Ultrafast nonequilibrium dynamics in warm dense noble metals

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Recent advances in intense ultrafast lasers have enabled the creation of highly excited states of matter that are far from thermal equilibrium. The ultrafast absorption of energy in condensed matter results in extreme material conditions, known as warm dense matter (WDM), in which the thermal energy is comparable to the Fermi energy and the ions are strongly coupled. WDM bridges the gap between condensed matter and plasma physics. Improving the fundamental understanding of ultrafast energy relaxation and the structural evolution in strongly out-of-equilibrium conditions is relevant for a wide range of fields, such as femtosecond laser ablation, micromachining, and even nanosurgery.

In terms of the response of systems irradiated by femtosecond laser pulses, the generally accepted concept is that an optical pulse directly excites electrons, which quickly thermalize, and that 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. This is the basic premise underlying the two-temperature model (TTM). This framework has been extensively applied to calculate the optical and thermophysical properties of laser-irradiated matter and to develop advanced models for thermal and nonthermal melting, as well as to interpret data from various experiments involving the interaction of matter with ultrashort laser pulses. However, the detailed dynamics of the electronic system might be more complicated than those described by the simple TTM. The hardening of phonon modes in noble metals is one of the interesting predictions in the highly excited two-temperature system. The excitation of localized d electrons may reduce the screening of the electron-ion potential. As a result, the d band may shift toward lower energies and the melting temperature increases. However, to the best of our knowledge, a direct measurement of modified electronic structure in such systems has not been reported to date.

In this talk, I will present a femtosecond measurement of the optical reflectivity of warm dense gold and X-ray absorption of warm dense copper irradiated with intense laser pulses. A series of ultrafast experiments using femtosecond optical and x-ray free-electron lasers reveal the rich dynamical features of nonthermal electrons and vacancies, and their interactions with lattice. I will also discuss an improved modeling of WDM dynamics, which includes recombination of nonthermal electrons and the dynamic shift of the excited d band. It can successfully reproduce the key features observed in the measurement and reveal the ultrafast population balance between conduction and localized electrons. These shed light on improving our understanding of material's ultrafast dynamics and related transport properties under extreme temperature and pressure conditions.

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Reference

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[2] M. Kim et al., Appl. Surf. Sci. 561 150073 (2021)