

What limits time resolution in AFM?

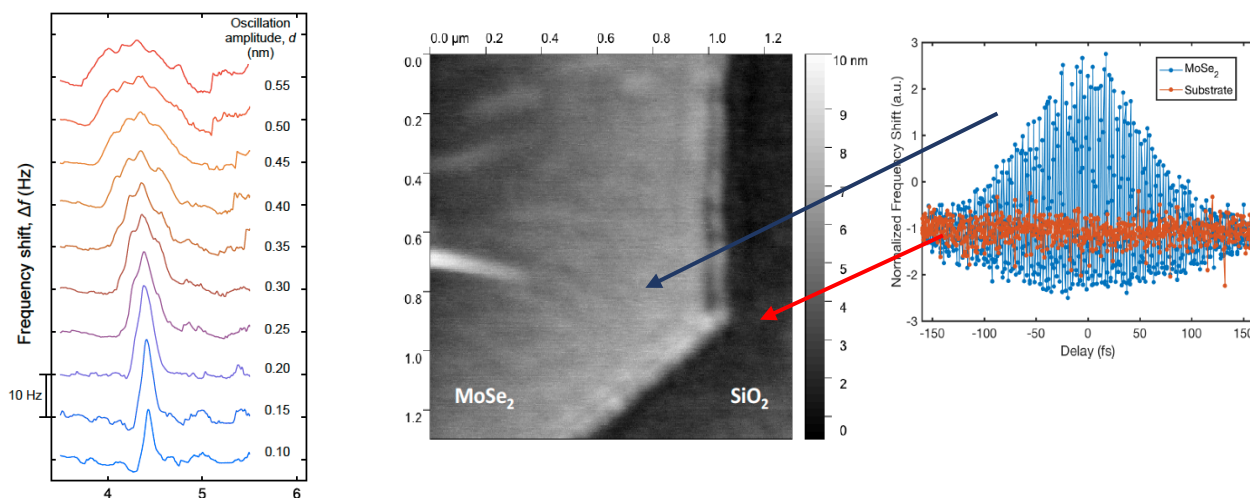
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Developing a technique that combines nanometer spatial and femtosecond temporal sensitivity is a crucial step towards exposing the inner mechanisms of chemical reactions, single molecule motion, electron dynamics in solids, and the effects of defects or trap states on electron motion and behavior, amongst a whole slew of other questions relating to the most fundamental processes in molecular systems.

In this presentation, I will give an overview of our recent successes at characterizing surface potentials using AFM-based techniques on time scales down to fs. The high spatial resolution of AFM in principle then allows the identification of rate limiting structures/defects, allowing the fundamentally important correlation of structure and processing with properties. Specifically, I will show how one can mechanically measure a change in the sample response as a result of a delay time as short as 25 atto seconds between a pump and probe stimulation pulse on systems such as LiNbO₃ or monolayers of MoSe₂. The realization of this ultrafast AFM opens the door to understanding ultrafast electron dynamics on surfaces, such as the new territory of atomic scale dynamics of polarization.

As a second example, I will discuss our most recent results on single electron transfer events at a single molecule-metallic electrode interface. Our results can be quantitatively understood by the transport of electrons between electrodes, suppressed by the Frank-Condon effect due to strong electron-phonon coupling (or molecular reorganization, in the language of redox chemistry). Our experiments are equivalent to a single molecule redox reaction in the language of chemistry – our experiments thus bridge single molecular charge transport with chemical rate theory (i.e. Marcus Theory).



Left image: Frequency shift spectra, with increasing AFM oscillation amplitude (i.e. coupling strength), taken 10 nm above a single ferrocene molecule at 4.8 K. From this data one can directly deduce (without assumptions) the molecular reorganization energy, nuclear-electron coupling constants and molecular vibration frequencies.