Femtosecond Laser Interaction with Metallic Tungsten and Nonequilibrium Electron and Lattice Temperatures

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High-intensity, 75-fs optical pulses have been applied to observe multiphoton and thermally enhanced photoemission from a tungsten metal surface. Experimental results suggest the presence of anomalous heating, a transient nonequilibrium temperature difference between the electrons and lattice. Pump-probe measurements indicate an electron-phonon energy relaxation time of several hundred femtoseconds.

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Multiphoton photoelectric emission from metals has been the subject of theoretical and experimental investigation for many years.[1] The advent of high-intensity ultrashort-pulse lasers made possible the extension of these studies into the picosecond time regime.[2,3] For intense optical pulses of duration shorter than or comparable to the electron-phonon energy relaxation time, it has been postulated that a nonequilibrium between the electrons and phonons may be achieved.[4] The smaller heat capacity of the electron gas would then allow the generation of an electron temperature much greater than the lattice temperature. Previous experimental investigations using high-intensity picosecond pulses have shown no evidence for this phenomenon of anomalous heating.[2,3] Recently, evidence for nonequilibrium electron temperatures has been reported from low-intensity picosecond reflectivity measurements in copper.[5] However, the temperature difference between the electrons and lattice was less than a few kelvins and the time resolution was insufficient to permit the measurement of the electron-phonon energy-transfer time.

In this Letter we report the observation of multiphoton and thermally assisted photoemission from a tungsten metal surface by means of high-intensity 75-fs optical pulses. Measurements of the integrated charge emission versus incident pulse intensity suggest a thermal nonequilibrium between the electrons and the lattice. Time-resolved pump-probe measurements indicate an electron-phonon energy relaxation time of several hundred femtoseconds.

Let us begin by considering the processes of multiphoton and thermally assisted photoemission in metals. At low incident laser intensities heating is negligible and the electrons are in a low-temperature Fermi distribution. The work function of tungsten is approximately 4.3 eV. With a laser photon energy of 2 eV, photoemission should occur primarily via the three-photon process. As the laser intensity is increased, heating of the electron Fermi distribution results in increased emission from thermally assisted lower-order processes.[6-8]

When a short optical pulse interacts with a metal, the energy is first absorbed by the electrons which thermalize rapidly through electron-electron scattering. The electrons then transfer energy to the crystal lattice through electron-phonon coupling via deformation-potential scattering. If the incident laser pulse is long compared to the electron-phonon energy-transfer time, the electrons and lattice will remain in thermal equilibrium and the temperature may be described by the equation

$$C_i \partial T/\partial_t = K\nabla^2 T + A(r,t), \tag{1}$$

where $A(r,t)$ represents the heat source term due to the laser pulse, K is the thermal conductivity, and C_i is the lattice heat capacity per unit volume.

Conversely, if the laser pulse duration is comparable to or shorter than the electron-phonon energy-transfer time, then the electrons and lattice will not be in thermal equilibrium. The time evolution of the electron and lattice temperatures T_e and T_i are then governed by a set of coupled nonlinear differential equations[4]:

$$C_e(T_e)\frac{\partial T_e}{\partial_t} = K\nabla^2 T_e - g(T_e - T_i) + A(r,t), \tag{2}$$

$$C_i \partial T_i/\partial_t = g(T_e - T_i). \tag{3}$$

The electronic heat capacity depends linearly on the electron temperature. The electron and lattice temperatures are coupled through the electron-phonon coupling constant $g \cong \pi^2 mnv_s^2/6\tau_{ep}T_i$, where m is the electron mass, n the electron density, v_s the sound velocity, and τ_{ep} the electron-phonon collision time.[4,9] Since $\tau_{ep} \sim 1/T_i$, g is a constant independent of temperature.

Experimentally, electron temperature and the electron-lattice energy-transfer time may be determined indirectly from measurements of the integrated photoemitted charge versus laser pulse intensity. In 1980, Yen, Liu, and Bloembergen[2] used 30-ps pulses at 1.06 μm to investigate thermally assisted photoemission in tungsten. No evidence for nonequilibrium electron and lattice temperatures was observed, and it was determined that the electron-phonon coupling constant g was greater than 2×10^{11} W/cm^3 K. Subsequent measurements with 2-ps pulses at 300 nm in zirconium also did not present any evidence for anomalous heating and placed similar bounds on the coupling constant.[3]

The laser system employed in our investigation consisted of a femtosecond, colliding-pulse, mode-locked ring dye laser and a four-stage dye amplifier[10,11] which generated 75-fs pulses at a wavelength of approximately 620 nm with a repetition rate of 10 Hz. Single pulse energies of up to 0.4 mJ were obtained to yield peak intensities of several gigawatts.

The laser pulses were focused at normal incidence onto the surface of a polished polycrystalline tungsten sample mounted inside a high-vacuum chamber maintained at 10^{-6} Torr. A 1-mm-diam wire was mounted 2 mm in front of the sample surface and biased to 5 kV to function as an anode and to collect the photoemitted charges. The integrated charge emitted from the sample was detected by a high-sensitivity, low-noise amplifier which had a detection limit of $\sim 10^4$ elementary charges. A portion of the incident laser pulse was directed to a reference photodiode which measured the pulse energy. Both the reference and photoemission signals were digitized and recorded by computer on a shot-to-shot basis. Pump-probe measurements were obtained by dividing the excitation into two pulses. Relative temporal delay between the two was varied by a computer-controlled, micron-step-size mechanical stage.

The experimentally measured, integrated photoemitted charge produced by 75-fs laser pulses is shown on a log-log plot in Fig. 1 as a function of the incident pulse intensity. The low scatter of the data points indicates that fluctuations in the durations of the amplified pulses are extremely small. The dependence of the photoemitted charge on the laser intensity differs significantly from what has been observed in picosecond photoemission experiments. At low intensities the dependence does not have the expected slope 3 of the intrinsic three-photon process but instead has a slope of approximately 4. At higher intensities the slope gradually decreases with increasing intensity up to the damage threshold. The damage threshold coincides with the onset of visible surface damage as well as the emission of positive ions from the sample.

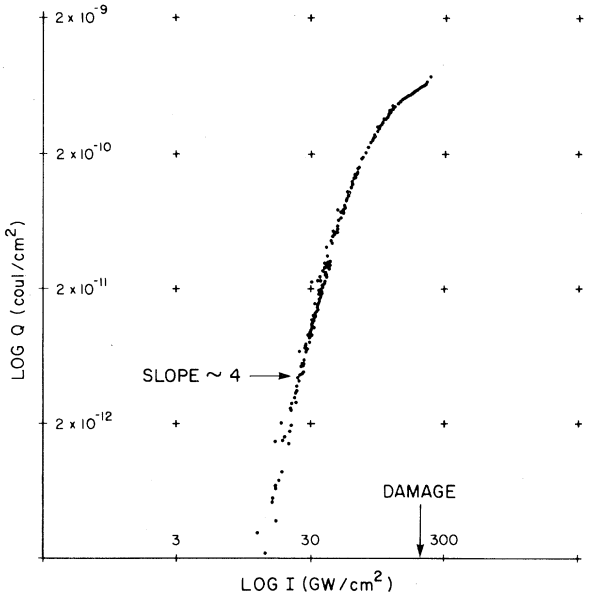

FIG. 1. Log-log plot of integrated photoemitted charge vs incident laser pulse intensity produced by 75-fs pulses on tungsten. Each point represents a single laser shot.

The decrease in the slope of the photocurrent versus laser intensity which occurs at high intensities is attributed to a charge saturation effect which suppresses the increased photocurrent and masks the signature of thermally assisted photoemission. Because of the extremely short pulse durations, peak currents in excess of 1000 A/cm^2 are produced in the high-intensity regime. The magnitude of the saturation effect was observed to vary with different anode voltages, becoming more severe at lower voltages. The time depedence of the saturation was investigated by measuring the total photoemitted charge produced by two equal-intensity pulses separated by a variable temporal delay. These pump-probe measurements indicate that the saturation recovers on a time scale of approximately 100 ps. These findings imply that the charge saturation results from a space-charge accumulation in

the vacuum rather than an effect which is internal to the metal.

Note that even at low intensities the photoemission is more nonlinear than the intrinsic three-photon process. As shown in Fig. 1, a dependence of approximately slope 4 is observed at intensities of < 30 GW/cm^2. In fact, the nonlinearity may be even greater because of possible charge saturation. Unfortunately, the observation of the photoemission at lower intensities is limited by the experimental detection sensitivity and the low electron yield produced by femtosecond pulse durations. If this enhanced slope is produced by thermally assisted photoemission, then energy arguments imply that there must be a thermal nonequilibrium between the electrons and lattice. If there is a transient nonequilibrium, then electron temperature increases of up to 2000 K would be possible even with fluences an order of magnitude below the damage threshold. However, if the electrons and lattice were in equilibrium, a temperature increase of only about $\frac{1}{10}$ of the melting temperature of ~ 3700 K would be possible. For such small temperature increases, thermal enhancement would be negligible. These results are in agreement with Fowler-DuBridge calculations of the photocurrent.[6-8] However, a quantitative comparison with the experimentally measured slopes or absolute electron yield is problematic because of charge saturation effects as well as uncertainties in the quantum-mechanical cross sections for the various-order nonlinear and thermally assisted photoemission processes.

Further evidence for disparate electron and lattice temperatures is provided by femtosecond pump-probe measurements of the transient electron temperature. Figure 2 shows pump-probe measurements obtained by the use of two pulses of equal intensity in the low-intensity, unsaturated regime. When the two pulses are temporally overlapped, separated by delays of the order of the pulse width or less, the photocurrent is determined by the instantaneous intensity of the coherently overlapped pulses. This yields what is effectivly a high-order autocorrelation function of the pulse intensity. The expected peak-to-background contrast ratios would be 10:1 and 35:1 for third- and fourth-order nonlinearities, respectively. The low contrast ratios obtained in the experimental data are the result of space-charge saturation which suppresses the photoemission at high intensities.

When the two pulses are several hundred femtoseconds apart and are not temporally overlapped, the pump-probe measurement shows the effects of a transient electron heating produced by the first pulse as it enhances the photoemission induced by the second pulse. The enhanced photoemission in the wings of the pump-probe correlation data indicates the presence of thermally assisted photoemission with electron temperature increases lasting several hundred femtoseconds.

The pump-probe data may be qualitatively com-

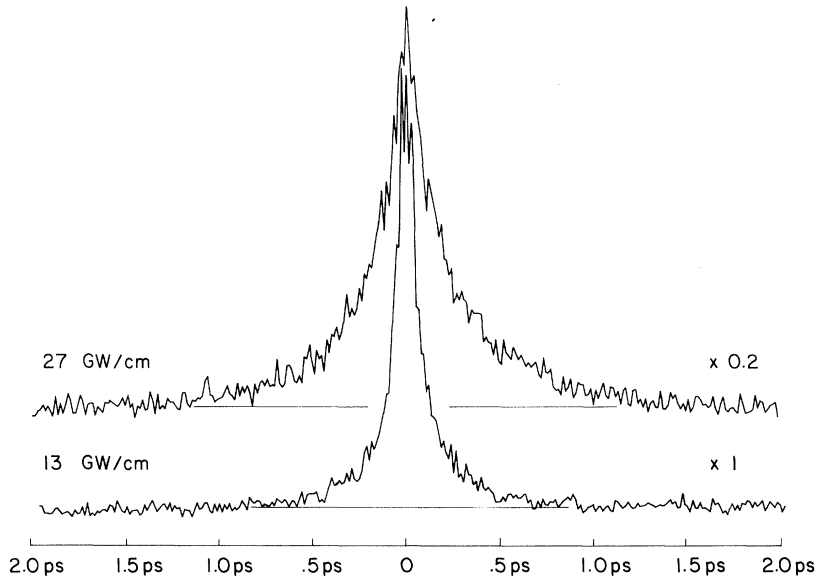

FIG. 2. Pump-probe measurement of transient electron cooling obtained by measuring integrated charge produced by two equal-intensity pulses separated by a variable time delay.

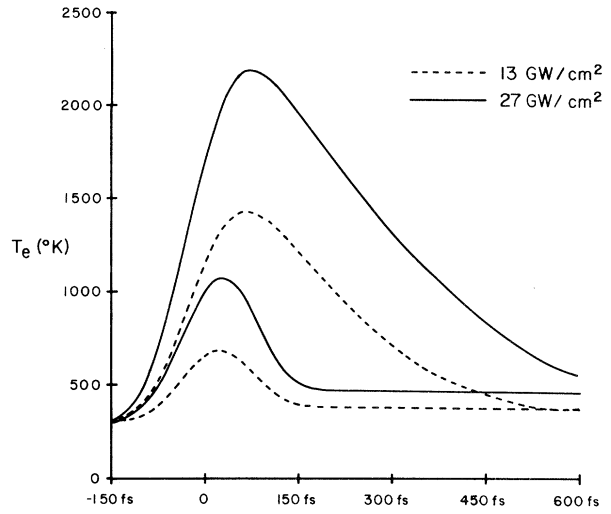

FIG. 3. Theoretically predicted behavior of electron temperatures for anomalous heating in tungsten produced by 13- and 27-GW/cm^2, 75-fs laser pulses. The upper and lower sets of curves correspond to values of the electron-phonon coupling constant of $g = 5\times10^{11}$ and 5×10^{12} W/cm^3 K, respectively.

pared to the behavior of the electron temperature predicted by the anomalous heating equations (2) and (3). Figure 3 shows the calculated transient electron temperatures for two different values of the electron-phonon coupling. When the coupling is very large, $g \sim 5\times10^{13}$ W/cm^3 K, the electron and lattice remain essentially in equilibrium and the temperature increase is governed by the lattice heat capacity. When the coupling is decreased, the electron temperature increases rapidly and requires longer to reach equilibrium with the lattice. For values of $g \sim 5\times10^{11}$ W/cm^3 K or less, the elevated electron temperature persists for several hundred femtoseconds. Note that both the electron temperature and cooling time increase with increasing laser intensity.

A qualitative comparison of the pump-probe measurements with the calculated electron temperatures may be made by considering the time required for the electrons and lattice to reach equilibrium. Experimental pump-probe data suggest that the electron-phonon coupling constant is of the order of $g \sim 5\times10^{11}$–1×10^{12} W/cm^3 K. Additional time-resolved studies and the elimination of charge saturation effects are necessary to determine the actual electron temperatures and more accurately measure the electron-phonon coupling constant.

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