Detecting and resolving individual adatoms, vacancies, and their dynamics on graphene membranes

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Graphene is a single atomic layer of graphite that has only recently become experimentally accessible in an isolated form [1]. We prepare graphene into free-standing membranes [2,3], i.e., a crystalline foil with a thickness of only one carbon atom. These membranes are highly promising for TEM studies, since (a) this membrane itself is of tremendous scientific interest, (b) adsorbates can be studied against a highly transparent and crystalline background, and (c) the precisely known structure is an ideal tuning and calibration tool for electron microscopy developments. Graphene membranes are very stable at low acceleration voltages, we and present results obtained on a conventional TEM (Jeol 2010 at 100kV) as well as initial tests on the new aberration-corrected TEAM 0.5 microscope operated at its lower limit of 80kV. The TEAM 0.5 microscope achieves sub-Angstrom resolution even at 80kV, thus resolving every single carbon atom in the graphene lattice.

Individual low-atomic number adatoms on a clean graphene membrane are detected, as well as individual vacancies or molecular scale adsorbates. The dynamics of these objects is shown, such as the replacement of vacancies from mobile adsorbates. By using the aberration-corrected microscope, even subtle effects such as the relaxation of the graphene lattice near an adatom or vacancy can be detected. Further, the binding configuration of an individual adatom with the graphene membrane can be discerned. Damage occors predominantly at the edges of the sheet, where individual carbon atoms can be seen to move around inbetween individual exposures. Fig. 1 shows graphene membranes prepared across 1 μm holes in a carbon film. Fig. 2 shows atomic resolution images of graphene samples. Fig. 2a shows a single- vs. a two-layer graphene membrane in one image for comparison. Since single-layer graphene is only half a unit cell in the c-direction of graphite, it has a unique signature both in the direct image as well as in a diffraction pattern. Fig. 2b shows an adatom that is centered between the positions of two carbon atoms of the graphene lattice, as is expected for a carbon adatom [4]. We expect that a controlled placement of atoms and molecules on graphene, in combination with aberration-corrected electron microscopy, will open new avenues to observe chemical interactions or structural modifications of low contrast molecules or nanoobjects.

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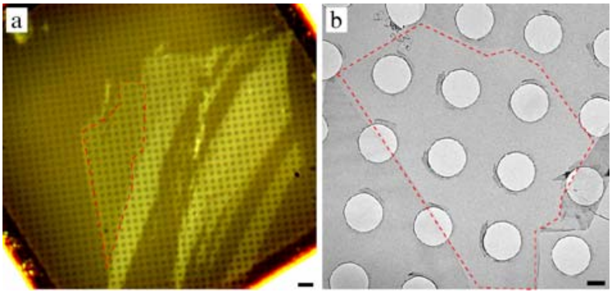

Figure 1. (a) Optical micrograph of graphene sheets on a perforated carbon film (c-flat, 1 µm hole size), and (b) low magnification TEM view of a graphene sample. The single-layer regions are indicated by red dashed lines. Scale bars: (a) 2 µm, (b) 500 nm.

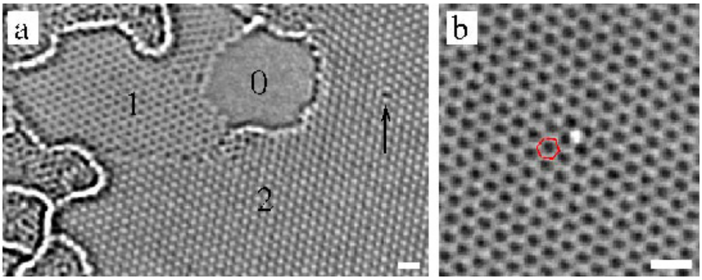

Figure 2. Atomic resolution images of graphene samples, obtained with the TEAM 0.5 microscope at 80 kV, Cs=-15µm. Atoms appear white. (a) Sample showing a zero- (vacuum), one- and two-layer graphene area. Arrow points to a vacancy. (b) Adatom on a single-layer graphene membrane. The hexagonal arrangement of the carbon atoms is indicated by the red hexagon. The adatom is centered between two carbon atoms of the graphene lattice, and induces a small distortion into it. Scale bars are 0.5 nm.