Helmholtz International Fellowship Report

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May 2 – June 30, 2017

I spent an exciting two-month period between May 2 and June 30, 2017 visiting CFEL/DESY Hamburg and being hosted by Robin Santra. On the day that I arrived the EuXFEL lased for the first time, and, on the last day of my stay, June 30, the EuXFEL was officially declared to be in operation by the Management Board. Indeed, I received an email from my SLAC accelerator physicist colleague, who incidentally was spending the same period at DESY, announcing the news of lasing. On June 29, as a member of the EuXFEL Scientific Advisory Committee (SAC), I accompanied the SAC Chair on a tour to verify readiness of the two end stations (FXE and SPB) proposed to be operational for the first run. Sandwiched between these two endpoints, I interacted fruitfully both with theory (Santra and Rohringer) and experimental groups, lectured in the UXSS 2017 Summer School and participated in the DESY Science Council meeting.

Interactions with Robin Santra's theory group were extremely productive on two separate fronts. First, we were working on the interpretation of results of an experiment designed to study inner-shell hole delocalization in ethyl trifluoroacetate, the iconic ESCA molecule. This molecule was chosen because there are four environmentally distinct carbon sites that are easily distinguishable by experiment. The question was whether the fragmentation that occurs after x-ray photoabsorption is site-selective and whether the degree of selectivity can be predicted. Given that photoabsorption is the dominant x-ray interaction mechanism, the importance of understanding the energy relaxation mechanism in molecules is paramount. Overall, the experiment showed a lack of site selectivity and an interesting dominant fragmentation mechanism: ion ejection from the ends of the molecule leaving a stationary neutral carbon dioxide molecule. With the inventiveness of Ludger Inhester, a novel and computationally "less expensive" methodology was developed to view these processes that predicted this mechanism and also gave the chemical insight that the weakest bonds in the molecule are further weakened, leading to dissociation along these channels. This work has now been published in Journal of Physical Chemistry Letters. Second, we furthered our collaboration on the dynamics and coherence in strong-field ionized water – as probed by resonant x-ray absorption in the water window. Here the interest is in understanding the long-lived electronic coherence in the condensed phase that was previously observed with optical probes of the hydrated electron. The x-rays provide a new window on the residual ion that is not available in the optical regime and an experiment is scheduled for May 2018 at LCLS.

A collaboration with Nina Rohringer was initiated to understand propagation of high-intensity resonant x-ray radiation in optically dense samples. Propagation effects on resonance at high x-ray intensity can lead to a number of interesting phenomena, e.g. self-induced transparency, and are of considerable importance for transient absorption spectroscopies at XFELs. Here interactions with Jens Viefhaus (at the time at DESY, now at BESSY) and Adrian Cavalieri may lead to experimental designs to detect these important phenomena in the time domain.

Finally, interactions with Henry Chapman and John Spence (visiting Helmholtz fellow) led to a new awareness and interest in the area of incoherent imaging as a method to enhance coherent diffractive imaging as a structural determination tool.