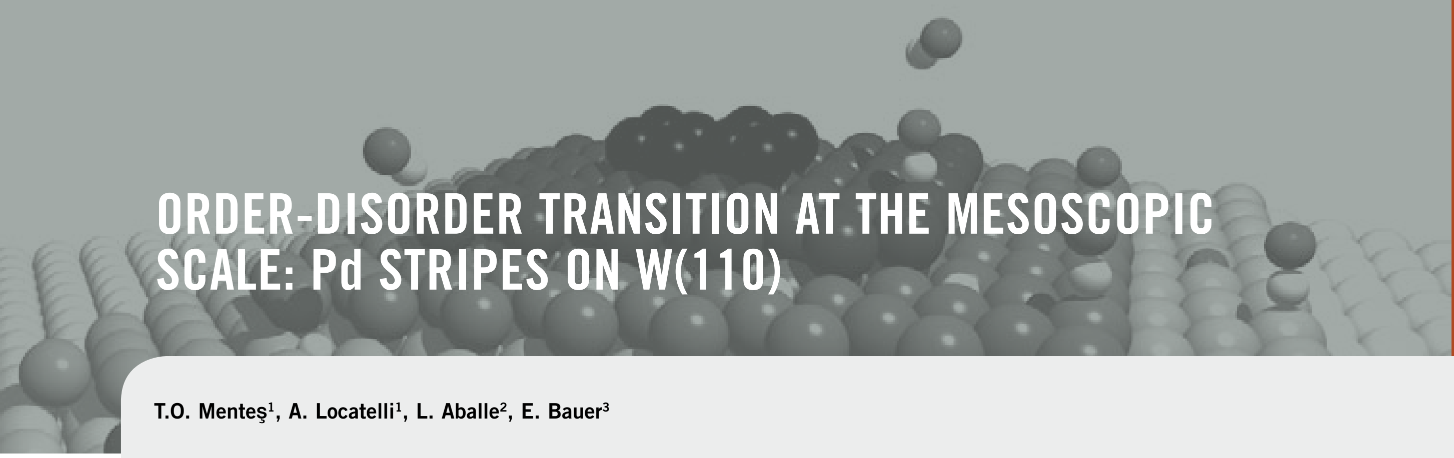

ORDER-DISORDER TRANSITION AT THE MESOSCOPIC SCALE: Pd STRIPES ON W(110)

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Spontaneous formation of patterns is a common occurrence in nature. From the Turing patterns during chemical reactions to the wavy stripes of sand dunes, one can find a plethora of examples with almost periodic shapes and forms. In addition to their aesthetically pleasant regularity, such patterns usually appear at length scales surprisingly larger than their building blocks (like the dunes made of small sand particles). This is generally due to the interplay between opposing processes, which may extend to long distances.

Most commonly, the pattern is formed out of equilibrium, and is kept in a steady-state by a driving force (the wind for sand dunes, a thermal gradient for convection patterns, etc) [1]. On the other hand, in some cases quasi-periodic structures can be observed in thermodynamic equilibrium, that is at the minimum of the free-energy. The mesoscopic order is stabilized by competing interactions with different range of influence. Short distance interaction can be bonding (in the case of atoms and molecules) or exchange between constituents, whereas at the long-range we generally find electrostatic, dipolar or elastic type interactions.

In this study we illustrate the formation of adlayer patterns induced by the elastic interaction between adlayer steps. Surface stress had already been shown to cause a periodic ordering of atomic steps on crystal surfaces [2]. The competition, which determines the pattern period, is between the energy cost of step creation due to bond breaking, and the elastic energy gain due to stress relaxation at the step. The period depends exponentially on the interaction parameters and easily reaches mesoscopic length scales above 100 nanometers.

Using low-energy electron microscopy (LEEM), a method which allows imaging structure of surfaces with a lateral resolution of about 10 nanometers, we have observed that a small amount of palladium (less than a single atomic layer) on W(110) spontaneously orders into monolayer stripes at high temperatures [3]. A series of images of Pd stripes on W(110) at different temperatures is shown in Figure 1. The alternating bright and dark areas correspond to regions covered with a Pd monolayer and those covered with a dilute Pd lattice gas, respectively. The pattern period and the contrast sharply decrease with increasing temperature.

The variation of the period as a function of temperature is plotted in Figure 2a. We have shown in ref [3] that this variation is due to the broadening of the stripe boundaries. As the boundary gets smeared out with increasing temperature, the energy cost of step creation decreases. However, the same broadening also reduces the elastic relaxation energy, counterbalancing the influence of the boundary energy at the highest temperatures before the stripes disorder. The pattern disorders at T_c = 1170 K.

Figure 1.
LEEM image series of Pd stripes on W(110) surface as a function of temperature. Regions covered with the Pd monolayer are bright. Image size is 2×2 μm^2. Temperature increases from left to right (1100 K, 1120 K, 1130 K, 1145 K and 1160 K). The curved lines correspond to monatomic steps on the tungsten substrate.

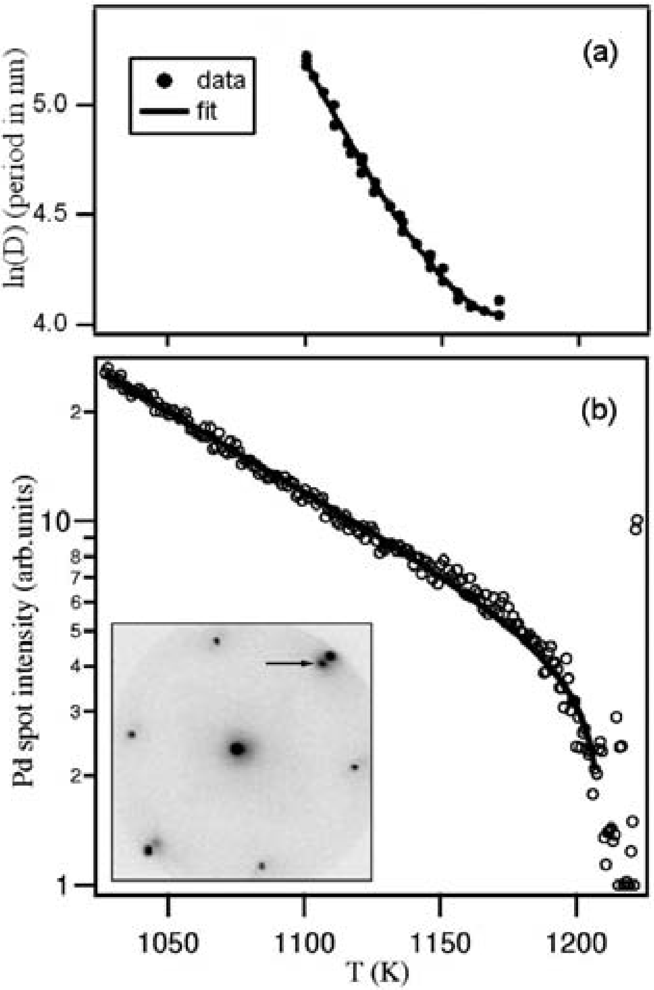

Figure 2.
The logarithm of the stripe period as a function of temperature is displayed in panel (a). Panel (b) shows the temperature dependence of the Pd low-energy electron diffraction spot intensity. The LEED pattern is seen in the inset.

We have shown that the temperature dependence of the pattern period can be described using the results from the two-dimensional Ising model [3]. As shown in Figure 2a, the variation is reproduced very well. On the other hand, the critical temperature found by fitting the stripe period, $T^0_c = 1202$ K, is considerably higher than the stripe disordering temperature T_c. This apparent discrepancy finds an explanation when we consider the order at the atomic scale. Condensed regions of Pd show a distinct diffraction spot, as marked by an arrow in the inset of Figure 2b. With increasing temperature, the intensity of the Pd spot decreases, as a consequence of Pd atoms subliming from condensed regions into the lattice gas. However, the spot is present, although increasingly weak and diffuse, all the way up to about T^0_c, as can be expected from the Ising model without long range interaction.

Our experiment is a direct confirmation that the short-range order at the atomic scale survives at temperatures higher than the disordering temperature of the mesoscopic pattern. Moreover, the difference between T_c and T^0_c depends on the ratio of the short and long range interaction parameters. These observations are relevant for order-disorder transitions in all pattern forming systems.

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