Correct Height Measurement in Noncontact Atomic Force Microscopy

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We demonstrate that topography measurements by noncontact atomic force microscopy are subject to residual electrostatic forces. On highly oriented pyrolitic graphite (HOPG) with a submonolayer coverage of C_{60}, we monitor the step height from C_{60} to HOPG as a function of dc bias between tip and sample. Because of the different contact potential of C_{60} and HOPG ($\sim$50 mV), the step height is strongly dependent on the dc bias. The presented results and additional simulations demonstrate clearly that for correct height measurements it is mandatory to use a Kelvin probe force microscopy method with active compensation of electrostatic forces.

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Since its development, atomic force microscopy (AFM) [1] has become a successful technique for imaging material surfaces, including insulators, down to the atomic scale [2–4]. Using a dynamic mode or noncontact AFM (NC-AFM) [5,6] the range of applications was extended to surfaces of soft samples, i.e., organic and biologic materials [7,8]. Many applications employ the AFM to study samples with regions of different materials, i.e., DNA on a substrate [9], semiconductor quantum dots on a semiconductor substrate [10], etc. and one information obtained is frequently the size of the objects studied. With respect to the lateral dimensions, the interpretation of the NC-AFM results is straightforward (except for the issues of lateral resolution and tip-sample convolution). However, for the interpretation of the height measurement a discussion of the vertical force gradient — the actually measured physical quantity — is mandatory. In normal feedback mode, the tip position is adjusted to maintain a constant force gradient, which is realized either by controlling the amplitude of the cantilever oscillation (usually applied in air), or by controlling the frequency shift Δf with respect to the free resonance frequency f_0 of the cantilever (FM mode, usually applied in vacuum) [11]:

$$\Delta f = -\frac{f_0}{2k}\frac{\partial F_{\mathrm{tot}}}{\partial z}, \tag{1}$$

where k is the spring constant of the cantilever. The total force F_{tot} between tip and sample is composed of several contributions:

$$F_{\mathrm{tot}} = F_{\mathrm{chem}} + F_{\mathrm{vdW}} + F_{\mathrm{el}}, \tag{2}$$

with the short ranged chemical forces, F_{chem}, the van der Waals force, F_{vdW}, and the long ranged electrostatic force F_{el}. In regular NC-AFM operation, the electrostatic force is minimized, by applying the correct bias voltage V_{bias} between tip and sample:

$$F_{\mathrm{el}} = -\frac{1}{2}\frac{\partial C}{\partial z}(V_{\mathrm{bias}} - V_{\mathrm{CP}})^2, \tag{3}$$

with $\partial C/\partial z$ being the capacitance gradient of the tip-sample system, $V_{\mathrm{CP}} = 1/e \cdot (\Phi_{\mathrm{tip}} - \Phi_{\mathrm{sample}})$ the contact potential (CP), and Φ the work function of tip and sample, respectively. The van der Waals force is independent of V_{bias} and decays faster than the electrostatic force [12]:

$$F_{\mathrm{vdW}} = -\frac{HR}{6d^2}, \tag{4}$$

where H is the Hamaker constant, R is the tip radius, and d is the tip-sample distance. The chemical forces are due to developing bonds between tip and sample atoms, and are relevant only at tip-sample distances below $\sim$5 Å [13]. This mechanism is presumably responsible for atomic resolution imaging.

As enhancement of the regular NC-AFM, the Kelvin probe force microscope (KPFM) was developed [14,15]; it allows one to image the contact potential of the sample by using an additional feedback to adjust the tip-sample bias during the scan.

In this Letter, we show that the height measurement on samples constituted of different materials is strongly influenced by the tip-sample bias. Using KPFM, the electrostatic forces are actively compensated resulting in a height measurement governed only by the van der Waals force [16], which we refer to as the correct height measurement. Experimentally, this effect will be demonstrated on a graphite-C_{60} sample and additional calculations show the general validity of this effect.

The present KPFM is a modified ultrahigh vacuum (UHV)-AFM (Omicron) operated at $p \leq 10^{-10}$ mbar [17]. The topography is measured using the FM mode at the first resonance frequency of the cantilever ($\sim$60 kHz). To detect the electrostatic force, an ac voltage $V_{\mathrm{ac}}\sin(\omega t)$ is applied between tip and sample, in addition to a dc bias V_{dc}. The ac voltage induces oscillatory electrostatic forces according to Eq. (3), where the spectral component at the frequency ω of the ac voltage results to [18]

$$F_{\omega} = -\frac{\partial C}{\partial z}(V_{\mathrm{bias}} - V_{\mathrm{CP}})V_{\mathrm{ac}}\sin(\omega t). \tag{5}$$

The oscillation at ω is detected using a lock-in amplifier and a controller is used to reduce the amplitude to zero by adjusting V_{bias} to match the CP. In the present case, ω is tuned to the second resonance frequency of the cantilever, thereby allowing a highly sensitive, simultaneous, and independent detection of the electrostatic forces. By this procedure, it is possible to use ac voltages as low as 100 mV maintaining a high energy resolution of the measured CP of $\sim$5 meV [17].

The sample consists of a highly oriented pyrolytic graphite (HOPG), which was cleaved in air immediately before mounting it into a vacuum chamber ($p \sim 10^{-8}$ mbar) for the C_{60} deposition. C_{60} was evaporated ($T_{\mathrm{source}} = 450\,^\circ\mathrm{C}$) at a rate of $\sim$0.25 Å/s onto the HOPG ($T_{\mathrm{HOPG}} = 120\,^\circ\mathrm{C}$) until a coverage of $\sim$5 Å, corresponding to $\sim$0.5 monolayer (ML). For the introduction into the UHV-KPFM, the sample was briefly exposed to air. Prior to the study, it was annealed for 30 min at 150 °C in UHV to remove the water contamination layer.

In Fig. 1, several images of the C_{60} on HOPG sample are shown, taken under different measurement conditions. Figures 1(a) and 1(b) show the topography of the sample imaged with NC-AFM at sample bias voltages of 0 and 1.34 V, respectively. The topography in (a) shows a C_{60} island on the flat HOPG substrate. The C_{60} island consists of up to 3 ML and has an irregular shape, in accordance with previous observation for the growth of C_{60} on layered materials [19,20]. In Fig. 1(b), the "island" actually appears below the substrate, still showing three different ML within the patch. However, the contrast between HOPG and C_{60} is inverted. This becomes more clear in the according line scans along the black/white line shown below the images. In addition to the pure NC-AFM topography, we also performed KPFM on the same position, where the topography is shown in Fig. 1(c) and the simultaneously determined CP image in 1(d). The latter image shows clearly the material contrast between C_{60} and HOPG, unambiguously identifying the C_{60} island. The according line scans (below the images) show a difference in CP of $\sim$50 mV between C_{60} and HOPG and the 3 ML island in the topography. From the images and line scans in Fig. 1, it is evident that the electrostatic forces strongly influence the height measurement in NC-AFM. A measurement free of electrostatic forces as conducted by KPFM reveals a height measurement based only on the van der Waals forces [16].

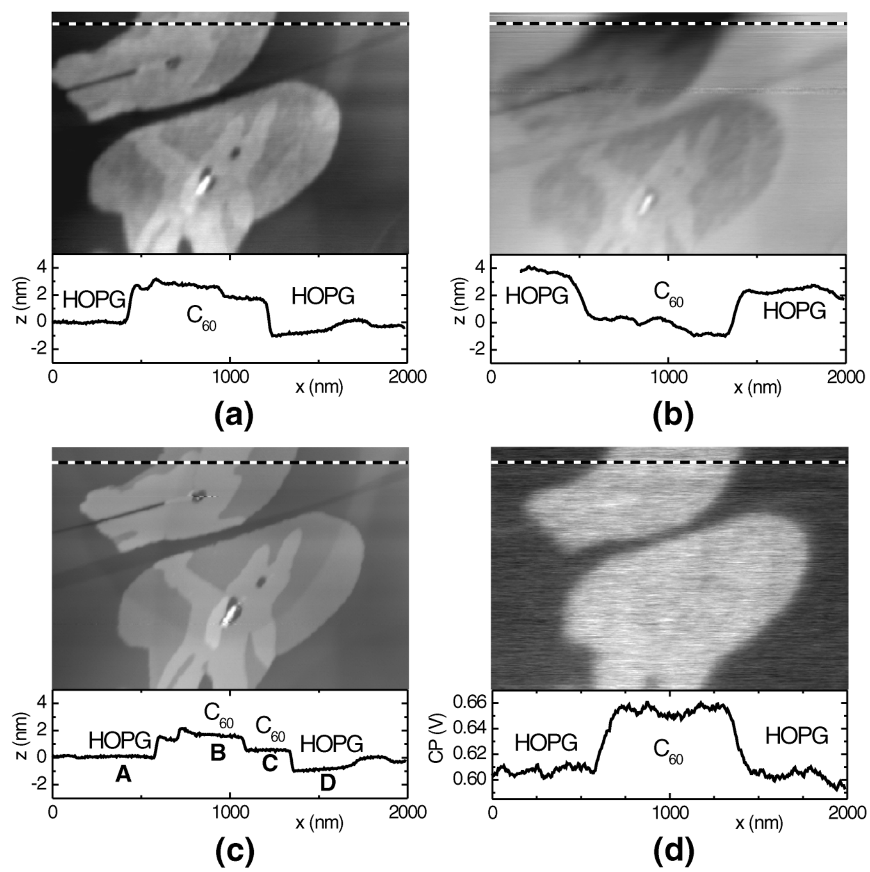

FIG. 1. NC-AFM measurement of C_{60} on HOPG. The topography is shown for $V_{\mathrm{bias}} = 0$ V (a) and $V_{\mathrm{bias}} = 1.34$ V (b). Topography (c) and simultaneously determined CP image (d) obtained by KPFM, with actively controlled V_{bias}. Corresponding line scans are shown below the images. All images are 2×1.35 μm measured at $\Delta f = -10$ Hz.

To examine the above described effect more closely, we studied in the following the topography for NC-AFM along the same line scan (see Fig. 1) as a function of the bias voltage V_{bias}. Figure 1(c) shows the topography along this line as determined in KPFM. Several plateaus for C_{60} and HOPG are labeled "A" through "D". In Fig. 2, we plot the step height between several of these plateaus (as indicated in the legend) as a function of V_{bias}. Exemplarily, three different cases are examined, a step from HOPG to HOPG (A–D), C_{60} to C_{60} (B–C) and C_{60} to HOPG (C–D). The HOPG-HOPG step A–D (open triangles) shows a step height of ~4.9 Å corresponding to 1 ML of HOPG [17], independent of V_{bias}. Also the C_{60}–C_{60} step (open squares) is independent of V_{bias}, showing a step height of ~9.2 Å in agreement with 1 ML of C_{60} [19,20]. However, the step from C_{60} to HOPG (C–D, open circles) shows a strong dependence on V_{bias} with measured step heights ranging from -1 nm to $+3.5$ nm. The step height determined from the KPFM measurement is indicated as the solid circle, giving a value of ~1.30 nm corresponding to 2 ML of C_{60} minus 1 ML of HOPG (a HOPG step is covered by the C_{60} island). This point was implemented in the figure at $V_{bias} = 0.63$ V, the mean contact potential between C_{60} and HOPG. As this point falls on the line through the bias dependent data, this shows that the correct height can be measured by NC-AFM only for an optimized bias voltage, set to the mean CP of the sample. Otherwise, the measured topography will be strongly influenced by electrostatic forces. However, we will show below that the bias can be optimized only for a sample with two different CP, i.e., consisting of two different materials.

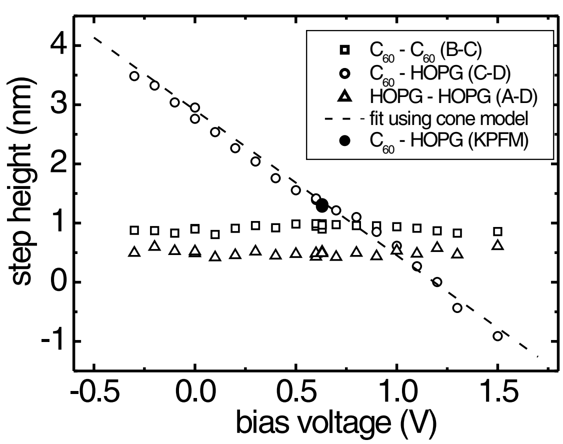

FIG. 2. Step height as a function of V_{bias} measured by NC-AFM. Plotted are steps from HOPG to HOPG (open squares), C_{60} to C_{60} (open triangles) and C_{60} to HOPG (open circles). The step height is determined between the plateaus "A" to "D" as indicated in the line scan of Fig. 1(c). The step C_{60} to HOPG shows a strong bias dependence. The dashed line is a fit describing the electrostatic force between the tip (modeled by a cone) and the surface. The solid circle represents the step height determined by KPFM. See text for details.

The slope of the bias dependence for the C_{60}-HOPG step (Fig. 2, open circles) contains information on the geometry of the tip. The electrostatic forces have been studied by different models for a variety of tip shapes [21–23]. Depending on the tip-sample distance, either the tip is more important (i.e., modeled by a sphere [21]), or the cantilever can be important [22]. The present data do not allow to favor one model over the other. Therefore, we adopt the model of a cone [22] in order to show that the slope of the step height from C_{60} to HOPG can be approximated using a reasonable tip shape (cone half opening angle $= 29°$) and a force gradient $\partial F/\partial z = 1 \times 10^{-12}$ N/nm. The determined cone half opening angle is in good agreement with the manufacturer's specifications, and the force gradient corresponds to a frequency shift $\Delta f = -10$ Hz, resonance frequency $f_0 = 60$ kHz and a spring constant $k = 3$ N/m, according to Eq. (1).

As mentioned above, the correct height can be measured if the sample bias is set to the mean CP. However, if a sample consists of several different materials with different CP, this is no longer valid as demonstrated in Fig. 3. In Fig. 3(a), the experimental situation from above is simulated using the same cone model [22]. A potential distribution as shown in the lower panel is assumed. The upper panel of Fig. 3(a) shows the correct topographic height for operation in Kelvin mode (open circles) and NC-AFM for three different bias voltages. It is clearly seen that the correct topography is also obtained if the bias voltage is set to the mean value (solid line). However, deviation from this voltage results in a change of the determined topographic profile. These results are in agreement with the experimental results shown in Fig. 1. In Fig. 3(b), a similar simulation is presented, however, this time using three "materials" with different contact potential (lower panel). Again, the upper panel gives the correct topographic profile obtained by KPFM (open circles) and NC-AFM under various applied biases. However, in this case the application of the mean contact potential does not result in a correct height measurement (solid line); the measured height in this case is 10% larger than the one determined by KPFM. This is due to residual electrostatic forces. In general, the effect of the electrostatic forces can be minimized by applying the mean CP computed as $V_{bias} = 1/n \sum_n V_{CP,n}$, where $V_{CP,n}$ is the CP of material component n of the sample. However, it has to be clear that the topography is subjected to residual electrostatic forces in this case, which results in a deviation from the correct height measurement.

In summary, we have shown that electrostatic forces have a strong impact on the topographic height profile measured in NC-AFM. Correct height measurement is possible for an optimized bias if the sample consists of only two different materials. To obtain a reliable height measurement in a sample with several different materials, the use of KPFM is mandatory, as with this technique the sample bias is controlled to match the CP at the present tip

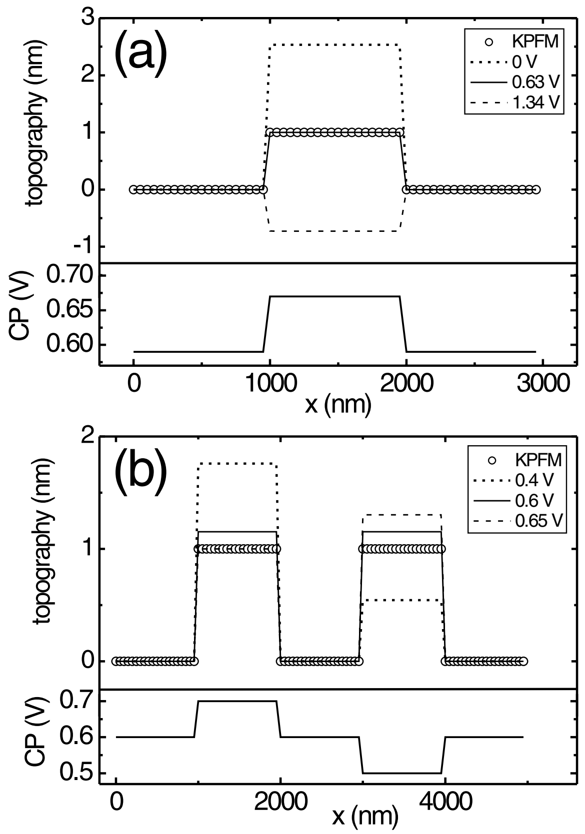

FIG. 3. The effect of electrostatic forces on the topography measurement in NC-AFM. (a) Simulation of two different materials (mimicking the experimental result of Fig. 1), where the CP is shown in the lower panel and the resulting topography for different V_{bias} in the upper panel. (b) Simulation of three different materials showing that for correct height measurements a KPFM method is mandatory. See text for details.

position. The described effect might also influence atomic resolution imaging by NC-AFM, as atomic scale variations of the CP have been reported recently [24].

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