F(\tau,l) = e^{-|l|a_\parallel/\xi} - \frac{1}{\pi} \frac{N a_\parallel}{4\omega^2} \int_{-\infty}^{\infty} C_k(\tau) \cos(k l a_\parallel) \mathrm{d}k. \tag{20}$$

We have written $F(\tau,l)$ in terms of the Fourier coefficients of the spatial correlation function $C_k(\tau = |t-t'|) = a_\perp^2 \langle |\tilde{\Delta}_k(\tau) - \tilde{\Delta}_k(0)|^2 \rangle$ defined by Bartelt *et al.*[15] For the EC model C_k is given by[15]

$$C_k(\tau) = \xi \frac{4\omega^2}{N a_\parallel} \frac{1 - \exp[-(\tau/\tau_{0e})\{(\xi k)^2 + 1\}]}{(\xi k)^2 + 1}, \tag{21a}$$

while for the SD model,[15]

$$C_k(\tau) = \xi \frac{4\omega^2}{N a_\parallel} \frac{1 - \exp[-(\tau/\tau_{0s})(a_\parallel/\xi)^2 \{\xi^4 k^4 + \xi^2 k^2\}]}{(\xi k)^2 + 1}, \tag{21b}$$

where

$$\tau_{0e} = \frac{kT}{2\Gamma_e c(m)} \tag{21c}$$

and

$$\tau_{0s} = \frac{a_\parallel^2 kT}{2\Gamma_s c(m)}, \tag{21d}$$

are the equilibrium times of a step disturbance. Also, the correlation length, ξ, for two points on a step separated by y, i.e., $\langle \Delta(0)\Delta(y) \rangle = \omega^2 \exp(-y/\xi)$ is given by[23]

$$\xi(T,m) \equiv \left[\frac{a_\parallel kT}{2b^2 c(m)} \right]^{1/2}. \tag{22}$$

Equation (17) shows that the diffraction intensity decays with a Debye-Waller-type exponent whose argument is $q_\parallel^2 \omega^2$. This should have been expected from the form of Eq. (3). For stepped surfaces, only the step edges contribute to the intensity when $\vec{q}$ is chosen to be at an out-of-phase condition. While the mean step position is constant, the step position fluctuates with a mean squared deviation of ω^2, where ω^2 is viewed analogous to the mean square displacement of an atom, $\langle u(T)^2 \rangle$, in the normal Debye-Waller theory.[24] This means that the ratio $b^2/c(m)$ can in principle be obtained by measuring the peak intensity as a function of temperature. The total intensity at the out-of-phase peak will be $I(\vec{q},T) \propto \exp\{-\langle [\vec{q}\cdot\vec{u}(T)]^2 \rangle - [q_\parallel \omega(T)]^2\}$. Since $\langle u(T)^2 \rangle \propto T$, a plot of $\log(I)$ vs T will be a straight line with a slope proportional to q^2 in the absence of step fluctuations. Deviations of this plot from a linear behavior would be due to the temperature dependence of b^2. In a Terrace-Step-Kink model for instance, $b^2 \propto \frac{1}{2} \sinh^{-2}(\varepsilon/2kT)$.[17] Using Eq. (18) for ω^2, this would contribute a $T^{3/2}$ term in the Debye-Waller plot when $kT \gg \varepsilon/2$. Assuming that $\vec{q}$ can be chosen small enough so that $[\vec{q}\cdot\vec{u}(T)]^2 \ll [q_\parallel \omega(T)]^2$, the deviation from a linear slope should be observable.

The magnitude of the expected TLS signal can be determined from the $\tau = 0$ value of the autocorrelation function, $G_p(0)$ (the mean squared intensity fluctuation from N_x steps). For both models at $\tau = 0$, $F(0,l) = \exp(-|l|a_\parallel/\xi)$ so that $G_p(0)$ contains only equilibrium information about the fluctuations. Assuming that $q_\parallel^2 \omega^2 < 1$, the cosh can be expanded in Eq. (19) for $a_\parallel/\xi \ll 1$ to give

$$\frac{G_p(0)}{\langle I_p \rangle^2} \approx \frac{2}{N_x(\zeta/\xi)} (q_\parallel \omega)^4 [1 - \exp\{-\zeta/\xi\}]. \tag{23}$$

Note that the size of the signal is proportional to $(q_\parallel \omega)^4$ and inversely proportional to a rescaled correlation length (ζ/ξ). The $(\zeta/\xi)^{-1}$ comes from the fact that points on a step separated by distances larger than ξ are independent of one another. Therefore, the number of independent scattering regions within the coherence length ζ are reduced by a factor of $(1/\xi)$. At low temperature where diffusion is slow and $\xi \gg \zeta$, each part of the step is correlated with all others in the coherent part of the beam diameter. In this limit the exponential in Eq. (23) can be expanded so that to first order the fluctuations are independent of both ζ and ξ. At high temperatures the diffusion is fast and ξ is small. If ξ is much smaller than ζ, the fluctuations from Eq. (23) are reduced by the number of statistically independent pieces along the step edge, i.e., (ζ/ξ). Note that Eq. (23) reduces to Eq. (5) of Ref. 3 in the limit $\xi \gg \zeta$.

Given that ζ is known and that $\omega^2(T,m)$ has been measured independently from a Debye-Waller analysis as discussed above, $\xi(T,m)$ can then be determined from the temperature dependence of the fluctuations. By combining the results of Debye-Waller and fluctuations measurements, the form of the step-step interaction potential $c(m)$ can be de-

termined since ω and ξ are related through Eqs. (18) and (22): $(\omega^2\xi)^{-1}=4c(m)(kT)^{-1}$.[23]

We also note that the fluctuations $G_p(0)/\langle I\rangle$ (compared to the statistical noise) depend on the step separation L through N_x, ξ, ω, and $q_\parallel$. Both ξ and ω depend critically on the form of the step interaction potential. For an elastic or dipole interaction[25] (with $U(L)\propto 1/L^2$), $G_p(0)/\langle I\rangle$ increases proportional to L when $\xi\ll\zeta$ but decreases as $1/L$ when $\xi\gg\zeta$. If, as proposed by Alerhand *et al.* for Si(001) steps,[26] the interaction potential has a form $\ln(L)$, then the fluctuation signal always decreases with larger terrace lengths [in this case $G_p(0)/\langle I\rangle\propto 1/L^2$ and $1/L^3$ for $\xi\ll\zeta$ and $\xi\gg\zeta$, respectively].

At this point we can estimate the size of the TLS signal. There has been extensive research on stepped Si(111) and Si(001) surfaces.[16] Alfonso *et al.* have studied the high temperature Si(111) ''1×1'' phase on a range of different step densities using REM.[10] From an analysis of their results values of $b^2=5.7$ Å^2 (at 900°C) and $c(m)$ between 0.72 eV Å$/(ma_\perp)^4$ to 1.32 eV Å$/(ma_\perp)^4$ have been estimated.[23,27] Assuming the larger value of $c(m)$ and a $ma_\perp=200$ Å terrace gives $\omega\sim 69$ Å and $\xi\sim 6400$ Å. If we take the coherence length of the beam to be $\zeta=1400$ Å for high-resolution LEED,[28] then $N_x=7$. The fluctuations from a single region are given by Eq. (23); $G_p(0)/\langle I_p\rangle^2=0.4$. From Eq. (3) the measured diffraction signal, $G(0)/\langle I\rangle^2$, is an average over the M regions plus a contribution from the statistical noise; $1/\langle I\rangle+G_p(0)/M\langle I_p\rangle^2$. For a beam diameter $D=50$ μm the number of coherent regions is $M\sim(D/1400\ \text{Å})^2$; giving $G_p(0)/\langle I\rangle^2\sim 3\times10^{-6}$. In order for $G_p(0)/\langle I\rangle^2$ to be measurable it must be comparable to $1/\langle I\rangle$. If we take the criteria for the minimal detectable signal to be greater than 10% of the statistical noise, we would require $\sim 30\,000$ counts per gate period. Finally, the necessary count rate depend on the gate time chosen for the experiment that in turn depends on a characteristic time scale for the step fluctuations. For a Si(111) stepped surface a typical correlation time, τ_{0e}, for step fluctuations is ~ 1.0 s.[15] Insisting that the time resolution of the TLS experiment be 2% of τ_{0e} (i.e., 0.02 s gate time), the scattered diffraction intensity would have to be $\sim 10^6$ counts/s in order to detect the step fluctuations. This type of count rate is certainly within the capability of a typical high-resolution LEED gun. Note that for standard LEED systems, the beam diameter and ζ are typically 500 μm and 200 Å, respectively.[29] This means that to obtain the same statistics available to a high-resolution LEED system, a count rate of $\sim 10^9$ counts/s would be needed. This is also an obtainable countrate but the 200 Å resolution would mean that G_p would be an average over many q's further reducing the signal. For an x-ray scattering experiment with both a beam diameter and transfer width equal to 5 μm,[2] 3000 counts/s would be required, which is about an order of magnitude higher than most undulator sources provide.

As an illustration of the temperature dependence of the fluctuations we have plotted $G_p(0)/\langle I_p\rangle^2$ in Fig. 2 for a stepped Si(111) surface with terrace sizes of 200 and 400 Å. The step interaction is chosen to be proportional to $1/L^2$ so $c(m)=1.32$ eV Å$/(ma_\perp)^4$ and $\xi\sim 6\,400$ Å at 900 °C. The temperature dependence of ω^2 and ξ are calculated from Eqs. (18) and (22) using either a

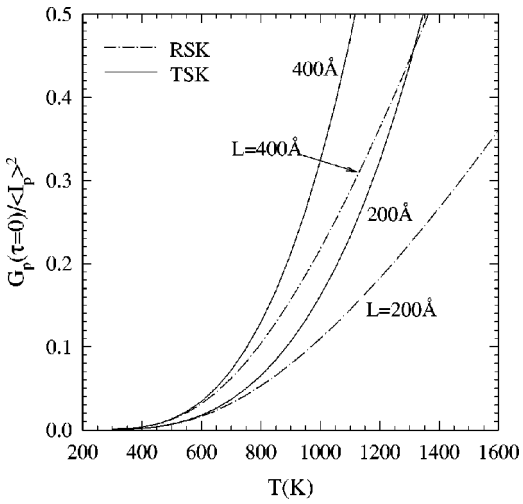

FIG. 2. Plot of the predicted relative correlation function vs temperature for a stepped Si(111) surface. Two terrace widths are assumed: $L=ma_\perp=200$ and 400 Å. The instrument transfer width is 1400 Å. The step diffusivity b^2 is calculated in both the TSK and RSK models.

Terrace-Step-Kink[17,25] (TSK) or Restricted-Step-Kink[30] (RSK) model to estimate the step diffusivity (assuming a kink energy of 200 meV). We point out that $\xi>\zeta$ over the entire temperature range in Fig. 2 so $G_p(0)/\langle I_p\rangle^2\propto L$. From the above discussion it is clear that TLS has sufficient sensitivity to measure the fluctuations of steps.

To illustrate the differences between the time dependence of the two models we have plotted $G_p(\tau)/\langle I_p\rangle^2$ for the EC and SD models in Figs. 3 and 4, respectively, for different values of ζ/ξ. The time behavior of the two models is quite different. The most striking difference, as already pointed out by Bartelt *et al.*,[12] is that equilibration by SD is much slower. Given $\tau_{0e}\sim\tau_{0s}$, the correlation function from the SD model decays over three orders of magnitude slower than $G(\tau)$ in the EC model.

At short times $G_p(\tau)/\langle I_p\rangle^2$ is not a simple function of

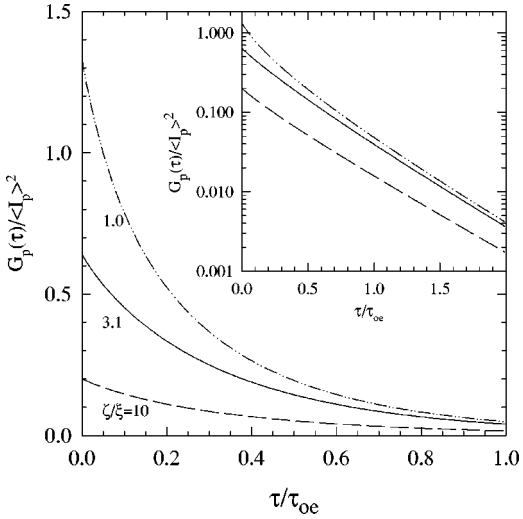

FIG. 3. A plot of $G_p(\tau)/\langle I\rangle^2$ vs τ/τ_0 for the EC model ($N_x=1$ and $q_\parallel\omega=1$). The different curves are for different choices of ζ/ξ. The insert shows a log plot of the same curves showing the long time exponential decay.

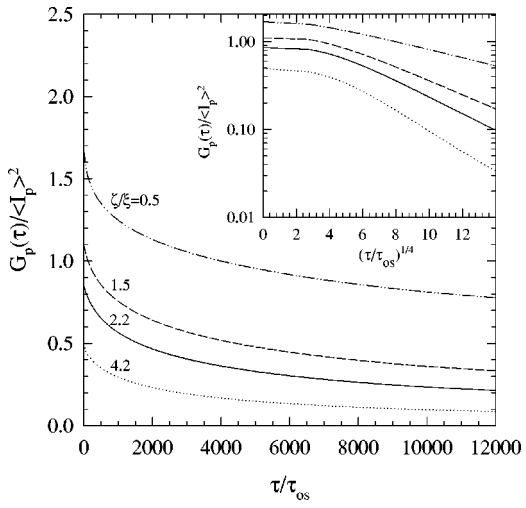

FIG. 4. A plot of $G_p(\tau)/\langle I\rangle^2$ vs τ/τ_0 for the SD model ($N_x=1$ and $q_\parallel\omega=1$). The different curves are for different choices of ζ/ξ. The insert shows a log plot vs $(\tau/\tau_0)^{1/4}$ of the same curves showing the long time exponential decay.

time for either model. The time at which the correlation function falls in half, $(\tau/\tau_0)_{1/2}$, is a function of the transfer width ζ, ξ and $q_\parallel\omega$. Figure 5 is a plot of $(\tau/\tau_0)_{1/2}$ determined from numerical calculation of Eq. (19) for various parameters in the EC model. The half time increases with ζ/ξ for small values of this ratio and then saturates at a constant for $\zeta/\xi>2$. Note that the scaling of the half time has a weak dependence on ξ at large ζ/ξ. This size dependence can be understood by looking at the k dependence of the spatial correlation function in Eq. (21a). The Fourier transform of C_k, when $(\tau/\tau_0)>1$, is dominated by its small k behavior. In this limit $F(\tau,l)$ is approximately a Fourier transform of $(\zeta/\pi)\exp(-\tau/\tau_0)\exp(-\xi^2k^2)$, which is an $\exp(-\tau/\tau_0)$ times a Gaussian who's width is $la_\parallel \sim(\tau/\tau_0)^{1/2}\xi$. In other words, for $la_\parallel=\zeta\gg\xi$, the sum in Eq. (19) can be extended to infinity in the long time limit and

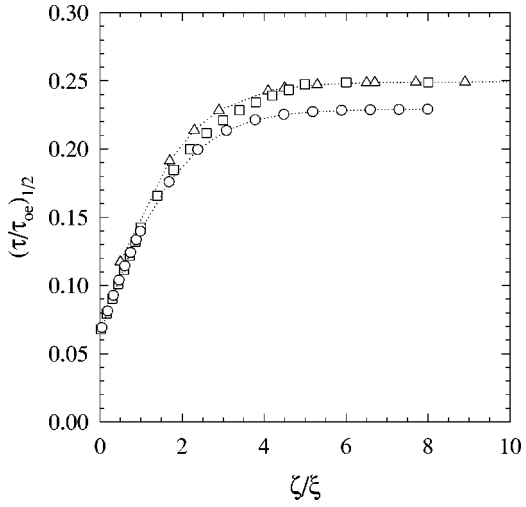

FIG. 5. A plot of the relative half time $(\tau/\tau_0)_{1/2}$ of $G_p(\tau)$ vs the relative beam coherence length ζ/ξ for the EC model ($N_x=1$ and $q_\parallel\omega=1$). The different curves are for different choices of the correlation length; $\xi/a_\parallel=10$ (○), 100 (□), 400 (△). The dotted line is a guide to the eye.

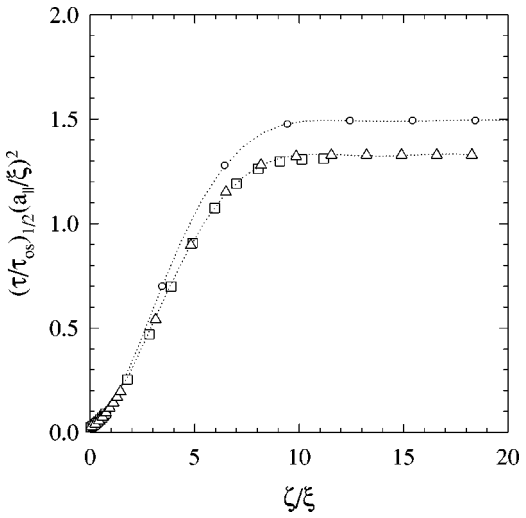

FIG. 6. A plot of the relative half time $(\tau/\tau_0)_{1/2}$ of $G_p(\tau)$ vs the relative beam coherence length ζ/ξ for the SD model ($N_x=1$ and $q_\parallel\omega=1$). The different curves are for different choices of the correlation length; $\xi/a_\parallel=20$ (○), 137 (□), 268 (△). The dotted line is a guide to the eye.

becomes independent of ζ. For $\zeta<\xi$ the sum, and thus the time constant, are a function of ζ. This is equivalent to noting that the longest time constant, $\tau_k=\tau_0[(\xi k)^2+1]^{-1}$, in Eq. (21a) is set by the smallest k probed by the diffraction, i.e., $k_{\min}\sim 2\pi/\zeta$. If $k_{\min}$ is of order $1/\xi$, then τ_k becomes independent of ζ and is completely dominated by the equilibrium time τ_0.

The characteristic decay time in the SD model has a similar behavior (becoming independent of ζ/ξ at large ζ/ξ) as seen in Fig. 6. Note that the time scale in Fig. 6 is normalized by $(a_\parallel/\xi)^2$. Unlike in the EC model, step-edge diffusion requires the concerted motion of atoms from one point on the step to another. This means that a simple $1D$ diffusion model would give the diffusion time to be proportional to $(d/a_\parallel)^2$, where d is some characteristic length. The length scale d is again set by ξ, since it is the distance over which step fluctuations are correlated.

The two models for equilibrium of the step can be easily distinguished in a TLS measurement. To show this we look at the long time behavior of the correlation function, $\tau/\tau_0\gg1$. While we have not been able to develop an analytic expansion of Eq. (19) in this limit, we have deduced an empirical expression from the numerical calculations of Eq. (19) for both models. For the EC model the correlation functions decay exponentially as can be seen in the insert of Fig. 3

$$\frac{G_p(\tau)}{\langle I_p\rangle^2}\underset{\lim\tau/\tau_{0e}\to\infty}{\propto}\exp\left\{-2\frac{\tau}{\tau_{0e}}[1+O(\xi/\zeta)+\cdots]\right\}. \quad (24)$$

Equation (24) is reasonably accurate for $\tau/\tau_{0e}>1$. For step-edge diffusion the intensity correlation function at long times is distinctly different from Eq. (24). From the insert in Fig. 4 the long time behavior is exponential but with an argument proportional to $(\tau/\tau_{0s})^{1/4}$. Empirically we find that the long time behavior can be approximately described as

$$\frac{G_p(\tau)}{\langle I_p\rangle^2} \underset{\lim \tau/\tau_{0s}\to\infty}{\propto} \exp\left\{-\frac{1}{3}\left(\frac{\tau}{\tau_{0s}}\right)^{1/4}[1+O(\xi/\zeta)+\cdots]\right\}. \tag{25}$$

To extract the characteristic atomic time constants τ_e or τ_s from the correlation function requires some apriori knowledge of ξ. For instance, in the case of EC kinetics, an asymptotic fit to a numerical calculation of Eq. (19) gives the slope of a $\ln[G(\tau)/\langle I\rangle^2-1/\langle I\rangle]$ versus τ plot to be approximately $-2[1+(\xi/9\zeta)^{1/2}]/\tau_{0e}$. Neglecting the second term in the slope would amount to a $\sim 50\%$ error in the determination of τ_{0e} if ξ was as large as 2ζ. In principle it would be possible to measure ξ by repeating measurements of $G(\tau)$ for different values of the beam coherence length, ζ, at fixed temperature. This could be done by defocusing the electron beam in order to degrade ζ.

IV. CONCLUSIONS

We have derived an analytic expression for the intensity correlation function in a TLS experiment from a stepped surface for both an evaporation/condensation and step-edge diffusion model. From these results we can determine the necessary count rates to extract the intensity autocorrelation function from statistical noise. We have also demonstrated that the two models for step equilibrium can be distinguished from the characteristic decay of the correlation function a long times.

In principle it may be possible to corroborate the mechanism for step fluctuations by looking for evidence of adatom diffusion on the terraces. In the EC model the step fluctuations must be correlated with terrace adatom fluctuations while in the SD model step fluctuations are only due to step-edge atom diffusion. To see fluctuations due to terrace adatom density changes the TLS measurements would be made at an in-phase diffraction condition but with $q_\parallel$ far from the peak. The signal would be small since the diffuse intensity in the wings would be lower by two orders of magnitude from the peak. Helium atom scattering may be more appropriate for this measurement than electron or x-ray diffraction because of helium's higher scattering cross section for point defects (100 times that of electrons). We are currently developing the theory of diffraction fluctuations from concentration fluctuations in adsorbed monolayers that would be directly applicable to this problem.

In this paper, we have ignored correlations between the motion of adjacent steps. The dynamics of step fluctuations in the strongly coupled limit has already been investigated by others.[31] We can postulate how strong coupling may effect the above results. For similar rate limiting kinetics we may assume that there will be substantial modifications to the correlation function. Because of the coupling between steps, correlated step motion will lead to both breathing and optical type modes for the relative positions of adjacent steps. Depending on the energetics of these two modes there may be more than one characteristic time scale in the correlation function. The mathematical problem of solving a system of coupled Langevin equations and then calculating the diffraction intensity is well beyond the scope of the present work. We leave the calculation of this more complicated problem to others.

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APPENDIX

Rather than solving Eqs. (10) or (14) directly using standard Fourier techniques, it is more convenient to generate a probability distribution for $\Delta(y,t)$ since we are interested in calculating averages of $\cos[q_\parallel a_\perp \Delta(y,t)]$. To do this we place $\Delta(y,t)$ on a discrete spatial lattice $y=na_\parallel$, where $n=1,2,\ldots,N$ (N odd) and write Δ and η as a discrete Fourier series

$$\Delta_n(t)=\sum_{k=0}^{N-1}\tilde{\Delta}_k(t)e^{i(2\pi k/N)n}, \tag{A1}$$

$$\eta_n(t)=\sum_{k=1}^{N-1}\tilde{\eta}_k(t)e^{i(2\pi k/N)n}. \tag{A2}$$

Substituting Eqs. (A1) and (A2) into either Eqs. (10) or (14) gives a linear equation

$$\frac{\partial}{\partial t}\tilde{\Delta}_k(t)=-\gamma_k\tilde{\Delta}_k(t)+\tilde{\eta}_k(t), \tag{A3}$$

where γ_k and $\tilde{\eta}_k(t)$ are appropriate for the EC or SD model. For the EC model γ_k is given by

$$\gamma_k=\frac{2\Gamma_e\tilde{\beta}}{a_\parallel^2 kT}\left\{1-\cos\left(\frac{2\pi k}{N}\right)\right\}+\frac{2\Gamma_e c}{kT}, \tag{A4a}$$

while for the SC model γ_k is given by

$$\gamma_k=\frac{4\Gamma_s\tilde{\beta}}{a_\parallel^4 kT}\left\{1-\cos\left(\frac{2\pi k}{N}\right)\right\}^2+\frac{4\Gamma_s c}{a_\parallel^2 kT}\left\{1-\cos\left(\frac{2\pi k}{N}\right)\right\}. \tag{A4b}$$

The Fourier components, $\tilde{\eta}_k(t)$, are determined from the statistical properties of the noise for each model [Eqs. (13) and (15)] and have the following properties:

$$\langle\tilde{\eta}_k(t)\rangle=0, \quad \langle\tilde{\eta}_k(t)\tilde{\eta}_k^*(t')\rangle=\frac{\kappa_k}{N}\delta_{kk'}\delta(t-t'), \tag{A5}$$

where κ_k in the EC model is given by

$$\kappa_k=\frac{2\Gamma_e}{a_\parallel}, \tag{A6a}$$

while for the SD model

$$\kappa_k=\frac{4\Gamma_s}{a_\parallel^3}\left\{1-\cos\left(\frac{2\pi k}{N}\right)\right\}. \tag{A6b}$$

Note, that the N real valued quantities $\Delta_n(t)$ have been mapped onto an equivalent number of independent Fourier quantities, $\tilde{\Delta}_k$, where $k=0,\ldots,k_{\max}=(N-1)/2$ for N odd. While $\tilde{\Delta}_0$ is real, $\tilde{\Delta}_k$ is in general complex. However, the number of independent k's is reduced by half because of the relationship between the real and complex terms: $\tilde{\Delta}^* = \tilde{\Delta}_{N-k}$.

We are after the joint probability $P_J(\underline{\tilde{\Delta}},t;\underline{\tilde{\Delta}}',t')d\underline{\tilde{\Delta}}d\underline{\tilde{\Delta}}'$ that at a time t the set of displacements $\underline{\tilde{\Delta}} = \{\tilde{\Delta}_0,\tilde{\Delta}_1,\ldots,\tilde{\Delta}_{k_{\max}}\}$ has a value between $\underline{\tilde{\Delta}}$ and $\underline{\tilde{\Delta}}+\mathrm{d}\underline{\tilde{\Delta}}$ and that at t' its value lies between $\underline{\tilde{\Delta}}'$ and $\underline{\tilde{\Delta}}'+d\underline{\tilde{\Delta}}'$. Also let $P(\underline{\tilde{\Delta}},t)d\underline{\tilde{\Delta}}$ be the probability that at time t the set $\underline{\tilde{\Delta}}$ has a value between $\tilde{\Delta}$ and $\underline{\tilde{\Delta}}+d\underline{\tilde{\Delta}}$. If we then define the conditional probability $P_C(\underline{\tilde{\Delta}},t|\underline{\tilde{\Delta}}',t')d\underline{\tilde{\Delta}}d\underline{\tilde{\Delta}}'$ that the value of $\underline{\tilde{\Delta}}$ is between $\underline{\tilde{\Delta}}$ and $\underline{\tilde{\Delta}}+d\underline{\tilde{\Delta}}$ at a time t, given that they were between $\underline{\tilde{\Delta}}'$ and $\underline{\tilde{\Delta}}'+d\underline{\tilde{\Delta}}'$ at an earlier time t', then the joint probability can be written as

$$P_J(\underline{\tilde{\Delta}},t;\underline{\tilde{\Delta}}',t') = P_C(\underline{\tilde{\Delta}},t|\underline{\tilde{\Delta}}',t')P(\underline{\tilde{\Delta}}',t'). \tag{A7}$$

Because of the statistical independence of each Fourier coefficient $\tilde{\Delta}_k$, P can be written as

$$P(\underline{\tilde{\Delta}},t) = \prod_{k=0}^{k_{\max}} P_k(\tilde{\Delta}_k,t). \tag{A8}$$

Since the Langevin Eq. (A3) is linear, the probability density evolves as a Gaussian with its center relaxing according to noise free dynamics with a width $\sigma_k(t)$ (Ref. 21)

$$P_k = \frac{1}{2\pi\sigma_k^2(t)}\exp\left\{-\frac{1}{2\sigma_k^2(t)}|\tilde{\Delta}_k-\tilde{\Delta}_{k\mathrm{o}}e^{-\gamma_k t}|^2\right\} \quad k\neq 0, \tag{A9a}$$

$$P_0 = \frac{1}{\sqrt{2\pi\sigma_0^2(t)}}\exp\left\{-\frac{1}{2\sigma_0^2(t)}|\tilde{\Delta}_0-\tilde{\Delta}_{0\mathrm{o}}e^{-\gamma_0 t}|^2\right\} \quad k=0, \tag{A9b}$$

where $\tilde{\Delta}_{k\mathrm{o}}$ is the initial value at $t=0$ and the time dependent width of the distribution is given by

$$2\sigma_k^2 = \frac{\kappa_k/N}{2\gamma_k}(1-\exp[-2\gamma_k t]) \quad k\neq 0, \tag{A10a}$$

$$2\sigma_0^2 = \frac{\kappa_0/N}{\gamma_0}(1-\exp[-2\gamma_0 t]) \quad k=0. \tag{A10b}$$

The width grows from zero at $t=0$ to its steady state value at $t=\infty$. In the long time limit the probabilities given by Eqs. (A9) approach their stationary values

$$P_k^{\mathrm{st}} = \frac{1}{2\pi\sigma_k^2(\infty)}\exp\left\{-\frac{1}{2\sigma_k^2(\infty)}|\tilde{\Delta}_k|^2\right\} \quad k\neq 0, \tag{A11a}$$

$$P_0^{\mathrm{st}} = \frac{1}{\sqrt{2\pi\sigma_0^2(\infty)}}\exp\left\{-\frac{1}{2\sigma_0^2(\infty)}|\tilde{\Delta}_0|^2\right\} \quad k=0. \tag{A11b}$$

In effect, therefore, the probabilities in Eqs. (A9) are themselves conditional probabilities since they incorporate (as a condition) the initial step distribution. With this in mind we can identify Eqs. (A9) as

$$P_{C,k}(\tilde{\Delta}_k,t|\tilde{\Delta}_k',t'=|t-\tau|) = \frac{1}{2\pi\sigma_k^2(\tau)}\exp\left\{-\frac{1}{2\sigma_k^2(\tau)}|\tilde{\Delta}_k-\tilde{\Delta}_{k\mathrm{o}}e^{-\gamma_k\tau}|^2\right\} \quad k\neq 0, \tag{A12a}$$

$$P_{C,0}(\tilde{\Delta}_0,t|\tilde{\Delta}_0',t'=|t-\tau|) = \frac{1}{\sqrt{2\pi\sigma_0^2(\tau)}}\exp\left\{-\frac{1}{2\sigma_0^2(\tau)}|\tilde{\Delta}_0-\tilde{\Delta}_{0\mathrm{o}}e^{-\gamma_0\tau}|^2\right\} \quad k=0. \tag{A12b}$$

Assuming that the system was set up at some time long in the past so that it has equilibrated by $t'=0$ and assuming that $t>t'$, the joint probability can be written as

$$P_J(\underline{\tilde{\Delta}},\tau;\underline{\tilde{\Delta}}',0) = \prod_{k=0}^{k_{\max}} P_{J,k}(\tilde{\Delta}_k,\tau;\tilde{\Delta}_k',0), \tag{A13}$$

where

$$P_{J,k}(\underline{\tilde{\Delta}},\tau;\underline{\tilde{\Delta}}',0) = P_{C,k}(\tilde{\Delta}_k,\tau|\tilde{\Delta}_k',0)P_k^{\mathrm{st}}(\tilde{\Delta}_k'). \tag{A14}$$

We are now in a position to calculate the appropriate averages in Eqs. (8) and (9). The average $\langle\tilde{x}(\tau)\tilde{x}(0)\rangle = g^2\Sigma_{nn'}\langle\cos[q_\parallel a_\perp\Delta_n(\tau)]\cos[q_\parallel a_\perp\Delta_{n'}(0)]\rangle$ becomes

$$\langle\tilde{x}(\tau)\tilde{x}(0)\rangle = g^2\sum_{n=-\zeta/2a_\parallel}^{\zeta/2a_\parallel}\sum_{n'=-\zeta/2a_\parallel}^{\zeta/2a_\parallel} \times\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}\cos[q_\parallel a_\perp\Delta_n(\tau)] \times\cos[q_\parallel a_\perp\Delta_{n'}(0)]P_J(\underline{\tilde{\Delta}},\tau;\underline{\tilde{\Delta}}',0) \times\prod_{k=0}^{k_{\max}} d\tilde{\Delta}_k d\tilde{\Delta}_k^* d\,\tilde{\Delta}_k' d\tilde{\Delta}_k'^*. \tag{A15}$$

Equation (A15) can be readily evaluated by writing the Δ_n's in terms of their Fourier coefficients, $\tilde{\Delta}_k$ [defined in Eq. (A1)] and using Eqs. (A9), (A13) and (A14), with the result[32]

$$\langle \tilde{x}(\tau)\tilde{x}(0)\rangle = g^2 N_y \exp\left\{ -\sum_{-k_{\max}}^{k_{\max}} \frac{\kappa_k}{2N\gamma_k} q_\parallel^2 \right\} \times \sum_{l=-(N_y-1)/2}^{(N_y-1)/2} \cosh \left\{ \sum_{-k_{\max}}^{k_{\max}} \frac{\kappa_k}{2N\gamma_k} q_\parallel^2 e^{-\tau\gamma_k} \cos\left(\frac{2\pi k l}{N}\right) \right\}, \tag{A16}$$

where $N_y = \zeta/a_\parallel$ and $l = n - n'$. Note that as $\tau \to \infty$ the step fluctuations become uncorrelated with those at earlier times. In this limit $\langle \tilde{x}(\infty)\tilde{x}(0)\rangle = \langle \tilde{x}(\infty)\rangle\langle \tilde{x}(0)\rangle$. In other words, Eq. (A16) also represents the average $\langle \tilde{x}\rangle^2$

$$\langle \tilde{x}\rangle^2 = g^2 N_y^2 \exp\left\{ -\sum_{-k_{\max}}^{k_{\max}} \frac{\kappa_k}{2N\gamma_k} q_\parallel^2 \right\}. \tag{A17}$$

By substituting Eqs. (A16) and (A17) into Eq. (8) the intensity correlation function from a single coherent region can be written as

$$\frac{G_{\rm p}(\tau)}{\langle I_{\rm p}\rangle^2} = \frac{4}{N_x N_y}\left[-N_y + \sum_{l=-(N_y-1)/2}^{(N_y-1)/2} \cosh \left\{ \sum_{-k_{\max}}^{k_{\max}} \frac{\kappa_k}{2N\gamma_k} q_\parallel^2 e^{-\tau\gamma_k} \cos\left(\frac{2\pi k l}{N}\right) \right\} \right]. \tag{A18}$$

[1] 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).

[2] S.D. Dierker, R. Pindak, R.M. Fleming, I.K. Robinson, and L. Berman, Phys. Rev. Lett. **75**, 449 (1995).

[3] E.H. Conrad, A. Menzel, S. Kiriukhin, and M.C. Tringides, Phys. Rev. Lett. **81**, 3175 (1998).

[4] G.S. Bales and A. Zangwill, Phys. Rev. B **41**, 5500 (1990).

[5] J. Villain, J. Phys. (France) **1**, 19 (1991).

[6] A. Rettori and J. Villain, J. Phys. (France) **49**, 257 (1988).

[7] L. Kuipers, M.S. Hoogeman, J.W.M. Frenken, and H. van Beijeren, Phys. Rev. B **52**, 11 387 (1995).

[8] M. Poensgen, J.F. Wolf, J. Frohn, M. Giesen, and H. Ibach, Surf. Sci. **274**, 430 (1992).

[9] L. Barbier, L. Masson, J. Cousty, and B. Salanon, Surf. Sci. **354**, 197 (1996).

[10] C. Alfonso, J.M. Bermond, J.C. Heyraud, and J.J. Métois, Surf. Sci. **262**, 371 (1992).

[11] B.S. Swartzemtruber and M. Schacht, Surf. Sci. **322**, 83 (1995).

[12] N.C. Bartelt, J.L. Goldberg, T.L. Einstein, and E.D. Williams, Surf. Sci. **273**, 252 (1992).

[13] F. Liu and H. Metiu, Phys. Rev. E **49**, 2601 (1994).

[14] B. Blagojević and P.M. Duxbury, in *Dynamics of Crystal Surfaces and Interfaces*, edited by P.M. Duxbury and T. Pence (Plenum, New York, 1997), p. 1.

[15] N.C. Bartelt, J.L. Goldberg, T.L. Einstein, E.D. Williams, J.C. Heyraud, and J.J. Métois, Phys. Rev. B **48**, 15 453 (1993).

[16] E.D. Williams and N.C. Bartelt, Science **251**, 393 (1991).

[17] N.C. Bartelt, T.L. Einstein, and E.D. Williams, Surf. Sci. **276**, 308 (1992).

[18] This result can be derived by using the probability distribution developed in Ref. 23 for noninteracting steps to calculate Eqs. (8) and (9) when $\tau = 0$. See Ref. 32.

[19] E.H. Conrad, in *Physical Structure*, edited by W.N. Unertl, Handbook of Surface Physics Vol. 1 (Elsevier, Amsterdam, 1996), p. 271.

[20] J. Villain, D.R. Grempel, and J. Lapujoulade, J. Phys. F **15**, 809 (1985).

[21] C.W. Gardiner, *Handbook of Stochastic Methods for Physics, Chemistry and the Natural Sciences* (Springer-Verlag, Berlin, 1983).

[22] N.C. Bartelt, T.L. Einstein, and E.D. Williams, Surf. Sci. **244**, 149 (1991).

[23] N.C. Bartelt, T.L. Einstein, and E.D. Williams, Surf. Sci. Lett. **240**, L591 (1991).

[24] R.W. James, *The Optical Principles of the Diffraction of X-rays* (Bell, London, 1962)

[25] C. Jayaprakash, C. Rottman, and W.F. Saam, Phys. Rev. B **30**, 6549 (1984).

[26] O.L. Alerhand, A.N. Berker, J.D. Joannopoulos, D. Vanderbilt, R.J. Hamers, and J.E. Demuth, Phys. Rev. Lett. **64**, 2406 (1990).

[27] E.D. Williams, R.J. Phaneuf, J. Wei, N.C. Bartelt, and T.L. Einstein, Surf. Sci. **294**, 219 (1993).

[28] Y. Cao and E.H. Conrad, Rev. Sci. Instrum. **60**, 2642 (1989).

[29] M.B. Webb and M.G. Lagally, *Solid State Physics* (Academic Press, New York, 1973), Vol. 28, p. 301.

[30] H. van Beijeren, Phys. Rev. Lett. **28**, 993 (1977).

[31] S.V. Khare and T.L. Einstein, Phys. Rev. B **57**, 4782 (1998).

[32] A. Menzel, Masters thesis, The Georgia Institute of Technology, 2000.