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Low energy electron microscopy investigations of kinetics and energetics on clean close-packed metal surfaces

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Abstract

Experiments are described in which low energy electron microscopy is employed to investigate the kinetics and energetics of clean close-packed metal surfaces, using step fluctuation spectroscopy. The data contain systematic trends of surface mass diffusion and step energetics that are identified and discussed. Further experiments and equipment development have been undertaken to explore the influence of accelerated mixing on surface evolution caused by ion beams, through the resulting kinetic and energetic perturbations of surface processes.

1. Introduction

Nanostructures play an important role in proposed materials that are structured for scientific or for technical purposes. Their synthesis and properties largely depend on fundamental behaviour related to the kinetics and energetics of surface thermal defects on the materials surfaces. Unfortunately, the pertinent behaviour is difficult to predict from theory, and experimental probes by which relevant properties of materials may be determined are scarce. Our focus here is on model nanoscale features that occur on the clean surfaces of vacuum-compatible metals. Kinetics on surfaces are largely determined by the surface mass diffusion coefficient [1–3]. Primitive surface features that contribute to surface energetics include surface steps and their mutual interactions, crystallographic surface facets, and more complex nanostructures [4, 5]. Important information about the energetics and the surface mass diffusion coefficient is contained in the equilibrium configurations of these surface structures, and in the amplitudes and decay rates of their fluctuations [6]. This information can be accessed by various microscopies such as low energy electron microscopy (LEEM) [7], scanning tunnelling microscopy [5] and reflection electron microscopy [8]. This paper describes the systematic application of LEEM to current and proposed investigations of surface kinetics and energetics on clean close-packed metal surfaces.

It requires recognition that surface science is endlessly complex and detailed. For each crystalline material, a variety of different surface orientations may be of interest, each with its particular characteristics. One response to this formidable breadth is to identify specific surfaces of important crystals that invite valuable applications, and to pursue the broad range of property measurements needed for fundamental understanding of behaviour. Silicon (001) and (111) provide examples in point that have attracted a great deal of attention, with experimental determinations of surface reconstructions [9, 10], surface defects [11], surface mass diffusion [7], ‘hut’ nanostructures [12, 13], and so on. A valid alternative is offered by studies focused on classes of similar materials, in the search for systematic behaviour and the comprehension of global trends in properties. Here we present an overview of such research using LEEM to explore systematic features of the behaviour exhibited by close-packed metal surfaces.

In what follows, section 2 reviews the step fluctuation spectroscopy employed in much of this recent research. Section 3 summarizes the results to date of the survey of surface properties entailed in this work. The results exhibit systematic trends that are discussed in section 4, which also outlines a perspective into which the observed behaviour fits. It is believed that a great deal more can be learned when the surface is, in addition, perturbed by an ion beam that accelerates surface mixing processes. Section 5 describes LEEM experiments showing how surfaces described in section 3 evolve in time when perturbed in this way by an ion beam. It also sketches current efforts to install an accelerator onto a LEEM in order to explore surface evolution *in situ*, under the stimulus of a beam of self-ions that perturb the chemically uncontaminated surface.

2. LEEM studies of step fluctuations

We have made experimental studies of step fluctuations on the (111) surfaces of Pt, Pd, Au and Ni, and the (011) surfaces of Mo and Fe; the Ni and Fe are not fully analysed so results are confined to the other four surfaces. All studies employed commercial bulk crystals except Mo(011), which used MBE single-crystal films grown on sapphire. The bulk crystals were cleaned by cycles of Ar⁺ sputtering and annealed, followed by annealing in O₂, as detailed elsewhere [14, 15]. Cycling to 1700 K in UHV served to clean the Mo films [16].

Microscope images of fluctuating step profiles at elevated temperatures may be processed to reveal Fourier amplitudes $y_q(t)$ of harmonic fluctuation, and the time sequence of Fourier amplitude in successive frames then allows the relaxation time τ_q to be calculated from [17, 18]

$$\langle y_q(t)y_q^*(t') \rangle = \langle |y_q(t)|^2 \rangle \exp -(t' - t)/\tau_q. \quad (1)$$

The Gibbs–Thomson [2, 4] effect creates a chemical potential $+\tilde{\beta}A d^2y/dx^2$ on both sides of a step oriented down along $+y$, and diffusion between step locations of opposite curvature then drives relaxation; here $\tilde{\beta}$ is the ‘step stiffness’ and A the area per atom. LEEM is particularly well suited to measurements of step fluctuations at temperatures of interest in growth phenomena. For a step of length L one finds [4] from equipartition

$$\langle |y_q(t)|^2 \rangle = k_B T / L \tilde{\beta} q^2, \quad (2)$$

and this yields step stiffnesses directly from measured Fourier amplitudes. Inset in figure 1 is an example that shows how data follow the q^{-2} dependence predicted for the capillary process [4]. The relaxation on metal surfaces derives from both surface and bulk diffusion, coefficients D_s and D_b , and it has been shown [4, 18, 14] that

$$\tau_q^{-1} = (2A\tilde{\beta}q^3/k_B T)[D_s + (\pi/2qa)D_b] = 2A\tilde{\beta}D_{\text{eff}}q^3/k_B T \quad (3)$$

with D_{eff} an effective surface diffusion coefficient.

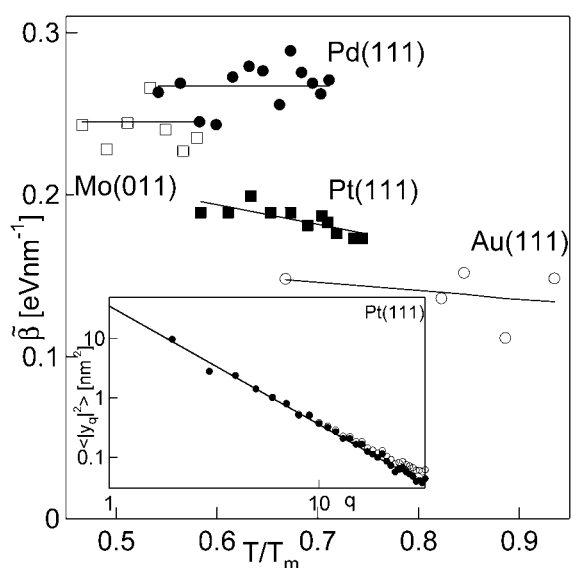


Figure 1. The measured step stiffnesses for particular orientations of steps on Pt(111), Pd(111), Au(111) and Mo(011) have similar magnitudes and depend only weakly on temperature. The inset shows the q^{-2} variation of the squared mode amplitudes; open circles, raw data; full circles, with corrections for spatial and temporal resolution.

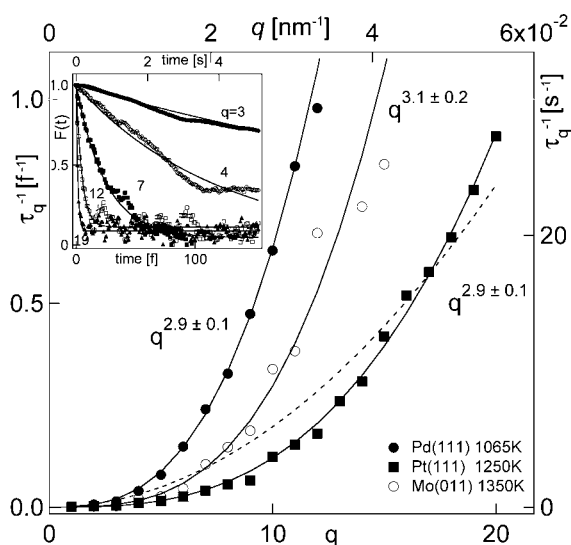


Figure 2. Measured relaxation times for step modes of Pt(111), Pd(111) and Mo(011) at about $T_m/2$, fitted to a power law q^α , with results indicated. $\alpha = 3$ corresponds uniquely to surface diffusion; a broken line contrasts the behaviour for $\alpha = 2$. Inset are data showing observed decays of correlation from which relaxation times are determined. $1s = 30$ frames and q (nm^{-1}) = $2\pi q/L$ with q integer and L typically $2.2 \mu\text{m}$.

Figure 2 shows how the q^3 regime predicted for dominant surface diffusion on metal terraces [18, 19] is identified for several metals at $T \sim T_m/2$; an observed crossover to q^2 at higher temperatures, that signals dominant bulk diffusion [14], is discussed in section 3. Inset in figure 2 are examples of $\langle y_q(t)y_q^*(t') \rangle$ fitted to exponential decays that yield the τ_q .

In our program, measurements have been completed on Mo(011) films [16], and bulk crystals of Pt(111) [14], Pd(111) and Au(111).

3. Energetics and kinetics on close-packed metal surfaces

3.1. Step energetics

Step stiffnesses derived from equation (3) provide step energies β calculated from [4] $\tilde{\beta} = \beta + d^2\beta/d\theta^2$. The angular dependence has been explored [16] by LEEM for Mo where the anisotropy of $\tilde{\beta}$ on the 2 mm surface is a factor 2. However, this yields a much smaller anisotropy of the step energy β . For fcc Pt(111) and Pd(111), with $3m$ symmetry, and at high temperatures, the stiffness itself is much more isotropic.

As exemplified by the results in figure 1 for Mo, Au, Pt and Pd, the stiffnesses near $T_m/2$ of all metals measured to date are within a factor of two of 200 meV nm^{-1} , and at most weakly temperature dependent. Both observations were unexpected. First, it was expected from simplified kink models that the stiffness in this temperature range would weaken towards a surface roughening transition [1] with $\tilde{\beta} \rightarrow 0$; instead, the $\tilde{\beta}$ appear stable. Second, stiffnesses determined at lower temperatures by various means [5] appear to have values larger than those determined in our work, by a substantial factor of three to ten. No explanation of this apparent discrepancy has as yet been identified.

3.2. Kinetics

The relaxation times observed in our work at $T \sim T_m/2$ provide unambiguous and quantitative determinations of the surface mass diffusion coefficient that are otherwise extremely difficult to obtain. The observed q^3 dependence is specific to processes limited by surface diffusion across terraces, a factor q^2 arising from the chemical potential and a further q from the path length, the latter reduced by area factors for bulk flow [14]. Figure 2 shows that relaxation times for three different metals fit a q^3 variation, corresponding to surface diffusion, in the lower temperature ranges of the present studies. The prefactors for surface diffusion determined independently from the temperature dependence in each case are $D_0 \sim 10^{-3}-10^{-4} \text{ cm}^2 \text{ s}^{-1}$, which appears reasonable, as discussed in section 4.

In figure 3, the measured surface diffusion coefficients D_s for the different metals are all displayed as a log function of T_m/T . These results are obtained from observed relaxation times over a wide range of T and q , using equation (3), as detailed elsewhere [14]. It is a striking fact that the several data sets lie quite close to each other in this figure. Together with the similarity of the observed values of D_0 , this establishes that the activation energies for surface mass diffusion scale among the different close-packed surfaces approximately as T_m . A similar behaviour for bulk diffusion has long been recognized, and is established securely by a wide range of experimental results [20], as discussed further in section 4.

4. A perspective on surface systematics

That surface diffusion coefficients (for the few known cases) are remarkably similar when scaled to homologous temperatures by T_m is unexpected; it is, however, similar to bulk diffusion, for which all simple metals, and other simple crystals like rare gases [20, 21], conform to

$$D_b = 0.3 \exp -17T_m/T \text{ cm}^2 \text{ s}^{-1} \quad (4)$$

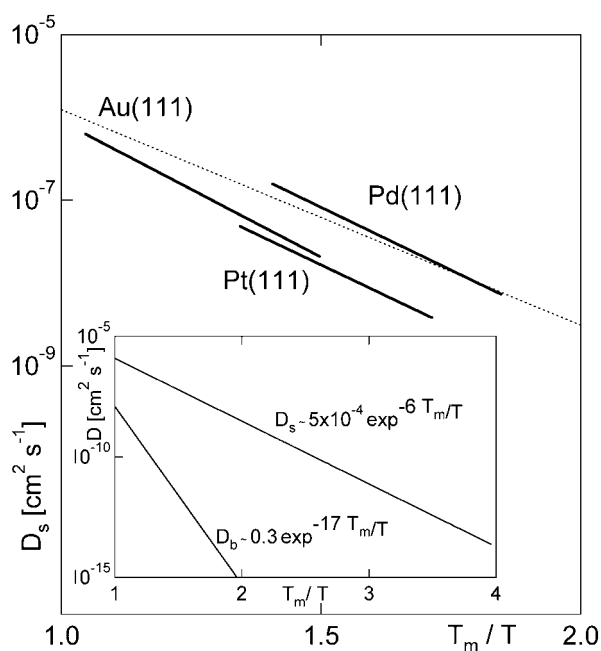


Figure 3. Surface mass diffusion coefficients for some close-packed surfaces shown as functions of T_m/T . The values of D_0 and the magnitudes of D are both similar among these metals, as is also the case for bulk diffusion, shown in the inset.

with generally good accuracy. The homologous behaviour goes further in that diffusion is vacancy dominated in all cases, and vacancy formation energies [22] conform well to $E_f = 10 k_B T_m$. Also, since formation entropies are 1–2 k_B , it follows from equation (4) that motion energies and entropies exhibit similarly reproducible homologous behaviour. These facts have led one of us [23] to identify the notion of a ‘Standard Metal’ with fixed diffusion parameters relative to T_m , from which specific metals have properties that depart to a greater or lesser (but not large) degree.

From the present observation of homologous behaviour for diffusion on close-packed surfaces has arisen [23] the more speculative idea of a ‘standard close-packed surface’. In our results, the activation energy for surface diffusion is about $6 k_B T_m$, and, largely from theoretical modelling [24–26], we assign this to a formation energy of about $4.5 k_B T_m$, and a motion energy of about $1.5 k_B T_m$. At present these diffusion parameters are not yet accurately determined. In measured cases the diffusion prefactor is $\sim 5 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1}$, so that the suggested equation comparable to equation (4) is

$$D_s = 5 \times 10^{-4} \exp -6T_m/T \text{ cm}^2 \text{ s}^{-1}. \quad (5)$$

The dominant defect whose properties these numbers reflect is widely supposed to be the adatom, but even this is not yet an established fact [1, 4]. Typical diffusion rates for the bulk and for close-packed surfaces of metals are contrasted in the inset of figure 3.

Even these rough estimates identify surprising properties with some confidence, and suggest a range of others for future attention as data accumulate. First, in the former category, the given formation energy together with an entropy of k_B determines the defect concentration at T_m as $\sim 3 \times 10^{-2}$; actual values can hardly be much smaller, because the identified formation energy is already three-quarters of the measured $6 k_B T_m$ activation energy for diffusion, and

the actual formation entropy may well be larger than our estimate because the vibrational frequencies of adatoms are reduced from typical lattice frequencies. Second, the density of 3×10^{-2} is so large that the surface advacancy is reaction controlled above about $T_m/3$, its life cycle dominated by spontaneous pair creation on terraces and its annihilation by encounters with diffusing adatoms. If, as is likely, the minority defect has similar if smaller population [26], the same remarks also apply to the majority defect. In any event, the defect concentrations are so large that mobile small defect clusters are expected to contribute significantly to mass diffusion (even clusters with tens of atoms diffuse [27]). For example, the dimer concentration $3c^2 \exp b/k_B T$ for $c \sim 3 \times 10^{-7}$ is comparable with c for reasonable binding energy b , and larger still if the adatom formation entropy is larger than k_B , as seems likely. Accordingly, diffusion on terraces at high temperature is a complex process involving defect reactions. These conclusions are proposed for all close-packed surfaces. Rougher surfaces can reasonably be believed [28] to exhibit reduced formation energies, and hence still larger defect populations.

The second important feature of these models is that their homologous diffusive behaviour relative to T_m reveals a remarkable universality that constrains physical processes of some interest. Two examples are epitaxial growth and step fluctuations. In the case of superlattice growth, the condition that surface diffusion is fast enough for deposited atoms to reach step edges, while bulk diffusion is slow enough to prevent layer-to-layer mixing, identifies an optimum growth temperature of about $T_m/3$. This agrees well with observations [29] that metal superlattices grow well at about $3T_m/8$. Again, in the fluctuations of step edges on close packed surfaces, equations (3)–(5) predict a crossover from surface to bulk diffusion [14, 17] for fluctuations of wavelength $1 \mu\text{m}$ near $0.6 T_m$; this agrees well with our observations of a transition from surface to bulk activated relaxation for steps on Pt(111) and Pd(111). Earlier research identified a similar crossover for smoothing of surface scratches [30] by surface and bulk diffusion, and the present perspective suggests that this transition should occur in the same range of homologous temperatures for all close-packed surfaces. The extent to which these predictions are in fact correct must be established by systematic experimental studies.

5. Ion beam acceleration of surface processes

Ion beams offer a means to perturb surfaces in the direction of enhanced defect populations and accelerated kinetics. It is expected that a regime of linear response will be followed by nonlinear processes when the defect concentrations undergo large fractional changes. A theory is available to describe the response [23]. The resulting behaviour offers an opportunity to probe defect concentrations and mobility, and also to detect minority contributions, depending on the particular defect parameters that prevail. We have undertaken a programme to install an accelerator on our LEEM to create beam of self-ions incident on samples with tunable low energy of impact. One specific focus of interest is the neutral condition in which the impact energy is so chosen that sputtered ions exactly compensate the incident flux, resulting in zero growth. It appears that step fluctuation experiments may thus be performed to determine the mass diffusion coefficient as a function of beam flux at various temperatures, for analysis in terms of the kinetics of reacting defect systems. Estimates of the type portrayed in section 4 suggest that ion currents of some $\mu\text{A cm}^{-2}$ are needed where metals grow at temperature $\sim 3T_m/8$. A second matter of interest near this impact energy is the analyticity of the surface evolution as the energy is tuned from growth to surface removal. We expect to employ these methods for the close-packed surfaces already studied under equilibrium conditions.

In initial experiments to investigate ion beam effects [31] we have employed a 1 keV Ar^+ ion beam to study the way that close-packed surfaces evolve under light sputtering at various temperatures. Together with a quantity of earlier work [32–35] in this area these results identify complex behaviour to be anticipated from future self-ion beam studies, including the

formation of mounds or ripples during sputtering. The central aim in our research was to identify a temperature above which surface recovery is sufficiently rapid that the step edges observed by LEEM remain well defined. Then the net effect of the ion beam is to cause steps to flow uphill at a rate consistent with the material removed by sputtering.

Experiments were performed at temperature, and the LEEM studied after sufficiently rapid cooling that the surface was essentially frozen in its sputtered configuration. The surface evolution depends somewhat on local miscut. We have studied Pd(111) and Pt(111), with selected results summarized below. In what follows sputter-exposures by a beam directed at 45° to the surface are quoted in ML, according to direct calibration with a surface profilometer.

Figure 4 shows a selection [31] of effects observed on sputtered Pd(111), with the initial surface shown in (a). Above 1010 K, the step edges remain clearly visible, and flow as expected in our interpretation; in figure 4(b), flow past fixed screw dislocations causes step profiles with bulges that have then decayed progressively with elapsed time, as expected from the combined effect of Gibbs–Thompson forces and terrace diffusion. Four different behaviours are observed. At 780 K, sputtering causes small islands of each antidefect to form, as in figure 4(c). There is a tendency for the sputtered surface to create alternating bands of lesser and greater step density (i.e., step bunching of a sort), where steps in the flatter areas develop strong undulations. At 780 K and only 18 ML sputtering, large portions of the surface develop sinusoidal profiles that, significantly, have similar phases on successive steps, as illustrated in figure 4(d). Under these conditions, the steps flow one terrace length each 7 s, while the relaxation time estimated by extrapolation from equilibrium fluctuations at high temperature is ~ 20 s. At least a fraction of this important effect arises as successive steps flow by sputtering through surface heterogeneities. This is demonstrated in figure 4(e), which shows several cases where a train of steps have clearly decayed after flowing past a heterogeneity. There is no evidence that the heterogeneities are pre-existing; they may in fact be sputter-induced structures made from slower-diffusing surface defects. Note that the erosion itself eventually brings to the surface all bulk defects created by the beam, so that bulk defects created by the ion beam are constantly brought to the surface by erosion.

At longer sputtering times the profiles change into sharp-crested 3D ripples, like wind-induced ripples on sand dunes, as shown in figure 4(f). The LEEM images give the impression of deep ripples, but in fact the aspect ratio of the ripples is a few step heights in fractions of $1 \mu\text{m}$, or about 1 in 10^3 . The sharp edges point downhill, so the ripples are sharp topped. They are remarkably similar in shape to cell profiles observed in the Mullins–Sekerka [36, 1] instability of solidification interfaces which, consistent with this analogy, are believed to exhibit the same type of instabilities that occur in step flow [1]. The type of profile observed is well reproduced by heuristic calculations based on flow of a slowly diffusing surface defect [1] to a flowing step that absorb defects differently from its uphill and downhill terraces (Ehrlich–Schwoebel effect [37, 38]). This explanation is therefore consistent with a steady state resulting from combined diffusion and sputter-induced erosion. Eventually, heterogeneities precipitate at the singular points, as seen in figure 4(g).

We remark that some of the same features appear when Pt(111) is sputtered, as in 4(h), although they occur over a narrower temperature range. It is apparent that ion beams create elegant and complex step structures, in addition to their role in accelerating surface kinetics. We expect that future use of self-ions at low impact energies to avoid all contamination will help to simplify the surface chemistry and make the various observed behaviours more easily understood. The ability to observe these processes in real time as they occur at various sputtering rates and impact energies will obviously make for a more powerful approach to these important issues.

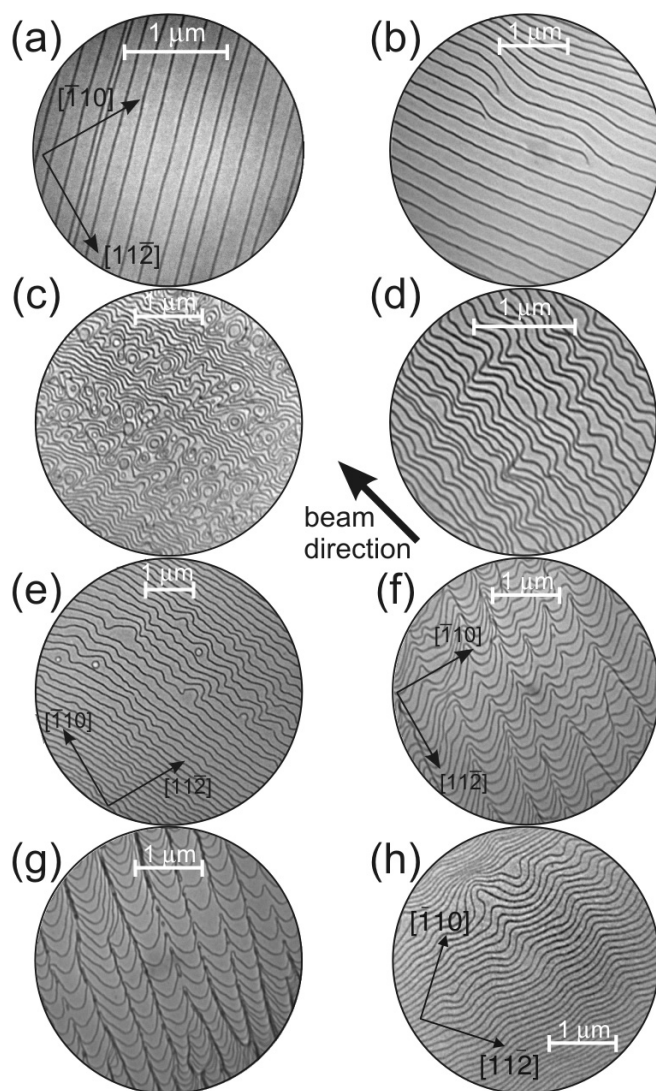


Figure 4. Evolution of Pd(111) under sputtering by 1 keV Ar^+ ions. (a) Initial surface at 1095 K. (b) Flow past screw dislocations during 180 ML sputtering at 1010 K, showing successive steps perturbed. (c) Step bunching occurs after 180 ML at 630 K, with islands and fluctuations in flatter bands. (d) After 18 ML at 800 K sinusoidal ripples appear, and (e) trains of correlated step bulges occur where successive steps have flowed past a surface heterogeneity (bright spots). After longer sputtering (45 ML and more at 800 K) the ripples become asymmetric (f) with sharp downhill edges, and eventually (g) develop heterogeneities next to the tips. (h) Pt(111) at 820 K and 270 ML, similar to (f) for Pd. Note that LEEM enhances the apparent structure; the true aspect ratio of the ridges is $\sim 10^{-3}$.

6. Summary

We employ LEEM in systematic investigations of kinetics and energetics on close-packed metal surfaces. From equilibrium measurements with step fluctuation spectroscopy we obtain surface mass diffusion coefficients and step stiffnesses through a temperature range that is of

interest in connection with growth and processing of these surfaces. Systematic behaviour of these properties from one surface to the next is beginning to emerge.

In future research, the addition of an accelerator that produces a beam of low energy self-ions will allow us to explore the changes in surface kinetics and surface evolution in the absence of chemical contamination. Initial efforts using an Ar⁺ ion beam have revealed a wealth of details relating to the interesting topological evolutions exhibited by close-packed surfaces in the temperature regime in which surface recovery is rapid enough that step edges remain intact.

In earlier research we observed steps interacting with reconstruction boundaries, crystallographic facets, threading dislocations as they glide and a variety of other surface nanostructures [39, 40]. From the equilibrium configurations, given forces in equilibrium, it is possible to infer their energies in terms of β . Thus the exploration of step fluctuations offers a rather wider eventual access to the energetics of small structures on surfaces, which are generally difficult to determine, than would otherwise have been apparent.

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