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X-ray Transient Grating & 2D Spectroscopy

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There are a great many motivations for extending nonlinear wave mixing experiments to the x-ray spectral range. Many are based on high x-ray wavevectors which enable transient grating (TG) measurements with cor-respondingly high wave wavevector magnitude q, i.e. extremely short spatial periods $\Lambda = 2\pi/q$ formed by crossed x-ray beams. We have conducted EUV TG experiments in collaboration with the FERMI team at the TIMER beamline [1,2]. The signals reveal surface and bulk acoustic waves with wavelengths of tens of na-nometers (equal to Λ) and thermal transport from the heated TG peaks toward the unheated nulls across dis-tances given roughly by $1/q$. Nanoscale thermal transport in insulators shows highly non-diffusive kinetics since most of the phonons that carry heat have nanometer mean free paths. Thermal transport may approach the ballistic limit at sufficiently short TG periods. We have also conducted experiments in which transient magneti-zation gratings are formed. We anticipate measurements with TG periods at or near those of modulated phases such as a charge-density or spin-density wave systems, allowing excitation of the characteristic modes in those phases.

Hard x-ray TG measurements using Talbot imaging of etched periodic patterns [3] are now possible. We conducted preliminary experiments at the SwissFEL BERNINA beamline, led by our conference organizer, with x-ray excitation and optical probing. All-x-ray experiments are anticipated, allowing TG measurements deep with-in bulk samples. Phase-coherent 2D x-ray spectroscopy will also be possible. Even without reaching the highest wavevectors, measurements on molecular electronic transitions will reveal new information that cannot be ob-tained using optical wavelengths [4]. 2D x-ray spectroscopy of nuclear transitions should also be possible. Angstrom TG periods and coherent excitation of collective modes at zone-boundary wavevectors may become possible.

Hybrid forms of 2D spectroscopy can be conducted using colinear low-frequency pulse pairs or pulse sequences and x-ray probes. This is 2D optical, IR, or THz spectroscopy with signals measured as x-ray diffraction (XRD), absorption, or induced emission which may reveal key information about the structural, electronic, or magnetic effects of phase-related excitation pulses at the lower frequency. Hybrid 2D spectroscopy of quantum phases may reveal the roles played by coupled degrees of freedom in excursions across the multiphase landscape in particularly incisive ways.

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