Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information

In situ scanning tunneling microscopy studies of carbonate-induced restructuring of Ag-decorated Cu(100) electrodes

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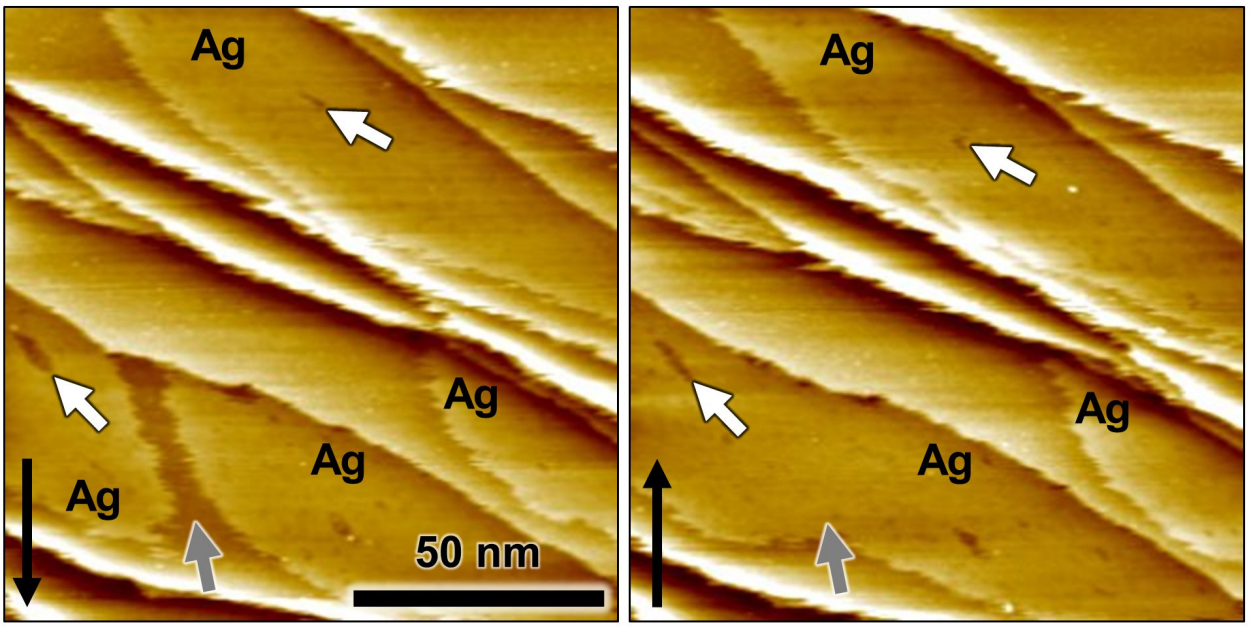

Figure S 1: Successive STM images of Ag-decorated Cu(100) in 0.1M H_2SO_4 at -0.04 V(time interval 126 s), illustrating the dynamic behavior of Ag islands on the mesoscale. The grey arrows indicate an area, where a larger island formed by merging of two Ag islands. The areas marked by white arrows are depressions that appear inside Ag islands, appear to have a limited mobility within them, and are attributed to holes within the Ag monolayer.

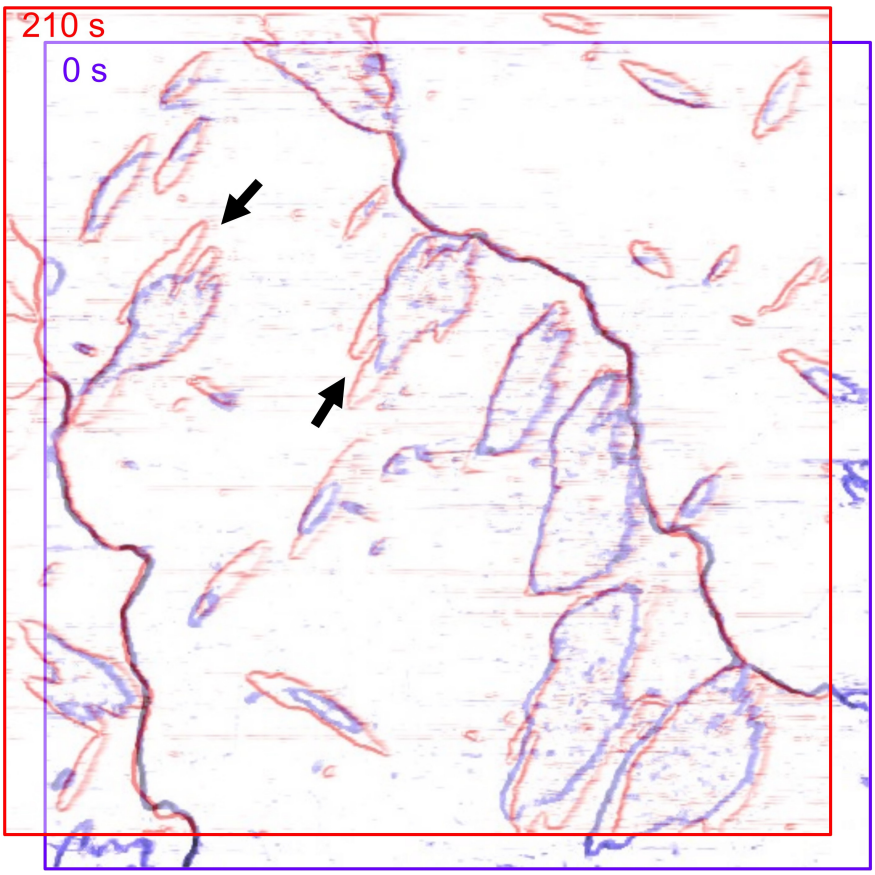

Figure S2: A direct comparison of the shapes of Ag islands and Cu steps in the two STM images in Figure 2d (blue) and f (red), respectively, recorded at a time interval of 210 s. The steps were determined using a Sobel edge enhancement kernel. Images recorded in the same scan direction were chosen to avoid artifacts due to thermal drift. While the Cu step edges show only minor smoothing, the Ag islands change significantly in shape and size. Examples where increases in the size of the islands are clearly visible are marked by black arrows.

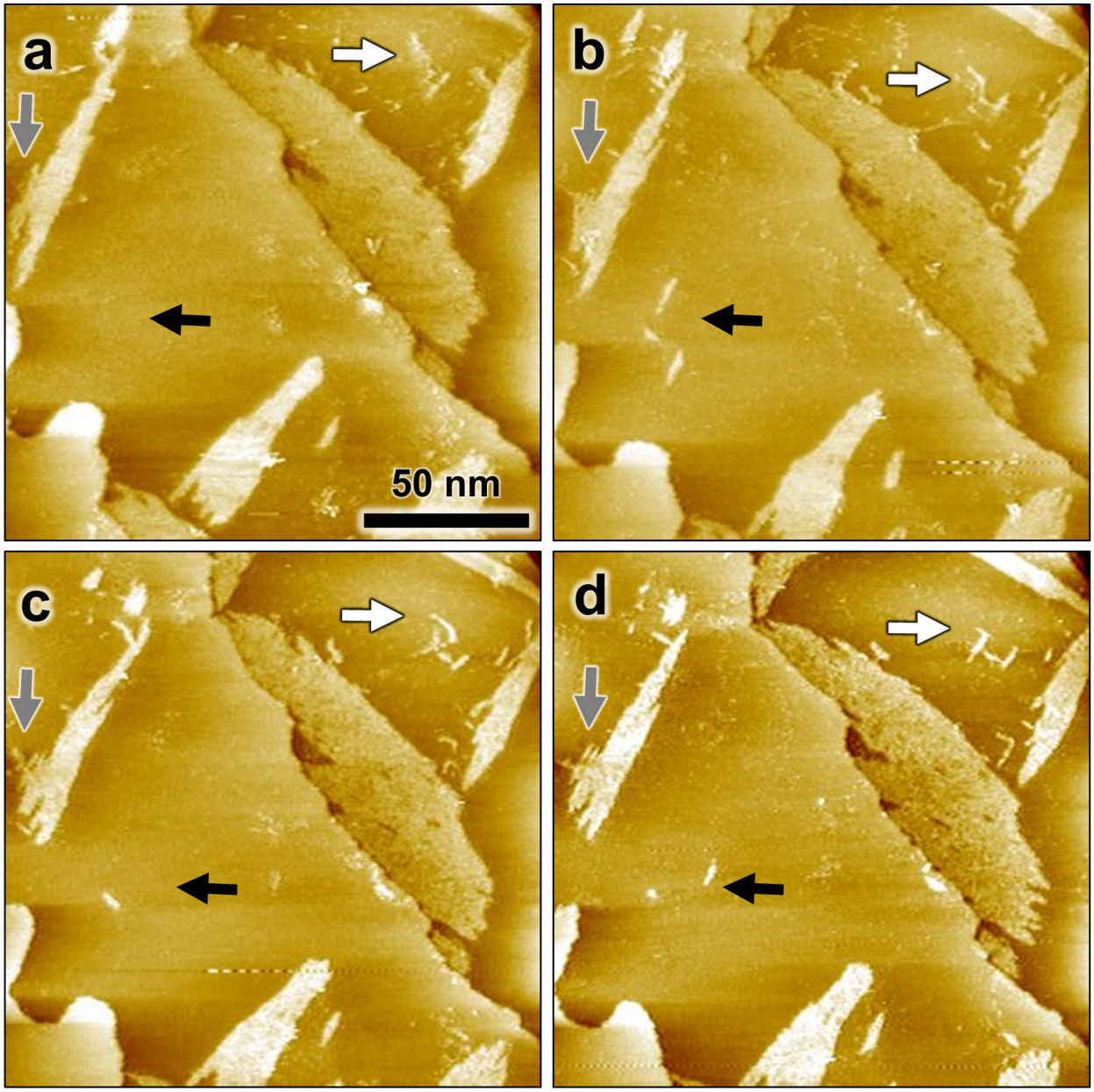

Figure S3: Subsequently recorded *in situ* STM images (time interval 84 s), taken at -0.01 V and about 100 min. after electrolyte exchange to 0.1M $KHCO_3$, that show moderate changes in the Ag island morphology on the Cu(100) surface. Occasionally, small anisotropic islands emerge on the Cu(100) terraces and restructure or disappear again (black and white arrows). In addition, some rearrangement at the edges of the Ag islands is observed (grey arrows).