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doi:10.1017/S1431927617011060
Microsc. Microanal. 23 (Suppl 1), 2017

Breaking the Time Barrier in Kelvin Probe Force Microscopy: Fast Free Force Reconstruction Using the G-Mode Platform

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The invention of the atomic force microscope (AFM)[1] effectively opened the doors to the nanoworld. The subsequent development of AFM techniques capable of simultaneously probing sample structure and material functionalities such as surface charge, electrochemical potentials, dielectric properties, made it possible to non-invasively characterize important material functionalities across relevant mesoscopic length (e.g. micron-atomic scale).[2] To date, the vast majority of applications of functional imaging using AFM focus on probing static or quasi-equilibrium (processes which are slow on time scale of the measurement itself (e.g. ms-hours)) processes. In this way, the spatial resolution and force sensitivity afforded by AFM techniques is counterpoise by the slow detection speeds compared to other common microscopy techniques (e.g. optical, scanning electron microscopy etc.).

Overcoming the temporal bottleneck is the next critical juncture in the development of AFM. We note that the temporal limitations of AFM are universally perceived to stem from (a) the mechanical bandwidth of the cantilever, (b) the nature of the heterodyne detection which requires averaging over multiple oscillationsl and (c) subsequent feedback operation. In this work, we overcome these restrictions by proposing and implementing an entirely new scheme of AFM imaging, referred to as fast free force recovery (FFFR or F^3R). F^3R allows direct reconstruction of the temporally resolved tip-sample forces from the noisy cantilever deflection signal with extremely high force sensitivity (pN) and fast temporal resolution (~μs). This approach is made possible by adopting the recently developed General acquisition mode (G-Mode) for AFM.[3] In this work we apply F^3R to the recovery of the electrostatic force in G-Mode Kelvin probe force microscopy (KPFM),[4-7] although this scheme is universally applicable for all SPMs, with stationary transfer functions, including KPFM, magnetic force microscopy (MFM), and other non-contact techniques. We demonstrate its usefulness for recovering the precise electrostatic force versus voltage dependence, ultimately allowing readout of electronic properties (e.g. electrochemical 'contact' potential difference (CPD) and capacitance information) having a temporal resolution (~10 μs) at least an order of magnitude higher than the state of the art. The F^3R methodology can be divided into three steps as outlined schematically in Figure 1. The first step involves capturing the photodetector response at very high sampling rates (4 MHz), with subsequent denoising and filtering.[3] Notice that unlike classical SPM methods where the filters are configured prior to measurement and stay constant in the process, in G-Mode SPM the multiple filtering routes can be applied to the stored signal, allowing significant flexibility and better information extraction. The second step involves removing the normalized cantilever transfer function from the detected photodetector response in the frequency domain. This requires knowledge of the full cantilever transfer function which can be measured as a calibration step. Once the transfer function of the cantilever is known, it can be used for the direct force recovery of G-mode data in Fourier space. The final step involves performing an inverse Fourier of the cantilever trajectory with the adapting filtering including only the frequency bins of interest corresponding to the tip-sample force. We will show that the F^3R approach overcomes the widely viewed time resolution bottleneck of AFM, i.e. the mechanical bandwidth of the cantilever, without the need for heterodyne detection or averaging of the signal, and hence preserving the tip

trajectory in space and time. As a first application we leverage the high potential sensitivity and fast readout for the detection of light induced surface photo-voltage in perovskite solar cell materials. As a model system we choose a single crystal of methylammonium lead iodide perovskite ($MAPbI_3$). We expect this approach to be valid for all modes of AFM operation and will be useful for probing kinetics and dynamics on the nanoscale [8].

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[8] This manuscript has been authored by UT Battelle, LLC, under Contract No. DE-AC0500OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript, or allow others to do so, for the United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).

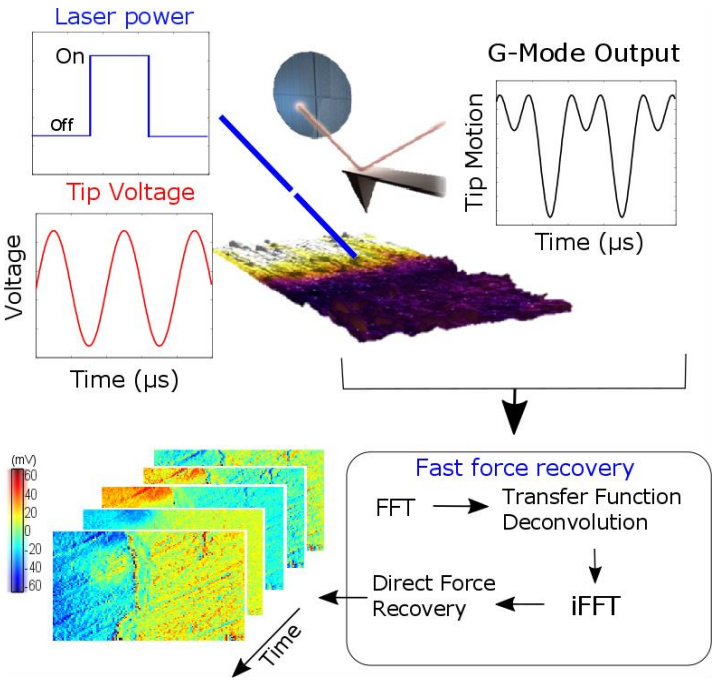

Figure 1. Schematic showing the principles behind G-KPFM with fast free force reconstruction.

https://doi.org/10.1017/S1431927617011060 Published online by Cambridge University Press