

3.B Time-Resolved Observation of Electron-Phonon Relaxation in Copper

The 1-KHz, synchronously amplified, colliding-pulse, mode-locked¹ laser at LLE is currently being used to study ultrafast phenomena in solid-state materials. Transient thermal modulation of the optical properties of metals is under investigation with the goal of time-resolving electron relaxation kinetics.²

Thermal modulation of the optical properties of metals is a widely used technique in studying critical points in band structure. Recently the modulation of reflectivity of copper has been used to observe nonequilibrium electron-lattice temperatures during picosecond (~ 5 -ps FWHM) laser heating of up to a few degrees.³ Although nonequilibrium heating was demonstrated in these experiments, the time resolution was insufficient to resolve electron-phonon relaxation. In a subsequent report, the phenomenon of thermally enhanced multiphoton photoemission was used to time resolve electron-phonon relaxation in tungsten.⁴ Results indicated that such relaxation is accomplished in a few hundred femtoseconds. Using amplified, 150-fs to 300-fs laser pulses, we have time resolved electron-phonon relaxation by monitoring the laser-heating-induced modulation of the transmissivity of 200-Å copper films.

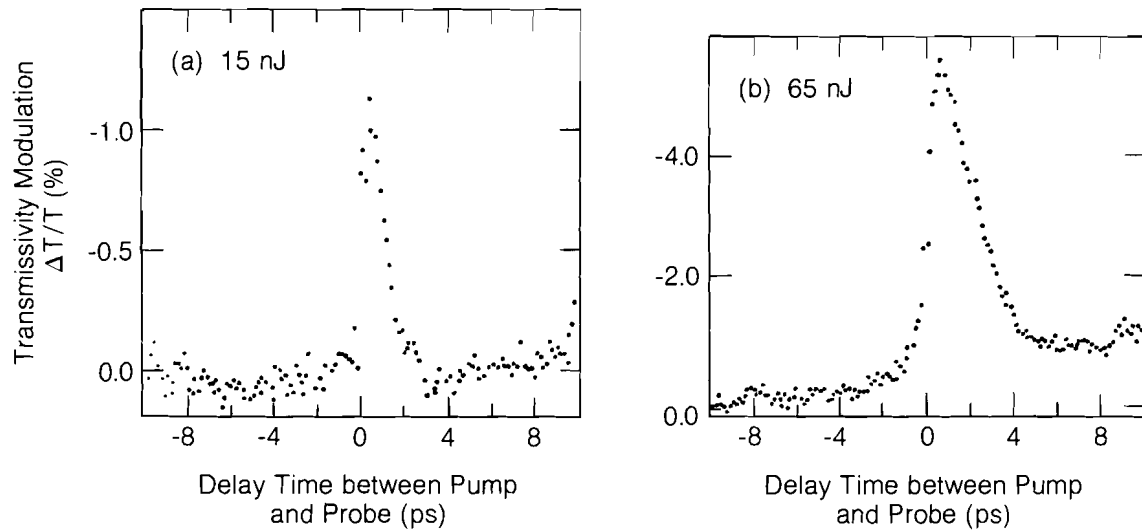
A 1-KHz, synchronously amplified, colliding-pulse, mode-locked laser ($\lambda \sim 620$ -nm) was used for the pump-probe experiments. The sample was heated by using the 620-nm fundamental laser frequency. Probing was accomplished at 620 nm or by using a 10-nm (FWHM) band from white light, generated by focusing the probe beam on an ethylene-glycol cell. The pump and probe were incident collinearly, normal to the copper film (polarized perpendicular to each other), and focused to ~ 27 - μm - and ~ 14 - μm -diameter spots respectively, such that the probe was near the center of the pump.

The transmissivity of the thin copper films at $\lambda \sim 620$ nm during laser heating (~ 300 -fs FWHM), for a pump laser fluence of 15 nJ (a) and 65 nJ (b), is shown in Fig. 27.15. The initial transmitted signal appears to integrate the heating pulse. The decay of the fast transient was found to be 1–4 ps and to increase with the heating pulse fluence. This effect is due to larger differences between electron and lattice temperatures for higher fluences, where more electron-phonon collisions are required for thermalization.

A simplified numerical model of nonequilibrium heating of copper was constructed and applied to the conditions in Fig. 27.15. This model is based on a solution of two coupled nonlinear differential equations in the form^{3,4}

$$C_e(T_e) \frac{dT_e}{dt} = P_o(t) - G(T_e - T_l), \quad (1)$$

$$C_l \frac{dT_l}{dt} = G(T_e - T_l). \quad (2)$$



268

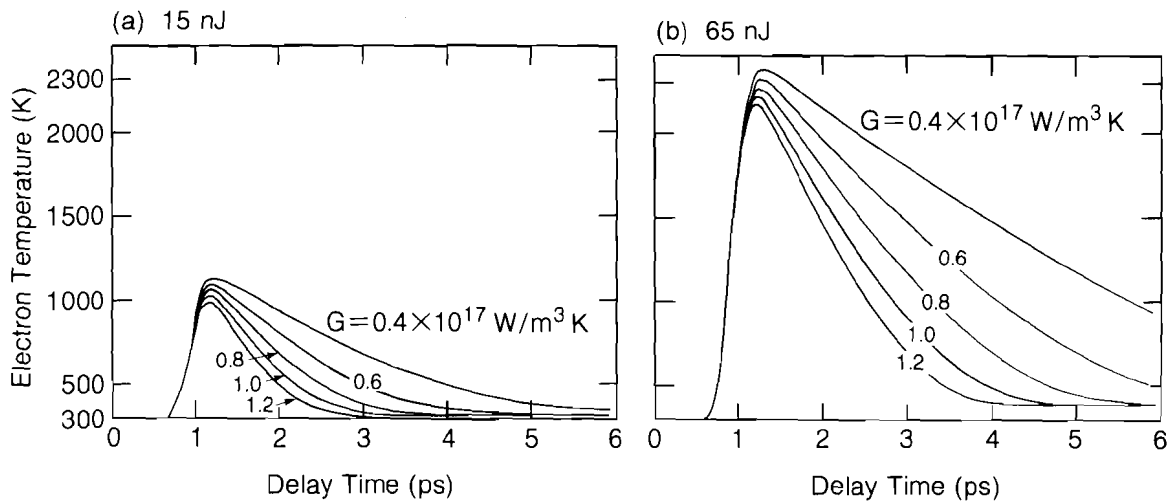
Fig. 27.15

Time-resolved transmissivity of 200-Å copper film during laser heating (~ 300 -fs FWHM at $\lambda \sim 620$ nm). Pump and probe focused to ~ 27 - μm - and ~ 14 - μm -diameter spots, respectively.

Here, $C_e(T_e)$ is the electronic heat capacity, which is directly proportional to the electron temperature; $P_o(t)$ represents the laser heating pulse; and G is the electron-phonon coupling constant. Thermal conductivity losses were ignored due to the thin-film geometry used in the present work. Simulation of conditions used in Fig. 27.15, for different values of electron-phonon coupling constant G , are shown in Fig. 27.16. Results indicate that G has a value of $\sim 1 \times 10^{17}$ W/(m³K). For a pulse energy of 65 nJ (peak fluence $\sim 3.8 \times 10^{10}$ W/cm²), the model predicts a peak electron temperature of 2200°K and an equilibrium electron-phonon temperature of 385°K.

Using white light in 10-nm spectral-band steps from $\lambda = 560$ nm to 640 nm ($\lambda = 590$ nm corresponds to an electron transition from the top of the d-band to the Fermi level) showed similar behavior as when probing at $\lambda = 620$ nm.

In summary, we have directly measured the electron-phonon relaxation time in copper as a function of pump-laser fluence and probe photon energy, for $\lambda = 560$ nm to 640 nm. We have demonstrated nonequilibrium heating with a large (few thousand degrees) difference between electron and lattice temperatures. Electronic and lattice effects on the optical properties of copper were separated in time. Extension of probe measurements to the near-IR and UV parts of the spectrum would serve to separate effects of bound and free electrons on the optical properties and provide considerable information on the band structure.



267

Fig. 27.16

Numerical modeling of the time evolution of electron temperature for the experimental condition of Fig. 27.15. Simulations were made for different values of the coefficient of heat transfer between the electrons and the lattice G .

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