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In-Situ Video-STM Studies of the Quasi-Collective Motion of Nanoscale Metal Strings in Au(100) Electrode Surfaces

Miguel Labayen, Olaf M. Magnussen

Institut für Experimentelle und Angewandte Physik, Universität Kiel, 24098 Kiel, Germany

Mass transport on metal surfaces is of central importance in homo- and heteroepitaxial growth, structural phase transitions, and surface restructuring processes. On the atomic scale it is usually described by diffusion of individual metal adatoms on atomically flat terraces and along steps, as well as the interaction of these adatoms with each other and with surface defects [1,2]. Here we report observations on Au(100) electrode surfaces in acidic solution during the potential-induced (1×1) → "hex" surface phase transition that reveal quasi-collective lateral motion of five atom wide, hexagonally ordered strings of Au atoms embedded in the square lattice of the Au(100)-(1×1) surface.

The measurements were performed using a novel high-speed in-situ scanning tunneling microscope (Video-STM), recently developed in our group [3,4]. With this instrument 1-3 min. long video sequences of atomic-scale processes at electrochemical interfaces can be recorded at image acquisition rates of 10-25 images per second. This technique was applied to studies of single crystalline Au(100) electrodes in Cl-containing Na_2SO_4 solution. Starting from a potential of +0.45 V vs. the saturated calomel electrode (SCE), where the Au surface exhibits a square (1×1) lattice, the potential was rapidly decreased to –0.1 V, where the "hex" reconstruction of Au(100) is formed [5-7]. In the "hex" phase the topmost Au layer is hexagonally ordered with a 25% higher packing density than Au(100)-(1×1). Formation of this potential-induced reconstruction proceeds via nucleation and growth of 14.5 Å wide strings roughly parallel to the {110} directions, in which six hexagonally ordered rows of Au surface atoms reside on five atomic rows of the underlying bulk (100) lattice, resulting in a uniaxial, vertical modulation of the Au surface layer.

As shown in FIG. 1, these elementary units of the Au(100) reconstruction are highly mobile objects, which exhibit fast quasi-collective lateral motion perpendicular as well as along the string direction. The perpendicular motion occurs in form of discrete jumps and can be explained by small displacements of Au surface atoms in the strings and the neighbouring atomic rows. As revealed by a detailed statistical analysis several jump mechanisms contribute to this type of string motion. In particular, the rate of jumps by one and by two Au lattice distances is almost identical. Motion along the string direction requires an exchange of Au atoms between the two string ends. The latter phenomenon may be explained by the propagation of structural distortions within the strings or quasi-onedimensional transport of Au adatoms on top of the strings. In addition, even more complex dynamic phenomena have been observed in Video-STM sequences, such as attractive interactions between neighbouring "hex" strings, the breaking of strings into two, independently moving parts, the recombination of two strings, and transient disordering of large sections of strings [8].

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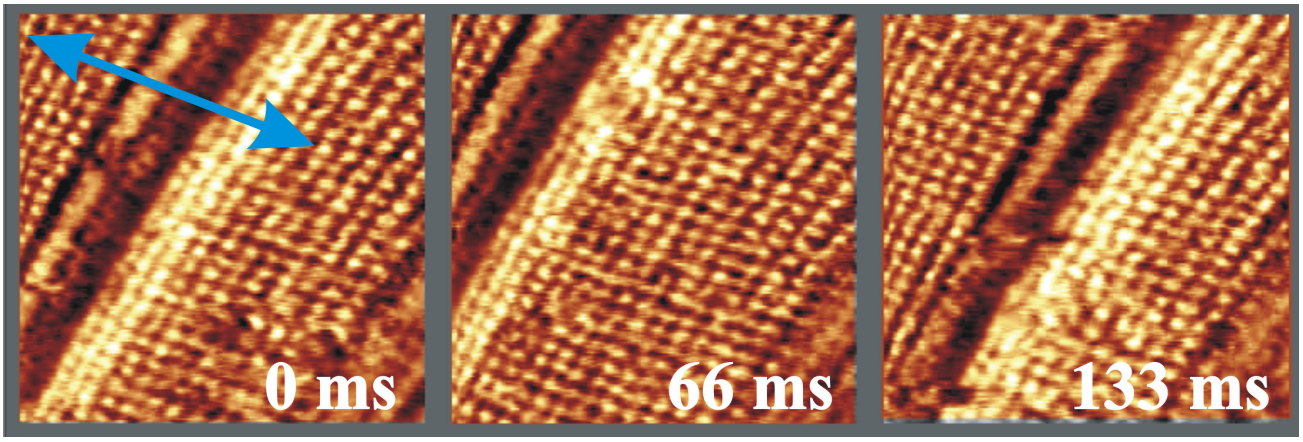

FIG. 1: Atomic-resolution Video-STM sequence of a single “hex“ string on Au(100), obtained from a 15 frame/s video (60×60 Å^2). The sequence was recorded at –0.1 V vs SCE in 10 mM Na_2SO_4 + 1 mM HCl. It illustrates the lateral motion perpendicular to the string direction (indicated by arrow) with a displacement of 3 atomic lattice distances during the first 66 ms and 6 during the following 66 ms.

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