Supplementary Information:
Torsional and lateral eigenmode oscillations for atomic resolution imaging of HOPG in air under ambient conditions

Anna L. Eichhorn[1] and Christian Dietz[1]*

[1]Physics of Surfaces, Institute of Materials Science, Technische Universität Darmstadt, Alarich-Weiss-Str. 2, 64287 Darmstadt, Germany

Email: eichhorn@pos.tu-darmstadt.de, dietz@pos.tu-darmstadt.de

Estimation of tip-trajectory from in-plane and out-of-plane deflection

The cantilever oscillation in out-of-plane (z) and in-plane (x) direction can be estimated by linear combinations of cosine functions including the amplitude setpoints A_i, the resonance frequencies $f_{0\,(i)}$, the time t and the phase Φ_{AM} of the oscillation used for the topographic feedback in amplitude modulation[1]. The static components z_0 and x_0 have been neglected for simplicity. For the calculations behind the figures of the tip-trajectories shown in the main text, equations (S1,S2) were used

$$z(t) = z_0 + z_{flex,2}(t) + z_{flex,3}(t) \approx A_{flex,2}\cos\left(2\pi f_{0\,(flex,2)}t - \Phi_{flex,2}\right) + A_{flex,3}\cos\left(2\pi f_{0\,(flex,3)}t - \frac{\pi}{2}\right), \quad \text{(S1)}$$

$$x(t) = x_0 + x_{tor}(t) + x_{lat}(t) \approx A_{tor}\cos\left(2\pi f_{0\,(tor)}t - \frac{\pi}{2}\right) + A_{lat}\cos\left(2\pi f_{0\,(lat)}t - \frac{\pi}{2}\right). \quad \text{(S2)}$$

Comparison of atomic resolution imaging on HOPG in the repulsive and the attractive regime

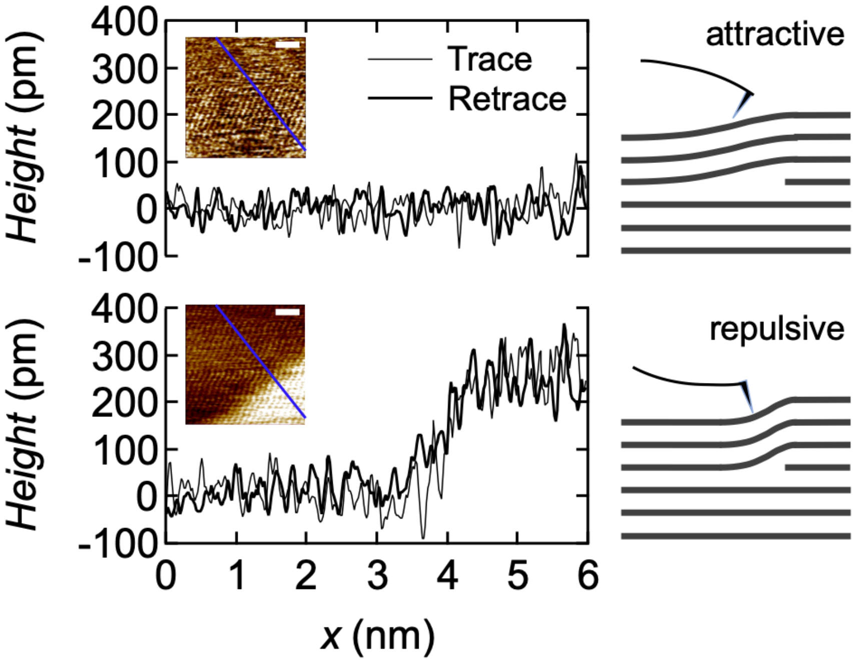

Figure S1: Cross sections through height images of HOPG at two different z-sensor positions and schemes of the alleged tip-sample interaction at the step edge in the attractive and the repulsive regime. The insets show the height images (retrace) of Figure 2j at Figure 2j at $z \approx 3.1$ nm (top) and Figure 2k at $z \approx 3.7$ nm (bottom). The position of the cross sections is marked in blue, scale bar: 1 nm. The thin black cross sections are drawn through the trace images (not shown) and the thicker cross sections through the retrace images at the same position. Scale bar: 400 pm.

Calibration of lateral oscillation amplitude inverse optical lever sensitivity (weighing factors)

In order to optimize the fit, we introduced appropriate weighing factors 1.25, 1.85 and 2.80. The smoothed height data was multiplied by and the smoothed lateral frequency-shift data was divided by these weighing factors, resulting in the black, blue and red dotted curves shown in Figure 3a and f, respectively. The wheighing factors $w_{lat,1}$ can be determined according to equation (1) which was found empirically

$$w_{lat,1} = 1.5^{\frac{A_{lat,1(large)}}{2A_{lat,1(small)}}}. \quad (1)$$

Comparison of tip trajectories in AMFlex2-FMLat1-FMFlex3 and AMFlex2-FMTor1-FMFlex3 mode

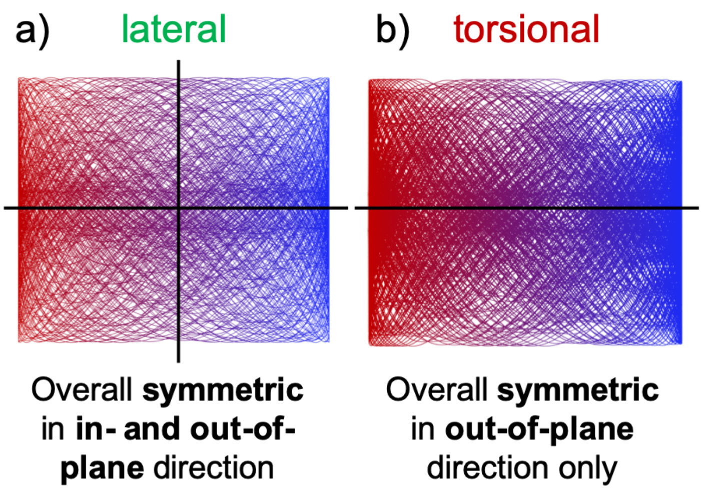

Figure S2: Comparison of the tip-trajectory resulting from (a) the AMFlex2-FMLat1-FMFlex3 mode and (b) the AMFlex2-FMTor1-FMFlex3 with $A_{flex,2}$ = 110 pm, $A_{flex,3}$ = 145 pm and $A_{lat,1} = A_{tor,1}$ = 328 pm.

Calibration of inverse optical lever sensitivities and force constants

The determination of the inverse optical lever sensitivities (invOLS) as well as the force constants of the flexural and the torsional eigenmodes were described in detail in our recent paper[2]. A method for the determination of the lateral invOLS was provided in the main text. The lateral force constant was calculated from the first flexural force constant, the width b and the thickness t of the cantilever[3]

$$k_{lat} = k_{flex,1}\left(\frac{b}{t}\right)^2. \qquad \text{(S3)}$$

References

1 Benaglia, S., Amo, C. A. & Garcia, R. Fast, quantitative and high resolution mapping of viscoelastic properties with bimodal AFM. *Nanoscale* **11**, 15289-15297, doi:10.1039/c9nr04396a (2019).

2 Eichhorn, A. L. & Dietz, C. Simultaneous Deconvolution of In-Plane and Out-of-Plane Forces of HOPG at the Atomic Scale under Ambient Conditions by Multifrequency Atomic Force Microscopy. *Advanced Materials Interfaces* **8**, doi:10.1002/admi.202101288 (2021).

3 Mullin, N. & Hobbs, J. K. A non-contact, thermal noise based method for the calibration of lateral deflection sensitivity in atomic force microscopy. *Review of Scientific Instruments* **85**, 113703, doi:10.1063/1.4901221 (2014).