Surface-supported metallic mesostructures through reaction-controlled phase separation

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Homogeneous multi-component systems far from thermodynamic equilibrium can undergo spontaneous phase separation to reduce the total free energy of the system. This process, known as spinodal decomposition, is accompanied by development and evolution of spatial periodic structures. For chemically reacting systems spinodal decomposition as a mechanism for structure formation was first suggested in the theoretical studies of polymer mixtures [1]. It has been predicted that also adsorbates on metal surfaces under reaction conditions can spontaneously re-organise into stationary or traveling periodic non-equilibrium nanostructures [2]. Stationary structures resulting from reactive phase separation driven by pure energetic factors were recently observed for metal adlayers on Rh surfaces [3]. These phenomena, described as "chemical freezing" of phase separation, persist only in the presence of a chemical reaction and can be controlled by varying the reaction conditions.

In the present work, we report the formation of a variety of stationary two-dimensional metallic and oxygen structures in Au and Au+Pd adlayers on Rh(110) under water formation reaction. We exploit combined use of LEEM and XPEEM to study the dependence of the morphology of metal adlayer structures on the reaction rate. We show that Au and AuPd structures result from chemically frozen spinodal decomposition, and are created, preserved or reversibly modified by tuning the reaction conditions. The wavelength of lamellar structures obtained at intermediate metal coverage is found to obey a power scaling law with respect to the reaction rate over more than two decades of the reactants pressure.

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