Relaxation of the electron temperature in a copper film excited by femtosecond laser pulses

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The relaxation of an induced optical response in a copper film has been studied by femtosecond laser spectroscopy in the spectral interval 500–640 nm. A method is proposed for directly determining the maximum heating of the electron subsystem. The measured relaxation time $\tau = 1.8$ ps and the electron temperature $t_e = 810$ K are used to calculate the electron–phonon coupling parameter: $\Lambda\langle\omega^2\rangle = 27 \pm 4$ meV^2.

The illumination of a metal surface with laser pulses, whose length is in the femtosecond range, drives the electron subsystem of the metal away from equilibrium with the lattice. The equilibrium is then restored through an electron-phonon coupling. The dynamics of the relaxation to equilibrium can be studied by measuring the transmission and/or reflection of a surface of the sample with the help of a delayed probe pulse. The time scale (τ) for the relaxation to thermal equilibrium in a metal—the electron–phonon relaxation time—is determined by the electron–phonon coupling parameter $\Lambda\langle\omega^2\rangle$ of the material of interest. This parameter plays a fundamental role in solid state theory.[1] According to Allen,[2] the relaxation rate is related to this parameter by

$$1/\tau = 3\hbar\Lambda < \omega^2 > /\pi k_B t_e, \qquad (1)$$

where t_e is the absolute temperature of the electron subsystem of the sample. In experiments with a femtosecond time resolution, one can thus measure the parameter $\Lambda\langle\omega^2\rangle$ if one knows the absolute temperature of the electron subsystem of the metal in the excited state.

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The temperature change resulting from excitation of a sample was determined in Refs. 3–6 on the basis of femtosecond spectroscopy of copper and other metals, through a numerical simulation of heat transfer in a metal.

Our purpose in the present study was to develop a direct method for determining the heating of the electron subsystem in a copper film. For this purpose, we supplemented the data found during excitation of a sample by ultrashort light pulses with data on the spectrum of the changes in the optical constants of copper. The latter data were obtained under equilibrium conditions, upon a precisely controlled change in the temperature of a sample in a cryostat. The electron-phonon relaxation time was measured, and the parameter $\Lambda\langle\omega^2\rangle$ was determined.

The test sample in the present experiments was a Cu film 20 nm thick on a glass substrate. We used a beam of amplified light from a colliding-pulse, mode-locked laser which generated a continuous train of pulses 120–180 fs long at a wavelength $\lambda = 605$ nm, with an average energy of about 100 μJ in the pulse, and with a repetition frequency of 3 Hz. This beam was split into two channels, for excitation and for probing. The exciting beam passed through an adjustable delay line and was focused onto the sample. The excitation energy ranged up to 50 μJ in the pulse. The probe beam was first focused in a cell filled with heavy water, in order to generate pulses of a continuum for use in probing the sample over a broad spectral interval (500–640 nm). The relative change in the transmission of the sample, $\Delta T/T$, was determined experimentally. The probe beam was focused inside the excitation spot. The spectrum of the probe pulse transmitted through the sample was measured by an optical multichannel analyzer. These measurements were carried out at room temperature.

In the difference spectra $\Delta T/T(E)$ we observe a dip (a decrease in transmission) at $E = 2.15$ eV. This dip corresponds to transitions from a d level in the valence band to the vicinity of the Fermi level. Figure 1 shows the response $\Delta T/T(\tau)$ measured at

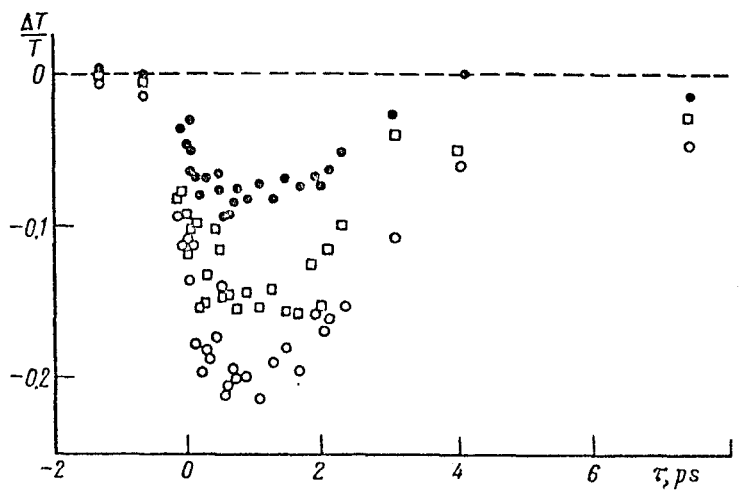

FIG. 1. Time evolution of the relative change in the transmission of a copper film for various photon energies. □—$E = 2.06$ eV; ○—$E = 2.15$ eV; ●—$E = 2.21$ eV.

three selected wavelengths in these experiments. The trailing edge on the plot was approximated by an exponential function. For $E = 2.15$ eV, i.e., for the wavelength at which the response has its maximum amplitude, the relaxation time of the response of the sample is $\tau = 1.8 \pm 0.1$ ps.

In order to determine the maximum electron temperature t_e achieved experimentally, we need to determine how the difference response $\Delta T/T$ at the selected wavelength λ_0 is related to the temperature t_e. To solve this problem we recorded transmission and reflection spectra of the test sample at various fixed temperatures. The sample was placed in a nitrogen-cooled cryostat and exposed to a collimated light beam from an incandescent lamp. The reflection and transmission spectra were recorded by an optical multichannel analyzer. We studied the spectral interval 1.9–2.4 eV and temperatures from 100 to 300 K. When we then subtracted the spectra corresponding to a reference temperature $t_0 = 291$ K from the spectra recorded at the various temperatures t, we found difference spectra $\Delta T/T(E)$ and $\Delta R/R(E)$, which depend parametrically on t. The spectra $\Delta T/T(E)$ and $\Delta R/R(E)$ were converted[7] into difference spectra of the real and imaginary parts of the dielectric constant of the sample, $\Delta\epsilon_1(E)$ and $\Delta\epsilon_2(E)$.

The solid line in Fig. 2 shows spectra of $\Delta\epsilon_1(E)$ and $\Delta\epsilon_2(E)$ for $t = 150$ K. In general, the optical response of the sample is determined by the changes in both the electron temperature and the lattice temperature. However, since the spectrum $\Delta\epsilon_1(E)$ has no sign-changing component, we conclude that the lattice component is small. The $\Delta\epsilon_2(E)$ spectrum is a sign-changing function, and it is shifted away from the zero line. The sign-changing component of the $\Delta\epsilon_2(E)$ spectrum results from a thermal rounding of the step in the Fermi distribution. The shift of the $\Delta\epsilon_2(E)$ spec-

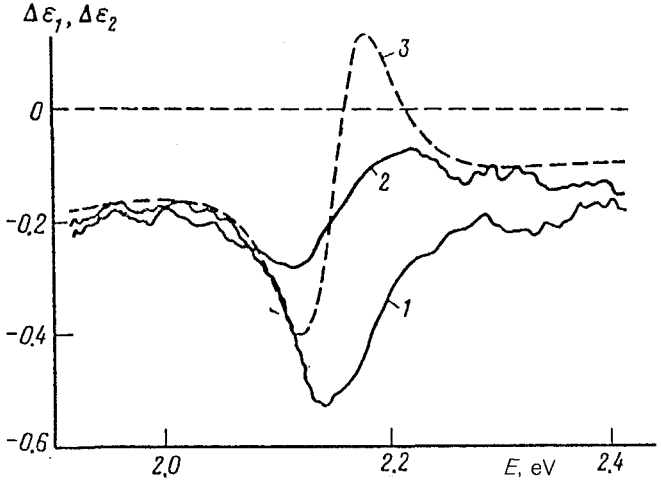

FIG. 2. Difference spectra (1) $\Delta\epsilon_1$ and (2) $\Delta\epsilon_2$ found from the experimental data for $t = 150$ K, along with (3) a theoretical curve for $\Delta\epsilon_2$ (see the text proper for an explanation).

trum away from the zero line stems from a change in the mean free time of the electrons upon a change in temperature (the Drude component). An estimate shows that the Drude component of the change in the imaginary part of the dielectric constant as the temperature is changed from 291 to 150 K is -0.16 for $\hbar\omega = 2.0$ eV and -0.09 for $\hbar\omega = 2.4$ eV. These figures agree with the size of the shift of the $\Delta\epsilon_2(E)$ spectrum from the zero line in Fig. 2.

The $\Delta\epsilon_2(E)$ spectrum can be calculated on the basis of a crude model based on the assumption that the interband component of ϵ_2 stems from transitions from a flat d level in the valence band to a parabolic conduction band.[7] The dashed line in Fig. 2 is a $\Delta\epsilon_2(E)$ spectrum calculated for $t = 150$ K for the case in which there is a free-electron component. The spectral width of this calculated $\Delta\epsilon_2(E)$ spectrum corresponds to the temperature $t_{\max} = 291$ K. However, the spectral width of the response of the test sample is considerably larger than expected. On this basis we assumed that there was a broadening of the optical response of the sample. This broadening might be caused by a finite spectral width of the d level, by possible indirect transitions from the d level to the conduction band, and by a physical nonuniformity of the copper film. After the broadening parameter was incorporated in the theoretical model, we fitted the theoretical $\Delta\epsilon_2(E)$ spectrum to the experimental spectrum and found a broadening of 0.07 eV.

On the basis of this model, with broadening, we calculated the amplitude of the optical response, $\Delta T/T(E = 2.15$ eV), as a function of the temperature t. The results are shown by the solid line in Fig. 3. The circles are experimental points.

We used this functional dependence to determine the electron temperature during ultrashort laser excitation of a copper film. The maximum electron temperature turned out to be 810 ± 30 K. Using the relaxation time for the excitation correspond-

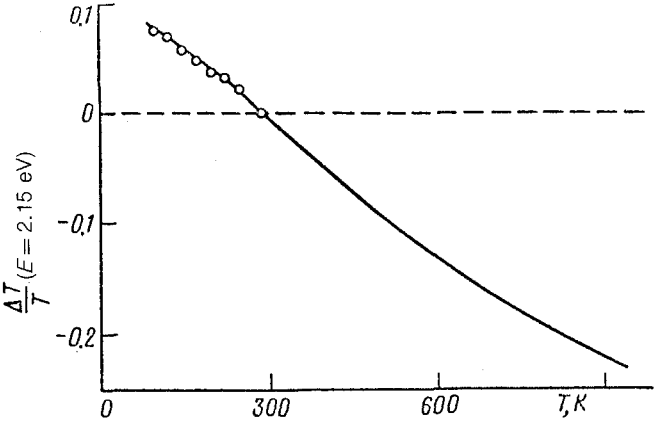

FIG. 3. Theoretical results on $\Delta T/T(E = 2.15$ eV) as a function of the temperature t. The circles are experimental data under equilibrium conditions.

ing to transitions near the Fermi level, $\tau = 1.8$ ps, we find $\Lambda\langle\omega^2\rangle = 27$ meV^2 from Eq. (1). The error in the determination of $\Lambda\langle\omega^2\rangle$ is 4 meV^2 and stems from the errors in the determination of τ and t_e. We finally find $\Lambda\langle\omega^2\rangle = 27 \pm 4$ meV^2. This result agrees well with data in the literature,[6] including some data found by other methods.[8] This agreement is evidence for the validity of the method used here to determine the electron temperature of a metal.

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