

Comment on “Electrostatic Force Microscopy on Oriented Graphite Surfaces: Coexistence of Insulating and Conducting Behaviors”

In a recent Letter [1], Lu *et al.* claimed to have identified insulating and conductive regions on a highly oriented pyrolytic graphite (HOPG) surface by differences in the surface potential. Because of the reasons given below, the results presented do not give sufficient evidence to support the claims made.

(1) The minimum of the force gradient signal corresponds to the contact potential difference (CPD) and appears to be at the same bias voltage for the tip on the dark and the bright regions [Fig. 1(a) in Ref. [1]]. This is in disagreement with the statement that the CPD is different by 0.25 V between the two regions. The square root of this curve does not result in a suitable calibration curve, as its slope depends on the tip-sample distance [Fig. 5(d) in Ref. [1]] and on the dielectric constant of the sample [2]. Variations in the tip-sample distance due to locally different CPD [3] or dielectric constant [2] will result in a change of this slope. The quantification of the CPD from the contrast in the electrostatic force microscopy (EFM) images is therefore not justified.

(2) It was shown by measurements under ultrahigh vacuum (UHV) conditions that graphite peeled off in ambient air shows a CPD lower by 0.5 eV compared to a sample peeled off in UHV (Fig. 1 in Ref. [4]). This result clearly proves that, besides the conditions during measurement, the conditions during sample preparation are also relevant.

(3) The shape of the majority of the regions with a higher EFM contrast appears to be circular. This could indicate the presence of a contamination film (i.e., water or hydrocarbon) on the surface, stemming either from the sample preparation in air or from the tip itself, which likely contains a contamination layer that could be transferred to the sample surface during the measurement. In fact, the images resemble the water films observed in Ref. [2]. Additional evidence can be observed in Figs. 2 and 4 in Ref. [1], where subsequent images show variations in the shape and size of the regions exhibiting the EFM contrast. This demonstrates that the sample is modified by simply scanning the tip across it. The authors explain the EFM contrast by the presence of defects in the graphite structure. In order to change the defect structure, more energy is likely required as could be provided by scanning with an AFM tip. However, modifying a contamination film on the sample requires less energy and is the more likely explanation.

(4) The inverted EFM contrast for positive and negative sample bias (Fig. 3 in Ref. [1]) does not exclude the presence of a contamination layer. In the present case of a conductive substrate (graphite), changing the polarity of the applied bias would change the polarization of the contamination layer (i.e., by reorientation), resulting in

the observed contrast inversion. This effect is absent when an insulating substrate is used, as shown in Ref. [2].

(5) The topography as shown in Figs. 2, 4, and 5 in Ref. [1] shows a curvature which is proposed to be a result of the presence of defects in single graphene layers. This curvature appears only along the x axis in all images and is likely an artifact occurring for tube piezos at large scan sizes. Additionally, the length scale of the topography curvature is on the order of several micrometers, whereas the length scale of the EFM contrast variations is on the order of 1 μm and below. If the same origin, namely, the presence of defects in the graphene structure, is made responsible for both effects, then both the topography and the EFM contrast should vary on the same length scale. In addition, the vertical resolution of the topography scan is fairly low; monolayer steps in graphite (~ 0.34 nm) or the height of a thin contamination layer cannot be resolved [the digital steps in Fig. 2(b) in Ref. [1] have a height of 0.5 nm].

(6) The electrostatic signal obtained in EFM is extremely sensitive to the sample surface and tip condition. The experiments conducted by the authors are subject to uncontrolled conditions, especially when operating in ambient air. Also, for the measurements in Ar atmosphere, the sample and the tip are introduced from ambient air and are likely contaminated. Even for measurements conducted under UHV conditions [4], the sample preparation influences the result (see point 2 above). As differences in the laboratory environment are not controllable, it is not scientifically justified to consider the absence of the EFM pattern on the HOPG-0.8 sample as a proof of different properties of the HOPG-0.8 and the HOPG-0.4 samples.

The arguments given above strongly indicate that the observed EFM contrast is the result of a surface contamination and cannot be attributed to an electronic property of the graphite sample.

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