## **Description of Additional Supplementary Files**

File Name: Supplementary Movie 1

Description: LSMO (001) surface in 0.5 Pa  $H_2O$ . Observation of a high Mn adatom mobility on flat A-terminated (001) surfaces due to partial solvation in  $H_2O$ . Frame rate is 4.4 fps.

File Name: Supplementary Movie 2 Description: LSMO (001) surface with a step in 0.5 Pa  $H_2O$ . Observation of a reduced Mn adatom mobility due to Erich-Schwöbel barrier at a step edge on the (001) surface. Frame rate is 4.4 fps.

File Name: Supplementary Movie 3 Description: LSMO (001) surface in high vacuum ( $10^{-5}$  Pa). Observation of the (001) surface with mixed A- and B-termination shows a reduced Mn mobility compared to H<sup>2</sup>O. Frame rate is 4.4 fps.

File Name: Supplementary Movie 4 Description: LSMO (001) surface in 100 Pa  $O_2$ . Highly ordered (001) A-terminated surface with a step edge and reduced adatom mobility compared to  $H_2O$ . Frame rate is 4.4 fps.

File Name: Supplementary Movie 5

Description: LSMO (001) surface in 10 Pa  $N_2$ . Ordered B-terminated (001) surface that shows a reduced Mn adatom mobility compared to  $H_2O$ . Frame rate is 4.4 fps.

File Name: Supplementary Movie 6

Description: PCMO (001) surface in 0.5 Pa  $H_2O$ . Ordered PCMO (001) surface that shows irreversible Mn dynamics with a pronounced leaching of Mn out of the surface and the formation of a Pr-rich bilayer. Frame rate is 4.4 fps.

File Name: Supplementary Movie 7

Description: PCMO surface close to (001) in high vacuum (10<sup>-5</sup> Pa). Disordered PCMO surface close to (001) typically observed in high vacuum, where leaching of Mn is absent. Frame rate is 4.4 fps.

File Name: Supplementary Movie 8.

Description: PCMO surface in 100 Pa  $O_2$ . Highly ordered surface with (001) facets and mixed termination, where leaching of Mn is absent. Frame rate is 4.4 fps.

File Name: Supplementary Movie 9.

Description: Preparation of ordered LSMO (001) surfaces in 100 Pa  $O_2$ . Initial state of the LSMO surface after ion milling before recrystallization in  $O_2$ . Frame rate is 4.4 fps.

File Name: Supplementary Movie 10.

Description: Preparation of ordered LSMO (001) surfaces in 100 Pa  $O_2$ . Final state of surface recrystallization in  $O_2$ . Frame rate is 4.4 fps.