Tabletop Optical Probes of MHz-GHz Longitudinal and Transverse Acoustic Waves in Glass-Forming Liquids

K. A. NELSON

Department of Chemistry, MIT, **kanelson@mit.edu**

Fundamental understanding of structural relaxation dynamics in glass-forming liquids continues to pose difficult challenges, in large part because of the experimental difficulty in accessing the extraordinary range of time scales over which relaxation occurs. For much of this range, shear and compressional dynamics are manifest in the corresponding acoustic properties. However, experimental measurements of longitudinal and transverse acoustic waves across their full frequency ranges are challenging for a variety of reasons including strong temperature-dependent acoustic damping and wavevector mismatch between light at optical frequencies and acoustic waves at both low and high frequencies.

We have developed two complementary time-domain spectroscopic approaches for tabletop optical characterization of longitudinal and transverse acoustic waves across much of their frequency ranges. The first uses a time-resolved four-wave mixing or “transient grating” geometry in which crossed excitation light pulses excite acoustic waves through impulsive stimulated Brillouin scattering (ISBS) or through mild heating (“impulsive stimulated thermal scattering” or ISTS). [1] In the former case, parallel or perpendicular light polarizations may be used to drive and monitor longitudinal or transverse acoustic waves respectively. In the latter case, slow non-oscillatory components of structural relaxation dynamics (the time-domain analog of the Mountain mode in quasi-elastic light scattering) as well as longitudinal acoustic responses are observed. This approach provides access to acoustic waves in roughly the 10 MHz – 10 GHz frequency range, and the slower non-oscillatory responses may extend out to almost 1 ms. The second approach is a variation of picosecond ultrasonics [2] in which a timed pulse sequence is used to irradiate a metal film and generate a several-cycle acoustic wave that propagates into an underlying sample layer and is detected at the other side. [3] This permits frequency-resolved measurements of acoustic waves up to several hundred GHz frequencies. Extension of this method to about 1 THz frequency may be possible. [4] Using a crystallographically canted iron layer, transverse as well as longitudinal waves can be launched. [5] Results from both approaches and their use to test models of supercooled liquid behaviour will be discussed.

References

[1] - Y. Yang and K.A. Nelson, J. Chem. Phys. **103**, 7722, 7732, (1995).

[2] - C. Thomsen, H. T. Grahn, H. J. Maris, J. Tauc, Phys.Rev. B **34**, 4129, (1986).

[3] - J.D. Choi, T. Feurer, M. Yamaguchi, B.Paxton, and K.A. Nelson, Appl. Phys. Lett. **87**, 081907, (2005).

[4] - C.-T. Yu, K.-H. Lin, C.-L. Hsieh, C.-C. Pan, J.-I. Chyi, and C.-K. Sun, Appl. Phys. Lett. **87**, 093114, (2005).

[5] - P. Frechard, S. Andrieu, D. Chateigner, M. Hallouis, P. Germi, and M. Pernet, Thin Solid Films **263**, 42, (1995).