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Tabletop time-resolved soft x-ray spectrometer

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Based on this DURIP grant, we purchased the necessary laser systems and other components (optics, vacuum components and optical mechanics), as well as a few customized-designed pieces to construct a table top time-resolved EUV (soft x-ray) spectrometer. We have successfully constructed a compact, ultrafast UV pump and EUV probe spectrometer, based on a fiber-based high-harmonic generation (HHG) based EUV/Soft X-ray beamline. Using this setup, we have obtained static absorption of -Fe2O3 thin film, reproduced literature reported transient absorption to track electron dynamics and polaron formation at Fe element in -Fe2O3. Recently, we also studied another different form of Fe2O3, which shows electronic dynamics that favors charge recombination. We are currently finalizing this study by characterizing structural difference between the two types of Fe2O3 and understand why one favors polariton formation whereas the other one facilitates charge recombination. In addition, a novel 2D UV-EUV spectroscopy to disentangle heterogeneity in solid state charge dynamics, contributing to my other AFOSR related project.

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Tabletop time-resolved soft x-ray spectrometer

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Final Performance Report

Based on this DURIP grant, we purchased the necessary laser systems and other components (optics, vacuum components and optical mechanics), as well as a few customized-designed pieces to construct a table top time-resolved EUV (soft x-ray) spectrometer. We have successfully constructed a compact, ultrafast UV pump and EUV probe spectrometer, based on a fiber-based high-harmonic generation (HHG) based EUV/Soft X-ray beamline. Using this setup, we have obtained static absorption of α-Fe_2O_3 thin film, reproduced literature reported transient absorption to track electron dynamics and polaron formation at Fe element in α-Fe_2O_3. Recently, we also studied another different form of Fe_2O_3, which shows electronic dynamics that favors charge recombination. We are currently finalizing this study by characterizing structural difference between the two types of Fe_2O_3 and understand why one favors polariton formation whereas the other one facilitates charge recombination. In addition, a novel 2D UV-EUV spectroscopy to disentangle heterogeneity in solid state charge dynamics, contributing to my other AFOSR related project.

Time-Resolved EUV/X-ray spectrometer and ultrafast dynamics of electrons of Fe_2O_3.

The time-resolved EUV spectrometer is shown in Fig.1a. We implement a compact design. From light source generation to spectrograph and CCD detector, the setup is only 2.5 m long and less 1 m wide. The driving lasers (Ti:sapphire, Coherent) couples to the hollow waveguide, which is filled with media gas for HHG (Figure 1b). The optical fiber effectively restrained unnecessary fast gas flow and therefore reduce the load for vacuum pump, which is a key for the compact EUV spectrometer. The generated HHG is then focused by a toroidal mirror at glancing angle (Figure 1c), and then focused onto the surface sample at glancing angle as well (Figure 1d). The HHG interacts with the sample with reflective geometry. The reflected HHG beam is then dispersed by a curved grating and then dispersed onto the EUV CCD (Figure 1e). The optical pump beam is also generated from 800 nm based of second harmonic generation (using BBO crystal, 130uJ) or nonlinear optical parametric amplification. It is focused into the sample at a shallow angle, to increase mode matching with the HHG beam. To ensure pump and HHG probe beam overlap, we home built an optical imaging setup to image the sample surface (Fig.2). We then first replace the sample with a phosphor screen, which can fluoresce under the EUV excitation. Then we will overlap the pump scatter on top of the EUV fluorescence to ensure the spatial overlap. The beam size of pump is 1mm*5mm and the beam size of EUV is 100um*1mm.

Using this setup, we first investigated α-Fe_2O_3 sample. The sample is home-fabricated and reach to high surface quality for EUV reflection (Fig.3a). The comparison between HHG beam reflected from Fe_2O_3 surface and SiO_2 surface clearly show a dip around 54 eV, which is originated from the absorption of Fe M edge (core level (3p) to 3d), agreeing with literature well. We then performed optical pump EUV probe experiment on the Fe_2O_3 to follow electron dynamics at Fe M edge. The raw data is shown in Fig.3c, whereas the reconstructed spectral dynamics is in Fig.3d. The pseudo color plot's z axis is ΔOD, in which we can clearly resolve a positive peak near 53 eV, and a small negative peak at 55 eV, due to polaron formation. By taking fine time step scans and taking a cut near 55 eV, we observed a positive peak close to t=0, e.g. UV pump and EUV probe are temporally overlapped or close to each other, which also decays fast with a lifetime of 600 fs (Figure.3e). This early time dynamics has been assigned to initial electron-hole pair generation in Fe_2O_3, which quickly stabilized with surrounding lattice to form polarons, which has a very long lifetime. As a result, a frequency shift occurs near 55 eV, appearing as the derivative feature

in the pump probe spectra, and is the origin of the negative signal in the dynamic cut. Overall, these experimental data agree with results from literature well. Thus, overall, our compact time-resolved EUV

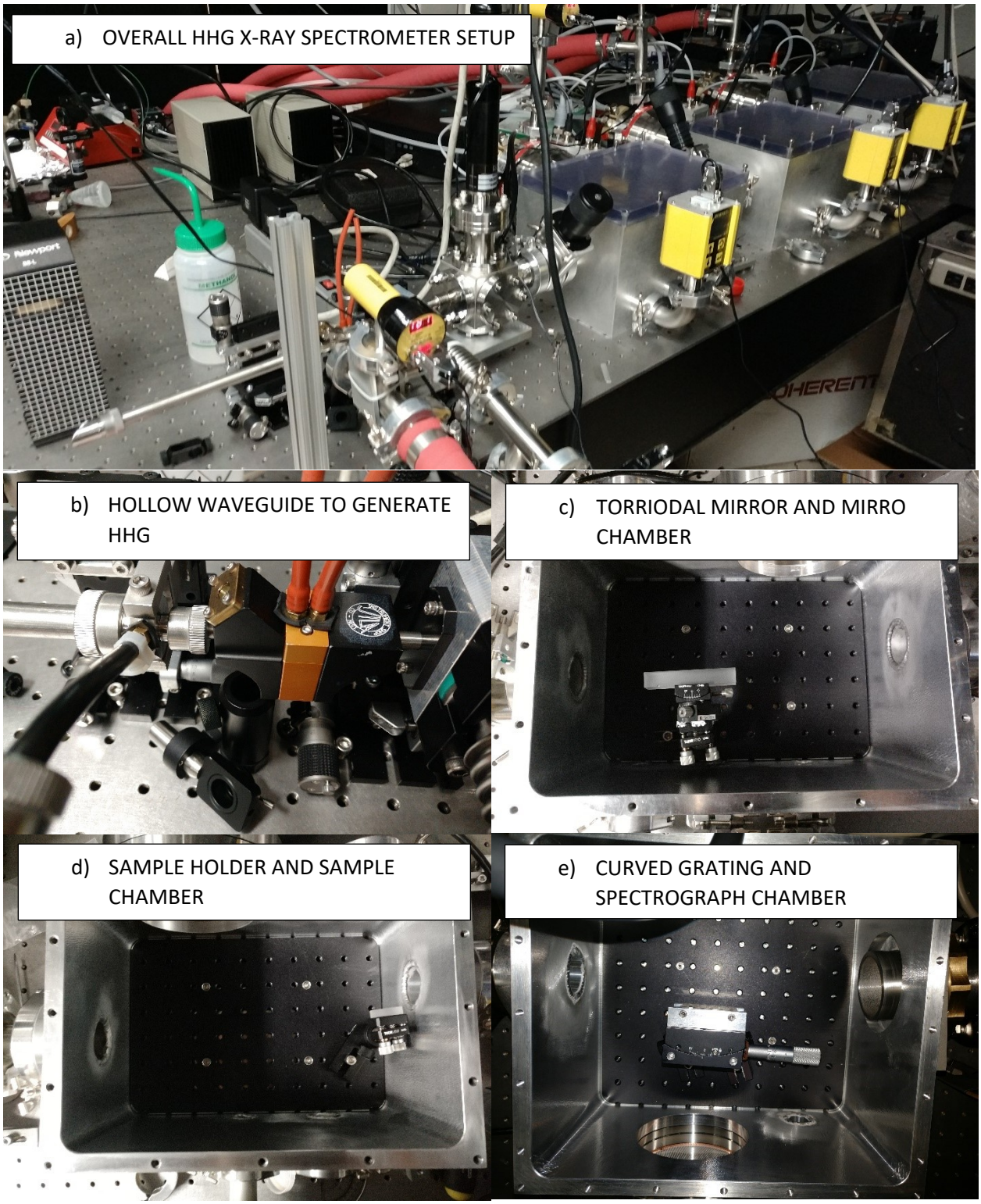

Figure 1. experimental setup of hhg EUV/x-ray beamline and pump probe setup.

spectrometer has been demonstrated to be able to follow element specific dynamics at solid state material surfaces.

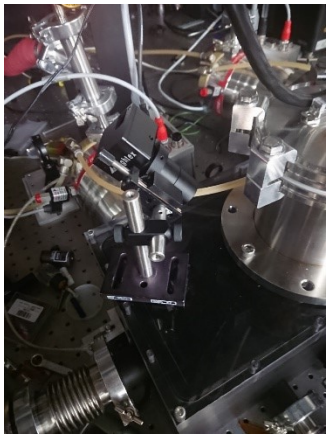

Figure 2. image system for pump probe overlap

Using this experimental setup, we found that not all Fe_2O_3 favors polaron formations. Indeed, aside from the polaron formation dynamics just discussed, which is similar to the findings from Leone and Baker's groups, we found that when Fe_2O_3 are stored under ambient environments for sometime, then their charge dynamics changes dramatically. Fig.3f shows a dynamic cut at 55 eV of the aged Fe_2O_3, and only a strong positive peak appears near t=0, with a fast lifetime of 230 fs. After that, the pump probe traces return nearly back to zero, close to the noise level. This result indicates that there is no peak shift happens to the Fe absorption and therefore no polaron formation. The fast decay lifetime and the negligible polaron signal indicate that there is a competing process that relax the excited electron and avoid it forming polarons. Such a process could be either electron and hole quickly recombine or electron transfer to other molecules. Thus, despite both samples are Fe_2O_3, certain lattice rearrangement or surface modification happens to the aged sample, which do not favor polaron formation anymore. The exact origin of the charge dynamic difference is still under investigation. To the best of our knowledge, this contrast in charge dynamics between Fe_2O_3 have not been reported before.

Currently, based on the support of DURIP in the past year, we are preparing two manuscripts: one reports the experimental setup, and the other one report the newly found competing electron-hole recombination channel. More importantly, this DURIP grant significantly helps the instrumentation situation for my AFOSR YIP project, as we now have a dedicated instrument for conducting the AFOSR projects.

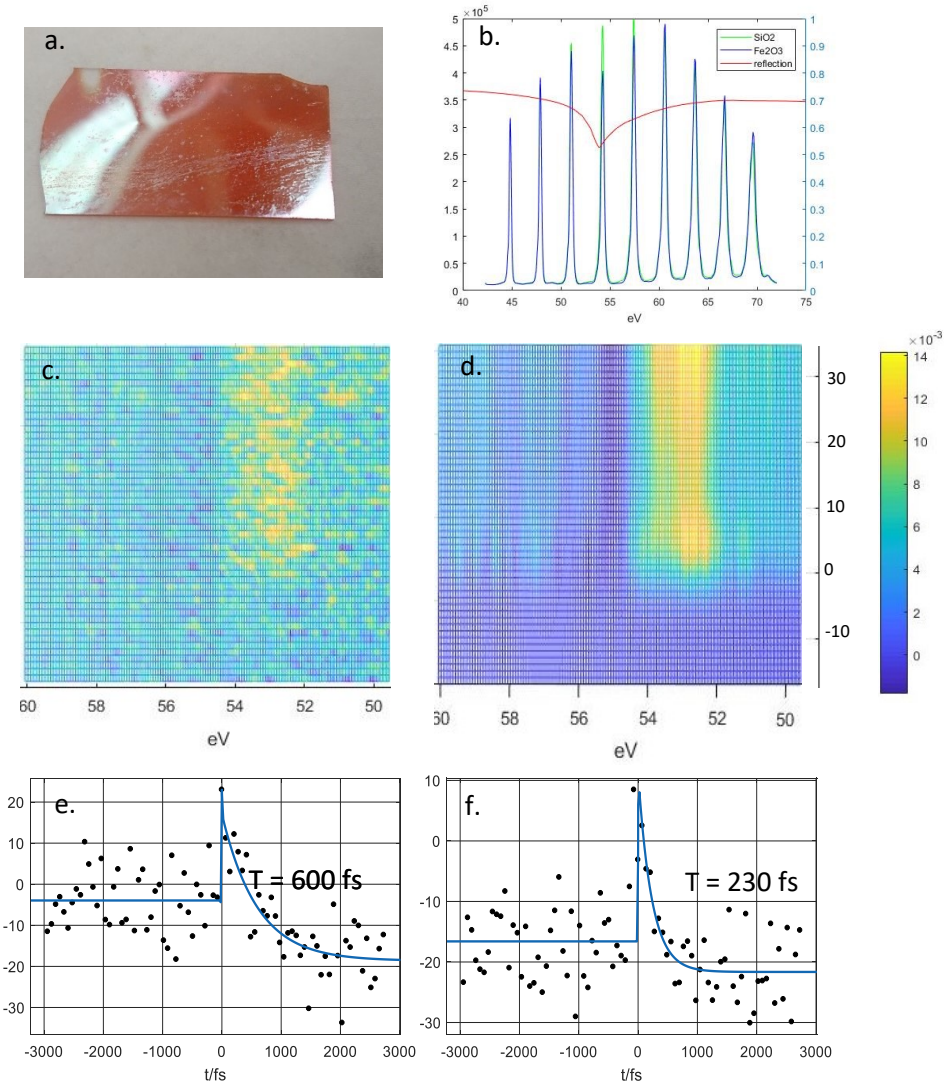

Figure 3. Fe2O3 sample and its EUV spectra. (a) image of hematite sample, (b) reflected HHG from Fe2O3, SiO2 and theoretical reflectivity of Fe2O3, (c) experimental raw and (d) reconstructed transient EUV spectra dynamics of Fe2O3 (e) finer temporal scan near 55 eV, of sample studied in (c and d), the dip is assigned to initial electron-hole pair generation, which quickly convert into a negative signal, because of polaron formation. (f) same scan as (e) but on an aged Fe_2O_3. The recovery dynamics is faster, and accordingly, the signal of polaron formation is nearly negligible. This might be due to that the fast relaxation provide a competing path way (charge recombination or electron transfer) that suppress polaron formation.