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Visualization of the ultrafast electronic dynamics in warm dense copper using femtosecond X-ray absorption spectroscopy

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The study of warm dense matter (WDM) is becoming more critical to understand the extreme plasma conditions in nature, such as planetary science, inertial confinement fusion, advanced laser ablation, and machining. The WDM generated by irradiation of the intense ultrashort laser on the solid density sample is highly non-equilibrium and lay in the intermediate state between the condensed matter and plasma.

Recently, time-resolved X-ray absorption spectroscopy (TR-XAS) has been developed to study the electron and lattice dynamics in such WDM. Some previous works showed that the TR-XAS technique could be applied to monitor the excited electron-hole distributions of WDM in broad energy ranges with picosecond resolutions [1,2]. However, in the ultrafast time regime (< ~ 1 ps), the electron dynamics is still questionable due to the lack of experimental resolution.

In this contribution, we present the femtosecond visualization of electron dynamics in warm dense copper using ultrashort X-ray pulses from PAL-XFEL. The rich dynamical features related to 3*d*-holes of WDCu were observed. In general, the energy transition process of the ultrashort laser-heated matter is described by the two-temperature model (TTM), which employs the concept of the quick thermalization of electrons. A finite electronic temperature is established in a few tens of femtoseconds while the lattice remains cold. Then, the two subsystems equilibrate through electron-phonon coupling. However, this simple TTM hardly explains the new experimental data in sub-picosecond regime.

The detailed dynamics of the electronic system might be more complicated than those described by the simple TTM [3]. We propose the improved model including the dynamical band-shifts and the recombination of the nonthermal electrons. This provides an improved explanation for the experiment. The use of femtosecond X-ray absorption spectroscopy here arguably probes the electronic structure of WDM, and provides evidence on the hardening of phonon modes of laser irradiated noble metals which undergoes ultrafast melting process [4].

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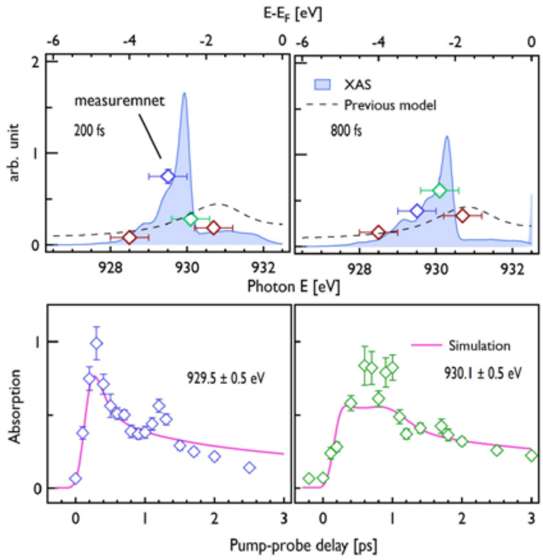

Figure 1. [top] Comparison of XAS calculation based on the traditional two-temperature model (dash) and the modified model (blue curve) with femtosecond X-ray absorption measurements for WDCu at the selected times (200 and 800 fs). [bottom] Temporal evolution of the X-ray absorption at 929.5 and 930.1 ± 0.5 eV are compared with improved calculations.

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