IN-SITU GROWTH OF GRAPHENE-LIKE TWO-DIMENSIONAL SILICA NETWORKS ON METALS

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Two-dimensional materials have attracted much attention in last years due to their unusual but very advantageous properties. Silicon dioxide films are interesting due to their use in many technological applications, e.g., as insulating barriers in devices, as a gate oxide in field effect transistors, for applications in catalysis, or for protecting metal surfaces. Recently it became known that a two-dimensional crystalline phase of silica exists. It is composed of single layer or bilayer corner-sharing SiO_4 tetrahedra in a hexagonal network. In top-view, the honeycomb-like lattice of the structure resembles that of graphene and shows exactly the same types of defects (pentagons, heptagons, octagons, etc.).

All studies reported until now have been ex-situ experiments and could not reveal the growth mechanism of hexagonal silica. Here we report a new route to grow the silica layers in a solid-state process that we have carried out *in-situ* in a transmission electron microscope [1]. The experiments allow us to obtain detailed information on the nucleation, growth, and the epitaxial relationship with the crystalline symmetry of the metal surface. The structure was grown on different catalytically active metal layers (Fe, Co, Ru) at temperatures around 500°C. The growth was monitored at high image resolution and by electron energy-loss spectroscopy. Whereas the cells form a hexagonally ordered network with an epitaxial match on the (0001)-surfaces of hexagonally close-packed Co and Ru layers, growth on other metal surfaces shows a disordered arrangement of the cells ("vitreous silica") and the appearance of defective structures. Furthermore, we have observed the same reconstruction of defects under electron irradiation as in graphene (Fig. 1).

A growth of a one-dimensional silica phase was also observed under similar conditions [2]. Lines of SiO_4 tetrahedra grow along surface steps on metal substrates (Fig. 2). The growth occurs in competition with the formation of two-dimensional structures. We have shown experimentally and by ab-initio calculations that linear growth along surface steps is energetically favorable.

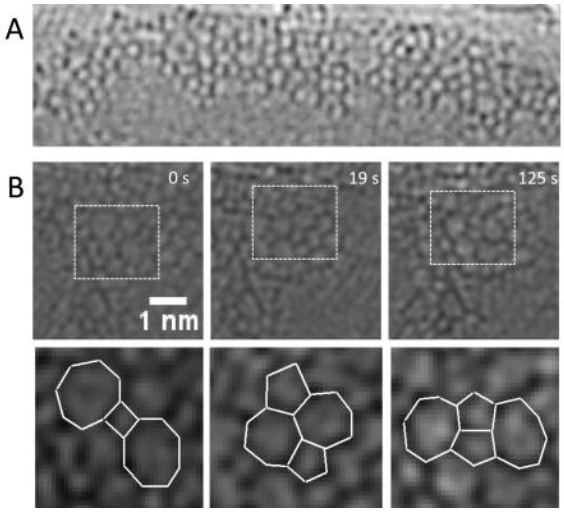

Figure 1: Structural defects in the hexagonal silica network grown at 470°C on a Ru surface. (a) Arrangement of vertical defect lines (pentagon pairs, interrupted by heptagons or octagons) in a partially ordered network. (b) Reconstruction of a typical defect as a function of time.

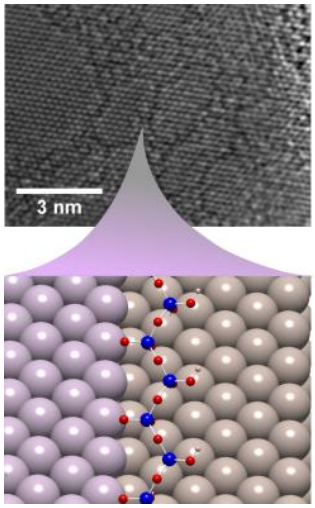

Figure 2: The growth of one-dimensional silica along steps on the surface of Co.

[1] F. Ben Romdhane, T. Björkman, J. A. Rodríguez-Manzo, O. Cretu, A. V. Krasheninnikov, and F. Banhart. ACS Nano, 2013, 7 , 5175–5180.

[2] F. Ben Romdhane, T. Björkman, A. V. Krasheninnikov, and F. Banhart. J. Phys. Chem. C, 2014, 118 (36), 21001-21005.