High Sensitivity Ultrafast THz Studies of Materials

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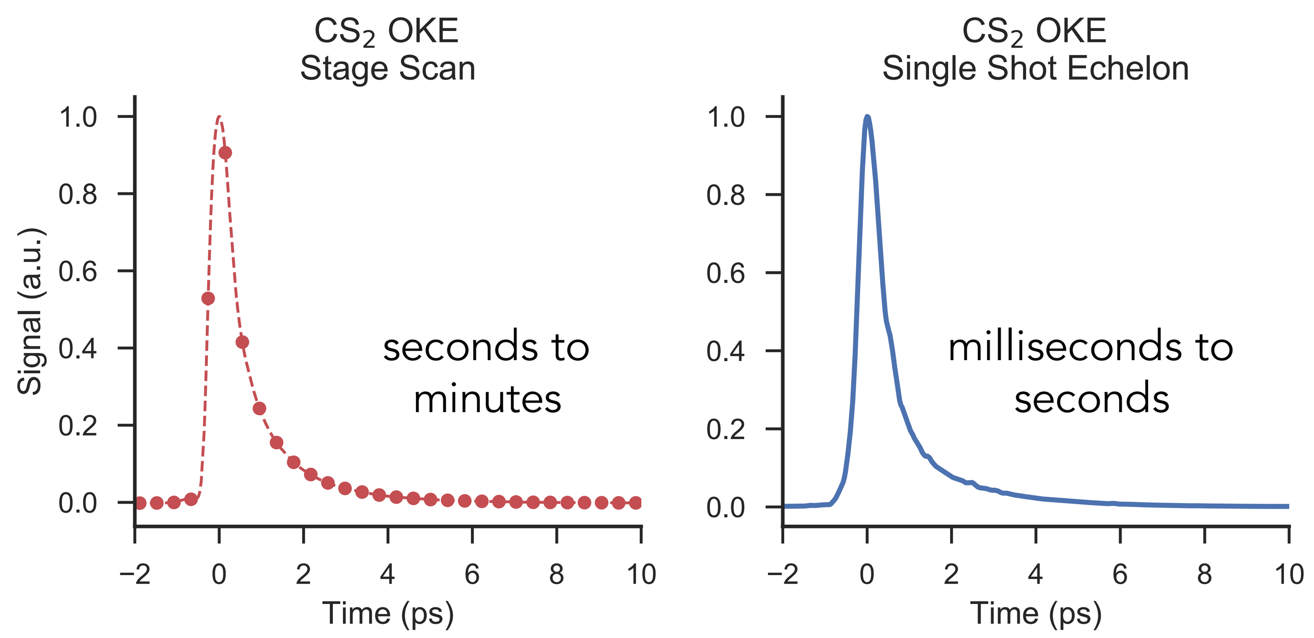
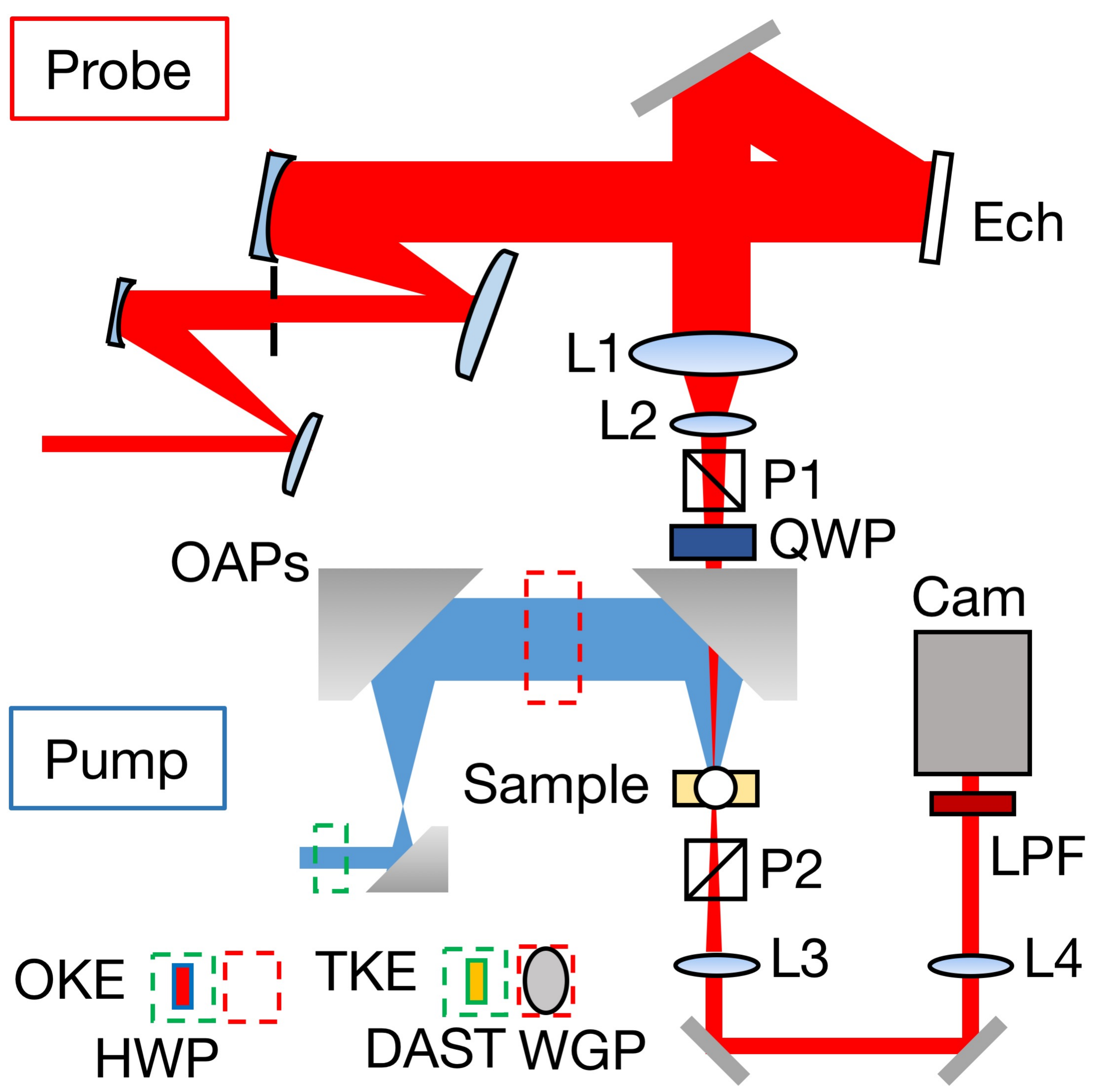
Speeding up ultrafast nonlinear spectroscopies of materials

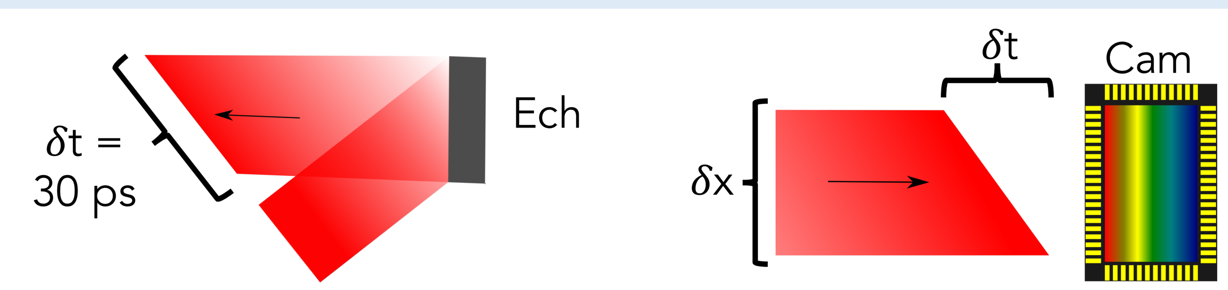


While offering unique insights into fundamental physical properties of matter, many ultrafast nonlinear optical and terahertz signals are too brief to directly measure. Instead, motorized stages change the relative delay time (distance) between a pump beam and a probe beam. An ultrafast signal can then be indirectly measured by stepping along many stage positions. This process can be quite slow (seconds to minutes). A single shot approach captures the same information much faster, often in milliseconds.

The single shot Kerr effect spectrometer

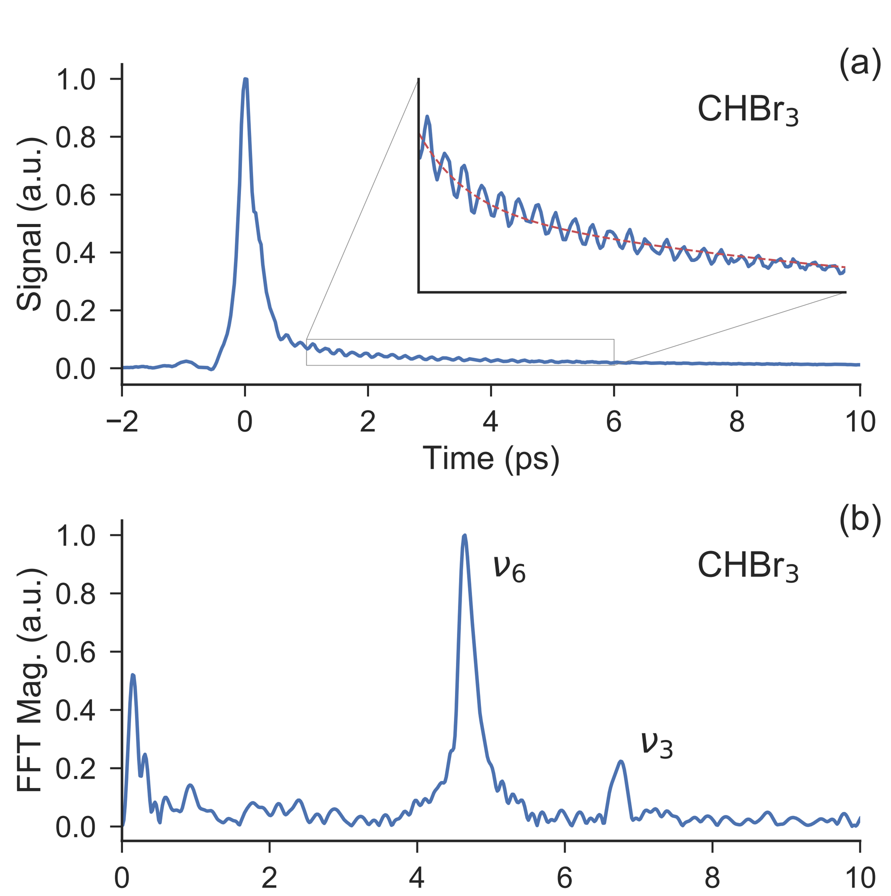


The spectrometer (left) is designed to measure the Optical and Terahertz Kerr Effect (OKE/TKE) responses of liquids. Intense optical/terahertz pump fields induce a picosecond transient birefringence in the sample. This transient birefringence rotates the polarization of a probe beam, and this rotation is detected by the camera. Green and red dashed squares indicate specific optics required for OKE and TKE measurements. Small modifications also allow for electro-optically sampled THz measurements.

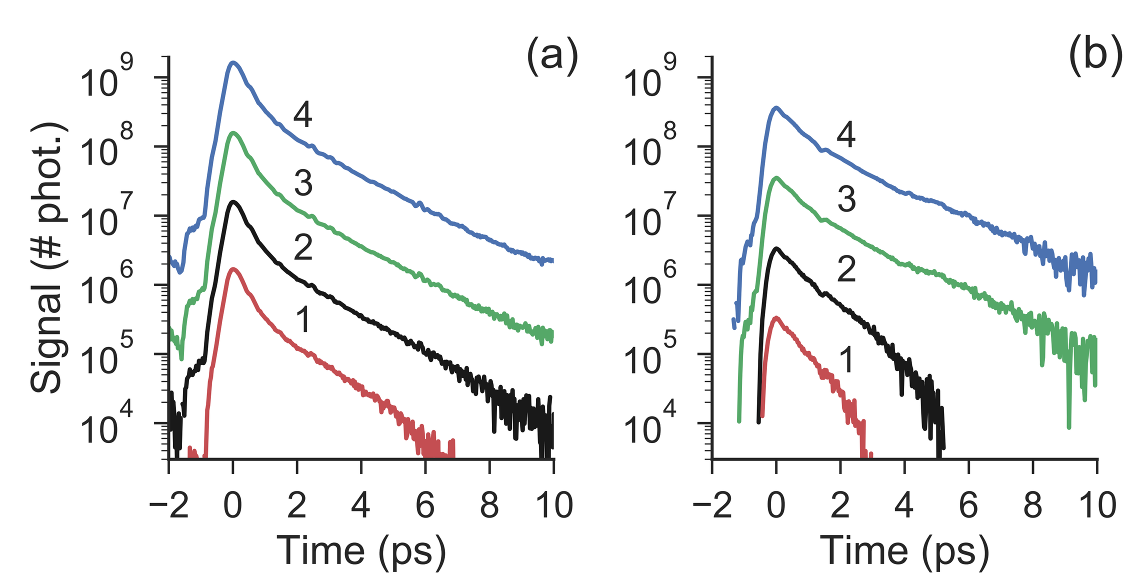


The two details (above) illustrate the generation and detection of a 30 ps duration probe pulse front that maps a nonlinear signal onto the camera's CMOS array.

Kerr effect spectroscopy of liquid dynamics



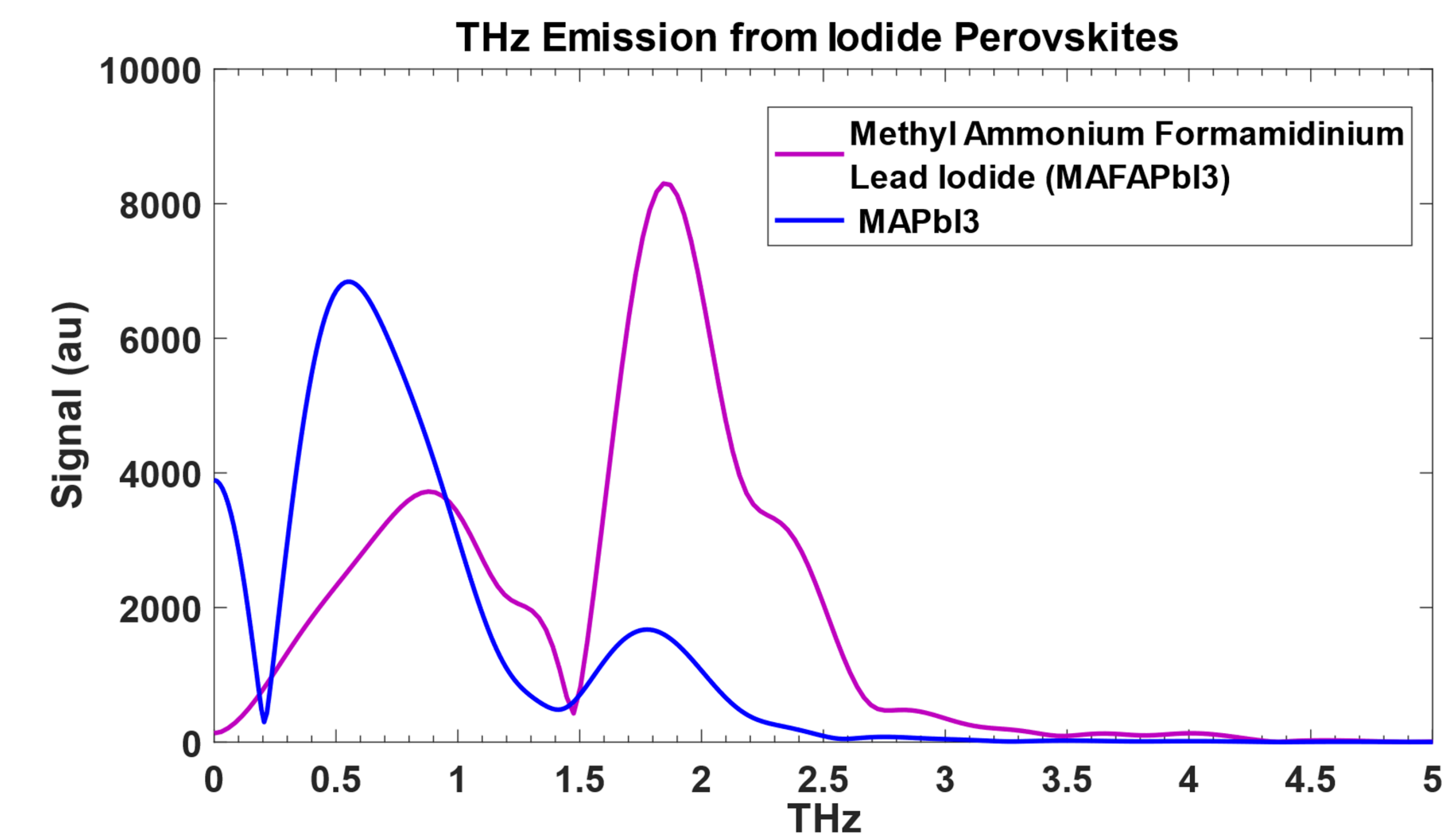
Bromoform, like many liquids, has THz-active intramolecular vibrational modes. Single shot OKE measurements (left, (a)) are sensitive enough to detect nonlinear molecular coherences (inset). The data's FFT (b) confirms the presence of two of bromoform's vibrational modes at 4.66 and 6.73 THz.

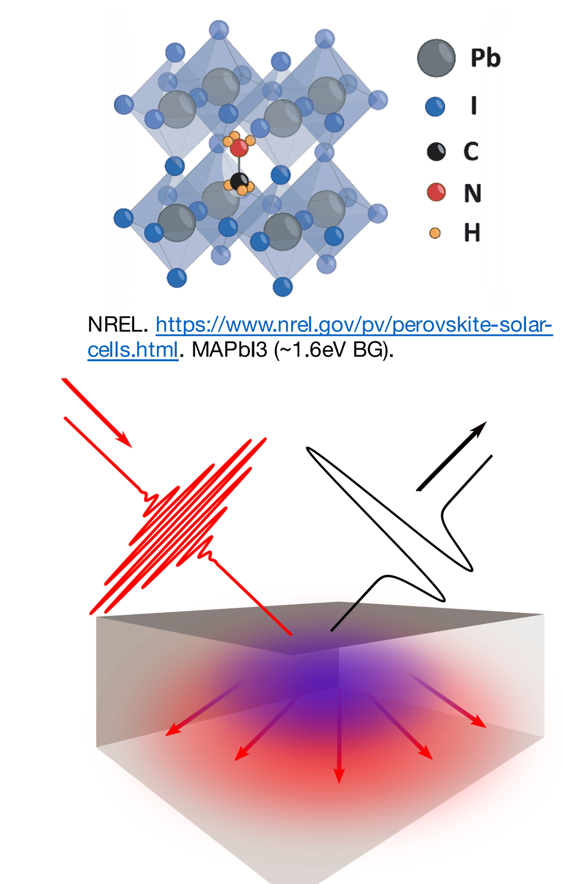


The signal scaling's of single-shot OKE (a) and TKE (b) measurements of CS_2, where $N_{shots} = 10^x$ (x = 1, 2, 3, 4).

Ultrafast THz spectroscopy of organohalide perovskites

Organohalide perovskites (OHPs) are promising candidates for optoelectronics, in particular photovoltaics. Understanding ultrafast carrier dynamics is critical for engineering higher-efficiency and more stable perovskite structures. Using a single shot approach to measure ultrafast charge carrier dynamics with electro-optic THz sampling may facilitate time-resolved studies of perovskite degradation and multidimensional studies of carrier-phonon couplings.





OHP THz emission spectroscopy

THz emission from OHPs (above) is thought to arise from the photo-Dember effect, where asymmetric electron-hole diffusion creates a transient oscillating dipole. Investigating this effect will provide a unique probe of carrier mobilities and dynamics.

Conclusions

The single shot echelon technique is a viable alternative to the conventional stage scan method. With millisecond-scale acquisition times for electro-optic THz measurements, we are interested in applying this approach to study time-resolved perovskite carrier dynamics and degradation pathways. Further efforts include applying single shot techniques to multidimensional spectroscopies (e.g. 2D TTR of liquids and perovskites), and investigating cheaper cameras and other cost-savings to spur adoption of the technique.

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