

## Time-resolved TEM beyond fast detectors

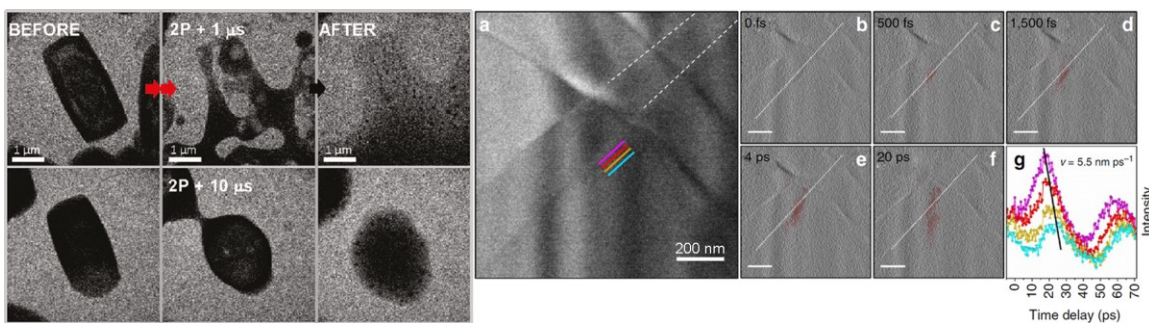
D. Flannigan, J. Chen, W. Curtis, D. Du, P. Engen, E. VandenBussche, Y. Zhang

Department of Chemical Engineering and Materials Science, University of Minnesota, 421 Washington Avenue SE, Minneapolis, Minnesota 55455, United States of America

flan0076@umn.edu

Timescales of dynamic processes in extended solids span many orders of magnitude owing to the large number of degrees of freedom. Additionally, scaling laws dictate that discrete temporal domains comprising the entire continuum consist of associated spatial domains within which specific dynamics are dominant. Ideally, one would be able to probe the entire spatiotemporal range on a single specimen spot with a single instrument. Modern TEMs are exceptionally versatile in this regard, providing access to spatial and energy ranges that span sub-Å to micrometres and sub-10 meV to 1,000s of eV, respectively. However, timescales of the associated physical phenomena span 100s of attoseconds ( $10^{-18}$  s) to minutes and longer, a range that cannot be fully covered by even the fastest direct detectors. Indeed, dynamics faster than  $\sim 0.1$  ms are largely inaccessible to detector-based TEM approaches.

Here, an overview will be provided of ongoing efforts aimed at pushing TEM temporal resolutions well beyond the limits imposed by detectors and by peak dose rates. Emphasis will be placed on laser-driven nanosecond single-shot and fs stroboscopic modalities, currently the two most widely used approaches (Figure 1) [1-3]. Common hardware configurations based on modified commercial TEM platforms will be described, and current state-of-the-art performance specifications will be discussed. This will be followed by a brief survey of discoveries and advances that have been made with imaging, diffraction, and spectroscopy. Particular focus will be placed on experiments that have led to deeper understanding of materials and to new physics [4]. The talk will conclude with a brief look toward new emerging approaches and expanded applications, such as pulsed-beam damage mitigation [5, 6].



**Figure 1.** Nanosecond and fs TEM. Panels on the left are 10-ns single-shot images of a photoinduced redox reaction [1]. Panels on the right (a-g) are 300-fs stroboscopic images of photoexcited phonon launch from a crystal step edge [2].

[1] Park, S. T., Flannigan, D. J. & Zewail, A. H. (2011). *J. Am. Chem. Soc.* 133, 1730.

[2] Cremons, D. R., Plemmons, D. A. & Flannigan, D. J. (2016). *Nat. Commun.* 7, 11230.

[3] Plemmons, D. A., Suri, P. K. & Flannigan, D. J. (2015). *Chem. Mater.* 27, 3178.

[4] Barwick, B., Flannigan, D. J. & Zewail, A. H. (2009). *Nature* 462, 902.

[5] VandenBussche, E. J. & Flannigan, D. J. (2019). *Nano Lett.* 19, 6687.

[6] VandenBussche, E. J., Clark, C. P., Holmes, R. J. & Flannigan, D. J. (2020). *ACS Omega* 5, 31867.

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