Experimental Persistence Probability for Fluctuating Steps

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The persistence behavior for fluctuating steps on the Si(111)-$(\sqrt{3} \times \sqrt{3})R30°$-Al surface was determined by analyzing time-dependent STM images for temperatures between 770 and 970 K. Using the standard persistence definition, the measured persistence probability displays power-law decay with an exponent of $\theta = 0.77 \pm 0.03$. This is consistent with the value of $\theta = 3/4$ predicted for attachment-detachment limited step kinetics. If the persistence analysis is carried out in terms of return to a fixed-reference position, the measured probability decays exponentially. Numerical studies of the Langevin equation used to model step motion corroborate the experimental observations.

With increasing interest in exploiting the special properties of nanostructures, characterization of the general stability or "memory" over long times of a fluctuating structure is likely to be valuable [1]. The idea of persistence that has been developed theoretically in recent years provides one way of addressing these issues [1–3]. The persistence probability $p(t)$ is defined simply as the probability that a random variable never crosses a chosen reference level within the time interval t. It is related to the First-Passage probability [4] and is not determined by a small number of moments of the random variable. The ideas of persistence can be directly related to the stability of crystalline structures by realizing that structural fluctuations occur at steps (e.g., the edges of crystal planes), and that the model classes that have been used to describe these one-dimensional boundaries can be related to the same model classes that have been investigated in theoretical studies of persistence [5].

In the past decade, the unifying concepts of the continuum step model and lattice models of varying complexity have been applied with great success to experimental observations of fluctuating steps on both metallic [6,7] and covalently bonded [8,9] materials. The resulting fundamental understanding has led to quantitative success in kinetic descriptions of experimentally measured structural transformations on the nanometer to micron scale [6,8,10]. However, in addition to the deterministic envelope of kinetic processes, the stochasticity of step fluctuations will become an increasingly important component of structural evolution at the nanoscale. The concepts of persistence are well matched to evaluating the impact of many interesting consequences of such stochasticity, such as the average time required for a structure to *first* fluctuate into an unstable configuration, or for neighboring structures to *first* fluctuate into contact. To develop such applications, it is necessary to demonstrate that the concepts of persistence are indeed applicable to step fluctuations. In this manuscript, we report an experimental demonstration of persistence phenomena on

a solid surface, specifically for the temporal fluctuations of steps with moderately complex structure.

For crystalline structures, persistence can be defined in terms of the fluctuation of a step with respect to a chosen coordinate of interest. Numerous analytical and numerical studies have indicated that the persistence probability has a universal scaling form

$$p(t) \sim t^{-\theta}, \qquad (1)$$

where θ, the persistence exponent, has a nontrivial value characteristic of the dynamics governing the evolution of the variable in question. Systems ranging from the simple diffusion equation [2] to kinetic Ising models [3] have been found to display the scaling form. Despite the number of interesting models studied theoretically, few experiments have been carried out to test the validity and utility of the persistence concept. Experiments on soap froth coarsening [11], liquid crystals [12], droplet condensation [13], and magnetization in spin polarized Xe gas [14] support the proposed universality of the persistence probability and agree with quantitative predictions of the persistence exponent.

We have applied persistence ideas to the case of fluctuating monatomic steps on a metal/semiconductor adsorption system, Si(111)-$(\sqrt{3} \times \sqrt{3})R30°$-Al [15,16]. A fluctuation in the position of the step edge requires correlated motion of several atoms to effect the smallest unit of displacement, that of a single kink. Because atomic processes in this system are more complex than in the lattice models often used in numerical studies, it provides a challenging test of universality.

Experiments were performed in UHV (base pressure $\sim 6 \times 10^{-11}$ Torr) by depositing $0.25-0.33$ ML of Al onto a clean Si(111)-(7×7) sample and imaging the resulting reconstructed surface at temperatures between 770 and 970 K with a variable-temperature STM (Omicron). More extensive experimental details have been published elsewhere [17,18]. Images were obtained by scanning the STM tip repeatedly over a single point on a step edge

for between 23 and 107 s. An example of one of these line-scan images is shown in Fig. 1. The digitized step positions extracted from the images generate a function, $x(t)$, that can be used to study the statistics of fluctuating steps, with the usual time correlation function

$$G(t) = \langle [x(t) - x(0)]^2 \rangle, \qquad (2)$$

as well as the persistence probability.

The time correlation function for equilibrium step fluctuations can be determined through a Langevin formalism, which has achieved great success over the past decade [6,8,18]. In this phenomenological approach, the coarse-grained step position evolves in time via

$$\partial_t x(y, t) = F[x(y, t)] + \eta(y, t), \qquad (3)$$

where F is the appropriate functional describing the relaxation of the step position and η represents random noise due to thermal fluctuations. The y direction runs parallel to the average step edge so that $x(y, t)$ describes the step's perpendicular excursions from its average. For the two most commonly observed step fluctuation mechanisms, random attachment-detachment and diffusion along the step edge, F is proportional to $(\nabla^2)^{z/2}$, where the dynamic exponent z equals either 2 or 4, respectively [6]. The statistical properties of the noise (volume conserving for $z = 4$ and white for $z = 2$) are chosen to satisfy conservation laws and the fluctuation-dissipation theorem. For the two cases mentioned, the correlation function has the simple form,
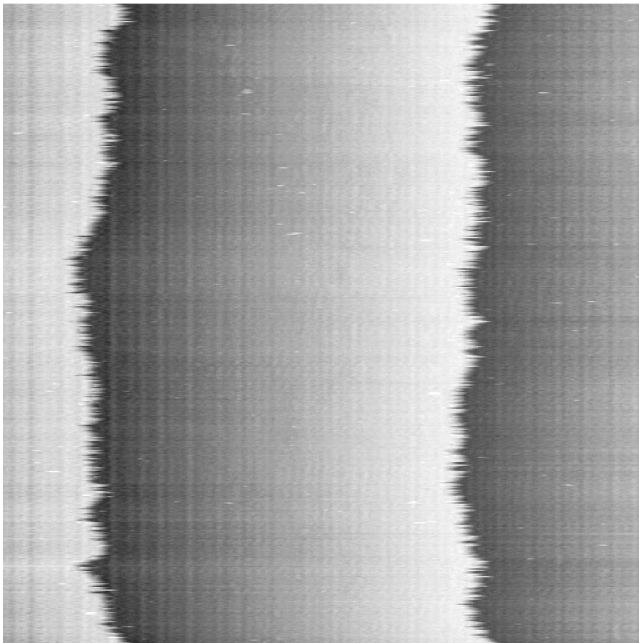

FIG. 1.   Line-scan STM image at 970 K—repeated measurement at a fixed y coordinate with a line-scan length of 100 nm and a line-scan time of 74 ms. Total time for the measurement is 38 s.

$$G(t) = c(T)t^{1/z}, \qquad (4)$$

where $c(T)$ contains the coarse-grained step stiffness and step mobility. A previous analysis of the time correlation function for this experimental system [18] yielded an average exponent of $z = 2.17 \pm 0.09$ from fits to Eq. (4). Supporting evidence suggests that the rate-limiting step fluctuation mechanism is most likely random exchange of mass between the step edges and the intervening terraces, and that the system thus corresponds to the case with F in Eq. (3) proportional to ∇^2 [6,8,18]. Extraction of $c(T)$ from the experimental data yielded time constants for attachment-detachment events that vary from 1.2 ms at 970 K to 260 ms at 770 K [18].

The persistence probability is nontrivially related to the correlation function of the position-time data, and must be determined through a separate analysis. To do this, the experimental persistence probability was calculated by dividing the time axis into intervals of length t and then determining the fraction of those intervals for which the digitized step position never crossed a chosen reference. This was done for a number of images (4–14) for each temperature, and the results were averaged to obtain the final probability.

The first-return question for each small time interval was investigated by choosing the reference as the initial step position in that interval. Figure 2 shows linear behavior on a double log scale, clearly indicating power-law decay. Between 770 and 970 K, the slopes in Fig. 2 yield an average effective persistence exponent of 0.77 ± 0.03. This result agrees with computational predictions of a value of 0.75 for the exponent for attachment-detachment limited step kinetics [5] and for other systems in the same dynamic universality class . Thus, the two exponents, z
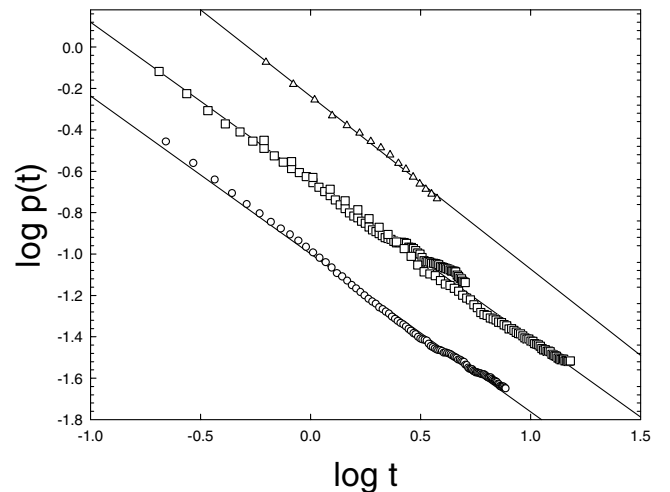

FIG. 2.   Log-log plot of the measured first-return persistence probabilities: 970 K (squares), $T = 870$ K (circles), $T = 770$ K (triangles). The slopes yield values of $\theta = 0.77$ for 970 K, $\theta = 0.76$ for 870 K, and $\theta = 0.84$ for 770 K, for an average (weighted by number of steps) value of $\theta = 0.77 \pm 0.03$.

and θ, computed independently from the same experimental data, lead to a consistent interpretation in terms of the model stochastic equation governing the rate-limiting step fluctuation mechanism.

The choice of the reference level used in the analysis above corresponds to a specific physical question: Given the position x of the step at some time zero, what is the probability that the step will not have returned to that same position within the time interval t? One can imagine a different physical question, relevant for equilibrated steps fluctuating about a known average position x_0. This question is: Given an arbitrary time zero, at which the step position may be any accessible value of x, what is the probability that the step position will not have returned to x_0 within time interval t? The second question corresponds to a persistence analysis with a fixed absorbing boundary. Experimentally, the choice of reference level will be dictated by the specific purpose for which persistence is intended. In particular, first passage into some fixed region of a surface could influence the placement of small structures in nanometer scale electronic devices.

It was possible to analyze the data with a fixed return point because the surface under study was near equilibrium, and the steps had well-defined average positions. Thus, the average position computed from the entire image was used as a reference level. The resulting persistence probability was observed to decay exponentially as shown in Fig. 3. At short times, the data is better fit to a power law with an exponent of ~ 0.4 as shown in the inset. At long times, very few segments of the image had avoided crossing the reference for the entire history of

the evolution and the computed probability dropped precipitously to zero.

To model the experimental system, the ∇^2 linear Langevin equation was solved numerically on small lattices (32 sites) using the discretization taken from Ref. [5]. STM line-scan data was simulated by following one point of the lattice over the full time evolution and performing the calculation of persistence as described above. Figure 4 shows the results averaged over 100 independent numerical solutions for each choice of reference level. The behavior in both cases agrees with the experimental observations. For the return to the initial configuration, a persistence exponent of 0.76 is obtained as expected from Ref. [5]. In addition, the rollover from power-law to exponential decay associated with the average reference level occurs in the numerical integration as in the experimental data.

The clear power-law decay of the first-return persistence probability for the fluctuations of steps on Si(111)-$\sqrt{3} \times \sqrt{3}R30°$-Al demonstrates the feasibility of obtaining useful first-passage statistics from STM data. The fact that the same exponent is observed for three temperatures that correspond to more than 2 orders of magnitude difference in the underlying time constant of the physical system [18] is a significant demonstration of the universal behavior predicted theoretically. Despite the complexity of the atomic-scale processes underlying the step fluctuations, the value of the first-return persistence exponent agrees well with theoretical predictions for a model Langevin equation expected to describe fluctuations governed by random exchange of atoms at the step edge [5]. This class of behavior is consistent with
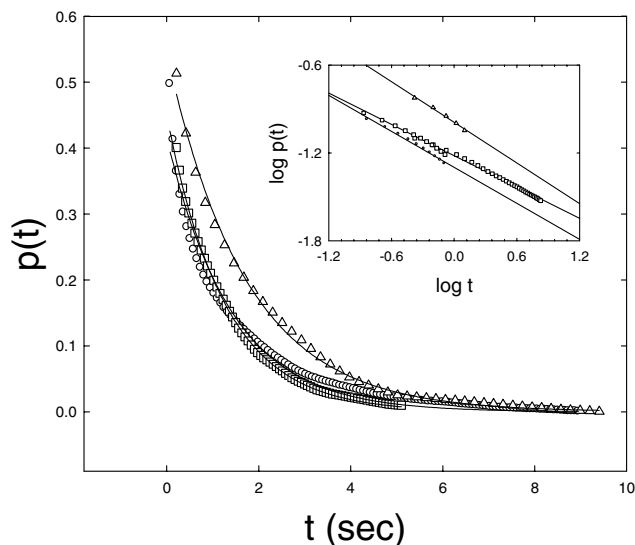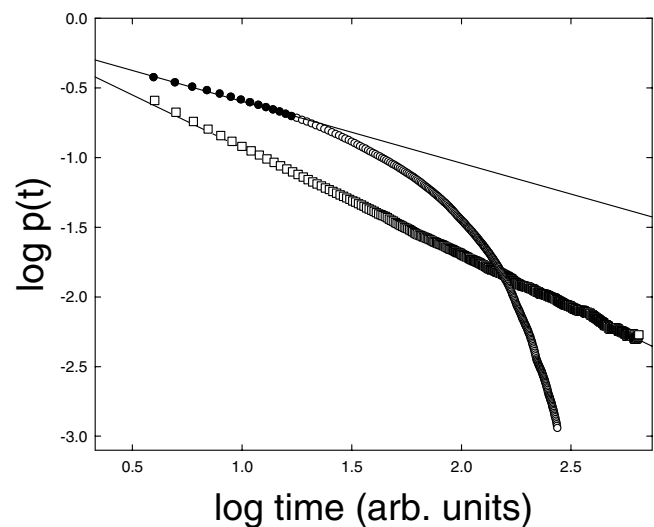

FIG. 3. Measured persistence probabilities with respect to the average step position fluctuations at 970 K (squares), $T = 870$ K (circles), and $T = 770$ K (triangles). The solid lines are fits to an exponential decay. The inset shows the initial power-law scaling of the persistence probabilities which have an average apparent exponent of $\theta = 0.40 \pm 0.03$.

FIG. 4. Persistence probabilities obtained from the numerical solution of the ∇^2 Langevin equation. The slope of the first-return reference level calculation (squares) gives a persistence exponent of 0.76. The average step position calculation (circles) shows early time linear behavior with a slope of 0.44 (solid circles) but at later time decays exponentially.

the previously measured scaling of the time correlation function [18]. Since different physical mechanisms will generally lead to different values of the persistence exponent, it provides corroborating evidence for concluding that the step fluctuations are dominated by attachment-detachment events.

The persistence probability associated with crossing the average step position was both qualitatively and quantitatively different from that of the conventional first-return problem. There have been few theoretical analyses of the dependence of persistence properties on the reference level [19] to guide interpretation of this result. However, it is interesting that the time constant of the exponential decay for the fixed-reference case is found to be independent of large changes in the physical parameters of the system. Rescaling the Langevin calculation to account for the change in the physical parameters of the experimental system with temperature suggests that this observation is physical [20].

As the number of experimental studies of persistence increases, it will be possible to evaluate the potential applications of the concept. At the least, it provides an independent dynamic exponent that can be associated with the physical mechanism underlying the observed kinetics. As noted by Cueille and Sire [21] for more complicated systems than considered in this study, the increased sensitivity of the persistence probability to the details of temporal correlations also allows a more complete understanding of the extent to which the usual dynamic exponents uniquely characterize universality classes. Recent generalizations of persistence [21–23] may bring added insight to the dynamics of complicated stochastic processes. From an applied perspective, given the increased importance of thermal fluctuations with decreasing size scale, experimental and theoretical studies of persistence may prove useful in controlling fabrication, stability, and response of nanostructures.

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