Ultrafast Time-Resolved Atomic Force Microscopy for Probing Photoexcited Dynamics

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Atomic force microscopy (AFM) probes a wide range of interactions at surfaces and interfaces on the nanoscale. Extending AFM to ultrafast timescales can reveal how local structure and electronic properties influence nonequilibrium dynamics. In optical pump–probe AFM, excited-state dynamics are reconstructed from time-averaged force measurements by varying the delay between two optical pulses. This approach resolves dynamics far faster than the sensor's mechanical response. Previous studies have combined frequency modulation AFM (FM-AFM) with femtosecond laser pulses to measure picosecond carrier relaxation [1]. However, modulating the optical intensity can introduce thermal expansion artifacts, particularly at high excitation intensities. This motivates a detection scheme that keeps the average optical power constant.

We developed time-resolved force microscopy by combining tuning-fork-based FM-AFM with electrically controlled optical pump–probe excitation and delay-time modulation [2]. The pulse delay is periodically switched between a measurement delay and a reference delay without changing the average optical power. Lock-in detection of the corresponding change in resonance frequency isolates the delay-dependent force response while suppressing thermal expansion artifacts. The optical system uses 45 ps pulses, enabling measurements on picosecond timescales. Electrical readout of the quartz tuning fork eliminates the need for a separate detection laser and facilitates optical excitation.

Measurements on bulk WSe_2 yield a stable time-resolved force signal with two decay components associated with surface recombination and carrier diffusion (Fig. 1). Force spectroscopy and a model based on photoinduced dipole–dipole interactions help elucidate the origin of the signal. These results extend AFM from static characterization to local measurements of photoexcited dynamics. Applying this approach to different AFM modes offers opportunities to investigate transient potentials, polarization, and carrier relaxation in materials and devices. In this talk, I will also present recent efforts to advance time-resolved AFM.

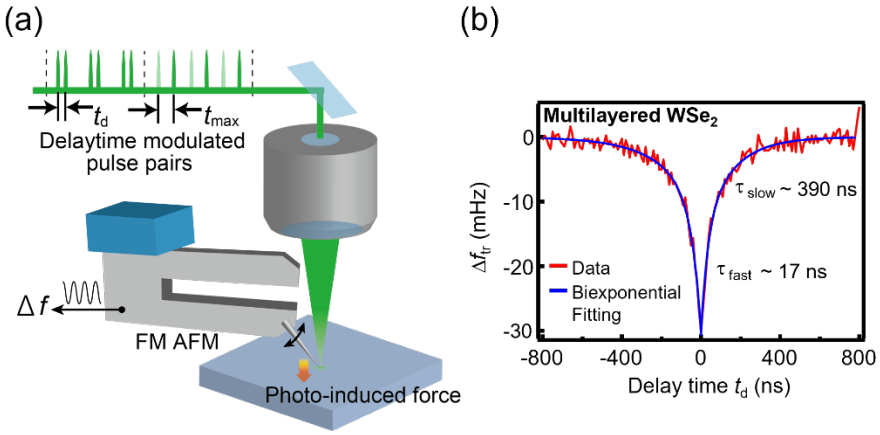


Figure 1. (a) Time-resolved FM-AFM setup and (b) time-resolved force signal measured on multilayer WSe_2 with a biexponential fit (right). Reproduced from Fig. 4(b) and (c) of Ref. [3].

References

[1] Z. Schumacher et al., Appl. Phys. Lett. 110, 053111 (2017).
[2] H. Mogi et al., Appl. Phys. Express 17, 015003 (2024).
[3] K. Iwaya et al., Imaging & Microscopy 1/2024, 34–36 (2024).