

Ultrafast AFM with Femtosecond Time Resolution for Study of Nonlinear Optical Metamaterials and 2D π -Conjugated Polymer Excitation Dynamics

D. Jacob^{1, #}, W. Behn¹, E. Ulyanov¹, D. Perepichka², and P. Grutter¹

¹*Physics Department, McGill University, Montreal, H3A 2T8, Canada*

²*Chemistry Department, McGill University, Montreal, H3A 3E8, Canada*

Presenting author's e-mail: daniel.c.jacob@mail.mcgill.ca

Ultrafast AFM is an experimental technique that utilizes a femtosecond pulsed laser directed onto a sample being measured by non-contact AFM to observe nonlinear effects, and measure excitation and relaxation timescales, with nanometer spatial resolution and femtosecond time resolution. Nonlinear metamaterials are an attractive alternative to current methods for second harmonic generation and associated phenomena due to their high Electric Field Induced Second Harmonic Generation (EFISHG) which allows for the conversion of the material's third order nonlinear susceptibility ($\chi^{(3)}$) into second order susceptibility ($\chi^{(2)}$) on application of an external bias [1–3]. Nonlinear effects have previously been detected by AFM on Lithium Niobate, and an investigation of nonlinear optical metamaterials at the nanoscale may allow for further EFISHG optimization [4]. The time and spatial resolution offered by Ultrafast scanning probe techniques also allow for measurement of carrier lifetimes in materials that exhibit optical excitations, this has previously been utilized for characterization of carrier lifetime in photovoltaics, nanoscale vibrational spectroscopy, and measuring plasmonic excitation timescales [5–8]. 2D π -Conjugated Polymers (2DCPS) are 2D films that are attractive for applications in thin film electronics, and azotriangulene monomer units are in particular attractive due to their predicted semiconducting nature, single band gap, and planar rigidity to enhance π -conjugation [9–11]. This AFM technique can be extended to the study of 2D π -conjugated homopolymers and copolymers with band gaps allowing for better information to further tune polymer films for optoelectronic applications.

References

- [1] W. Cai, A. P. Vasudev, and M. L. Brongersma, *Science* **333**, 1720 (2011).
- [2] L. Kang, Y. Cui, S. Lan, S. P. Rodrigues, M. L. Brongersma, and W. Cai, *Nat. Commun.* **5**, 4680 (2014).
- [3] Y. Zhao, Y. Yang, and H.-B. Sun, *Photonix* **2**, 3 (2021).
- [4] Z. Schumacher, R. Rejali, R. Pachlatko, A. Spielhofer, P. Nagler, Y. Miyahara, D. G. Cooke, and P. Grütter, *Proc. Natl. Acad. Sci.* **117**, 19773 (2020).
- [5] Z. Schumacher, Y. Miyahara, A. Spielhofer, and P. Grutter, *Phys. Rev. Appl.* **5**, 044018 (2016).
- [6] M. Takihara, T. Takahashi, and T. Ujihara, *Appl. Phys. Lett.* **93**, 021902 (2008).
- [7] R. Gutzler, M. Garg, C. R. Ast, K. Kuhnke, and K. Kern, *Nat. Rev. Phys.* **3**, 441 (2021).
- [8] M. Garg and K. Kern, *Science* **367**, 411 (2020).
- [9] G. Galeotti et al., *Nat. Mater.* **19**, 874 (2020).
- [10] Y. Jing and T. Heine, *J. Am. Chem. Soc.* **141**, 743 (2019).
- [11] P. Agarwala and D. Kabra, *J. Mater. Chem. A* **5**, 1348 (2017).