Ultrafast XES During XFEL Heating of Fe_3O_4: Watching Magnetism Melt and Electrons Delocalize While the Lattice Stays Cold

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The study of warm dense matter (WDM), i.e., solid-density samples at electronic temperatures comparable to their Fermi energy, is an emerging field at the middle ground of condensed matter physics and traditional dense plasma physics. The WDM regime encompasses interesting examples in astrophysics, planetary science, and fusion energy research. While most WDM studies rely on compression and heating by laser-plasma methods, XFEL heating is an interesting alternative, especially in cases where crystallinity of the sample may be maintained during the heating pulse, resulting in a (briefly) frozen lattice with extremely hot electrons in varying degrees of local thermodynamic equilibrium.

Here, we report an x-ray free electron laser heating study performed at the MEC endstation of LCLS for the classic magnetic material Fe_3O_4. Using the heating pulse also as a time-averaged probe pulse, we measure XRD, Fe Kβ XES, and also Fe VTC XES. This combination of methods allows independent monitoring of crystallinity, electron localization, and d-shell magnetism.

We find good crystallinity during the incident pulse. As pulse intensity is increased a sequence of spin-flip and then progressive charge-transfer effects can be observed by the comparison of XRD and the two XES methods. Temporospatial simulation of the ionization process is used to aid quantitative interpretation of the observed charge-transfer and spectroscopic signatures.

These results demonstrate the importance of multimodal studies while also providing an interesting example where solid-state physics concepts and environments remain relevant up to 1,000,000 K.

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