# Brief Reports

*Brief Reports are accounts of completed research which, while meeting the usual* **Physical Review** *standards of scientific quality, do not warrant regular articles. A Brief Report may be no longer than four printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

## Electron thermalization in gold

W. S. Fann, R. Storz, and H. W. K. Tom
*AT&T Bell Laboratories, Holmdel, New Jersey 07733*

J. Bokor
*AT&T Bell Laboratories, Murray Hill, New Jersey 07974*
(Received 2 June 1992)

We report the direct observation of the thermalization of electrons in gold following 180 fs optical pulse excitation. The evolution of the electron energy distribution from the nascent (as photoexcited) to a hot Fermi-Dirac distribution was measured by time-resolved photoemission spectroscopy. Depending on the excitation density, thermalization times as long as $\approx 1$ ps were observed. A model incorporating both electron-electron and electron-phonon scattering, and using Fermi-liquid theory to properly account for screening is found to reproduce the main features of the experiment.

Electron-electron (*e-e*) scattering in metals has usually been studied by transport[1] measurements. The contribution of *e-e* scattering to resistance can only be observed at low temperature, because above the Debye temperature electron-phonon (*e-p*) scattering completely dominates the resistivity. According to Landau's Fermi-liquid theory,[2] the resistance due to *e-e* scattering is $\rho_{e\text{-}e} = AT^2$, where $T$ is the temperature and $A$ is a constant. However, even at low temperature, extraction of $\rho_{e\text{-}e}$ from the measured resistivity is complicated by electron-phonon (*e-p*) and defect scattering.[3] Observation of the thermalization of electrons excited by ultrafast optical pulses provides an alternative means to study *e-e* scattering. The relaxation of an optically excited, non-Fermi-Dirac distribution to a hot Fermi-Dirac distribution is mainly through *e-e* scattering due to the large momentum exchange and large phase space available for the process which involves quasiparticle energies in the range of an electron volt.

In this paper, we report the first direct measurement of the thermalization process in an optically excited metal. We are able to observe the nascent (as photoexcited) electron energy distribution, and the time evolution from the nascent distribution to a Fermi-Dirac distribution. The thermalization process is found to take up to $\sim 1$ ps for low optical excitation levels, and proceeds more rapidly for higher optical excitation levels. Because thermalization and electron-phonon energy relaxation occur on similar time scales (on the order of ps), we find that even in this regime it is necessary to simultaneously include both *e-e* and *e-p* scattering to fully understand the dynamics. A model based on the Boltzmann transport equation under the relaxation-time approximation is proposed to explain the experiment. Fermi-liquid theory is used to properly account for Coulomb screening.

Time-resolved photoemission spectroscopy was used to measure the time evolution of the electron energy distribution following ultrashort laser pulse excitation of a gold sample. The sample was a room temperature 300-Å-thick gold film held in vacuum at $5\times10^{-11}$ torr. The experimental apparatus was similar to that reported previously,[4] except that the temporal resolution was improved significantly. The 1.84-eV visible excitation laser-pulse duration in the present experiment was 180 fs, and the 5.52-eV UV probe pulse duration was 270 fs.

Figure 1 shows the time evolution of the distribution function for an absorbed fluence of $120\pm40$ $\mu$J/cm$^2$. The

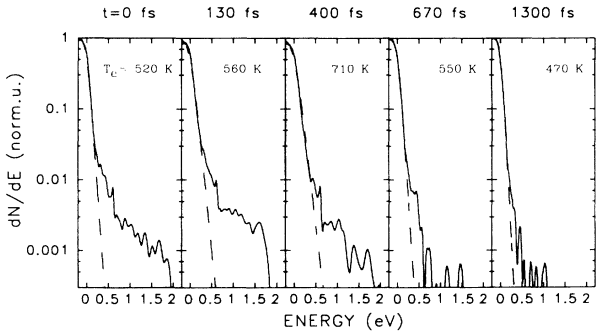


FIG. 1. Electron energy distribution function vs energy with 120 $\mu$J/cm$^2$ absorbed laser fluence at five time delays. The dashed line is the best Fermi-Dirac fit and the corresponding electron temperature $T_e$ is shown. The vertical scale is in units of the density of states.

 

best-fit Fermi-Dirac function is shown by the dashed line, and the energy is referenced to $E_F$, the Fermi energy. The procedure for deriving the joint density of states, and using it to extract the energy distribution function from the photoemission spectra has been described previously.[4] At $t=0$ fs, a nearly flat distribution extends from 0 to 1.84 eV (the pump photon energy). The one-photon cutoff, which is the signature of nascent distribution, can easily be observed. At $t=130$ fs, a plateau from 0.7 to 1.8 eV reaches its maximum value. With this pump fluence, if there were no energy relaxation, then a flat distribution extending from 0 to 1.84 eV (the pump photon energy), with a value of 0.0035, would be observed. The level in Fig. 1 is within a factor of 2 of what we would expect with no relaxation. This observation suggests that the energy relaxation time for the 1.8-eV electrons is about $\frac{1}{2}$ the pump pulse duration, e.g., $\approx 90$ fs.

We have investigated the possibility that the plateau region is due to a "coherent artifact" arising from two-photon ($\omega+3\omega$) photoemission with no intermediate relaxation. This effect would vary in time as the cross correlation of the $\omega$ and $3\omega$ pulses. Using a density-matrix calculation, we find that for the 1.8-eV signal at $t=0$ fs, the ratio of the two-photon coherent contribution to the contribution arising from single-photon photoemission from the real population of photoexcited electrons is roughly equal to the ratio between the dephasing time, $T_2$, and the energy relaxation time $T_1$ of the 1.8-eV electrons. Since $T_1 \geq 2T_2$, the maximum artifact is $\frac{1}{2}$ of the observed signal. Thus, the maximum artifact is $\approx 0.0005$ at $t=0$ fs.

At $t=400$ fs, the best-fit Fermi-Dirac temperature reaches its maximum, 710 K. At this time approximately 30% of the energy remains in the hot tail although most of the electrons $>1.0$ eV are gone. The energy relaxation time for an electron with energy $E$ should scale as $(E-E_F)^{-2}=\delta E^{-2}$ according to the Fermi liquid theory. Qualitatively, this is what we observe in the experiments. It is clear from these data that the electron system loses energy to the lattice before it can thermalize.

Figure 2 shows data similar to that in Fig. 1 except the absorbed fluence is 2.5 times higher, $300\pm 90$ $\mu$J/cm$^2$. The plateau from 0.8 to 1.84 eV has the same shape as in the lower fluence case and approximately twice the amplitude, nearly consistent with the fluence increase. In addition, the formation of a thermalized distribution is clearly faster in this case. By 670 fs, the thermalization is essentially complete. There are several factors which contribute to the faster thermalization of the distribution. First, since a larger energy density has been deposited, the temperature of the thermal part of the distribution increases more rapidly. This increases the number of available electrons with which the nonthermal electrons can scatter, further increasing the scattering rate. Another effect is that as the rapidly heating thermal component overtakes and overlaps the nonthermal part of the distribution, only the higher energy electrons remain in the nonthermal part, and these electrons have the shortest energy relaxation time. The net result is the thermalization time decreases as the excitation density increases.

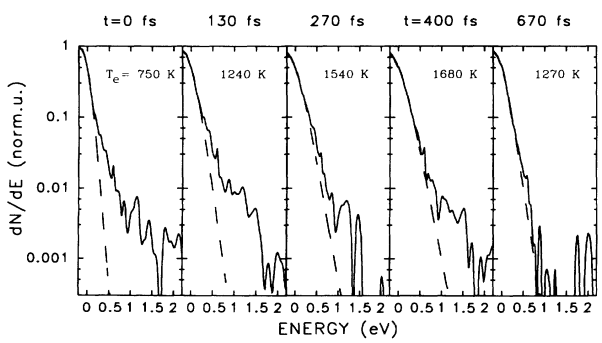


FIG. 2. Electron energy distribution function vs energy with 300 $\mu$J/cm$^2$ absorbed laser fluence at five time delays.

There have been a number of studies of the relaxation of electrons in metals excited by ultrashort laser pulses.[5-11] All of these have been analyzed within the two-step relaxation picture,[12,13] which assumes the electrons first thermalize among themselves and subsequently lose energy to the lattice. It has already been suggested[14,15] that the time scales for electron thermalization and for electron-phonon transfer are potentially overlapping and that the electron temperature $T_e$ and the lattice temperature $T_1$ are not well defined. A recent study[16] of the lattice temperature dependence of the $e$-$p$ relaxation time in Ag and Au has shown that the electron distribution is indeed nonthermal on the time scale of the $e$-$p$ energy relaxation time. This effect is clearly demonstrated in the present experiment. The $e$-$p$ mean free path[17] is $\approx 300$ Å. With Fermi velocity $\approx 10^8$ cm/s, the average collision time is 30 fs. Thus within 1 ps the electron has experienced approximately 30 phonon collisions. The Debye temperature of gold[2] is 170 K, so on average an electron will lose energy equivalent to 85 K at each phonon collision. Thus a substantial amount of energy is transferred into the lattice during the $\sim$1-ps thermalization time.

Now we turn to the relaxation of nonthermal electrons. The lifetime of an excited electron in a degenerate system due to both elastic and inelastic $e$-$e$ collisions is given by Fermi-liquid theory under the random-phase approximation (RPA),[18] as

$$\tau=\tau_0\left[\frac{E_F}{\delta E}\right]^2 . \tag{1}$$

The square factor in the denominator comes from the available phase space and $\tau_0$ is the proportionality constant. This scaling is strictly valid only at $T=0$ when the phase space of available initial and final states is strictly given by $\delta E$, but is a good approximation at finite temperature for electrons with $\delta E \gg kT_e$. $\tau_0$ has been calculated using the Lindhard dielectric function.[18]

$$\tau_0=\frac{128}{\pi^2\sqrt{3}}\frac{1}{\omega_p} , \tag{2}$$

where $\omega_p$ is the plasma frequency. With $\omega_p=8$ eV for gold, $\tau_0\approx 2$ fs. We can make an estimate of $\tau_0$ by noting that, experimentally, electrons 0.3 eV above the Fermi energy are observed to have a lifetime of $\approx 700$ fs. Thus,

$\tau_0$ is experimentally determined as $\sim 5$ fs. We believe that the difference between the theoretical and experimental values for $\tau_0$ is attributable to the fact that this experiment measures only the energy relaxation time, while the theory describes the lifetime due to all $e$-$e$ collisions, and also that the theory neglects the effect of $d$-band screening.[19]

We now attempt to model the full evolution of the energy distribution function by considering the Boltzmann transport equation under the relaxation-time approximation.[2] The electron distribution function is divided into thermal and nonthermal parts,

$$f = f_{\text{nonthermal}}(E,t) + f_{\text{thermal}}[T_e(t)] \ . \tag{3}$$

The thermal part is the Fermi-Dirac distribution,[20] $f_{\text{thermal}} = [1+(E-E_F)/k_B T_e(t)]^{-1}$. The nonthermal part is a function of both electron energy and time. We choose the nascent distribution as the initial condition for the nonthermal part. It extends from the Fermi energy to 1.84 eV above the Fermi energy. The relaxation of the nonthermalized part is governed by the equation

$$\frac{\partial f_{\text{nonthermal}}}{\partial t} = -\frac{1}{\tau_0}\left[\frac{\delta E}{E_F}\right]^2 f_{\text{nonthermal}} \ . \tag{4}$$

The solution for the above equation is

$$f_{\text{nonthermal}}(\delta E,t) = f_0 \exp\left[-\left[\frac{\delta E}{E_F}\right]^2 \frac{t}{\tau_0}\right] , \tag{5}$$

where $f_0$ is the nascent distribution. Energy is transferred from the nonthermal distribution into the thermal distribution while $e$-$p$ coupling transfers energy from the thermal distribution to the lattice. The equation governing the energy changes for the thermal distribution is

$$C_e(T_e)\frac{\partial T_e}{\partial t} = -G(T_e - T_1) + \frac{\partial}{\partial t}\int (f_{\text{nonthermal}})(E^{3/2})dE \ , \tag{6}$$

where $G$ is the electron-phonon coupling constant.

The formulation of Eqs. (4)–(6) neglects the dependence of the relaxation time for $f_{\text{nonthermal}}$ on $T_e(t)$ in order to simplify the calculation.[2] As mentioned earlier, this is expected to be valid for electron energies $\delta E >$ a few times $kT_e$, e.g., for the higher energy electrons within the nonthermal part of the distribution. We also ignore the direct transfer of energy from the nonthermal distribution to the lattice because it is small compared to either of the terms in Eq. (6).

Figures 3 and 4 show the calculations for the two fluences used in the experiments assuming the excitation is instantaneous at $t=0$ fs. We have used the $G = 4\times 10^{16}$ W m$^{-3}$ K$^{-1}$ (Ref. 4) and experimentally determined value of $\tau_0 = 5$ fs. Figure 3 is calculated for an absorbed fluence of 80 $\mu$J/cm$^2$. This value was chosen to match the peak calculated $T_e$ with the peak temperature fit to the data in Fig. 1 (corresponding to the dashed line). Two important features are reproduced in the calculations. First, there is still 30% of the excitation energy in the nonthermal part of the distribution at 400 fs. Second, the full Fermi-Dirac distribution is not established until $\sim 1$ ps. In Fig. 4, the thermalization time is about half that exhibited in Fig. 3, which is consistent with Fig. 2.

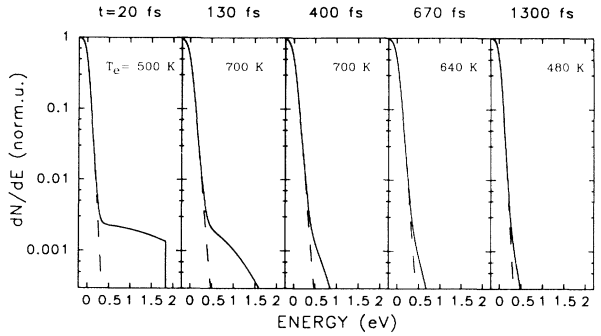


FIG. 3. Model calculations corresponding to the low fluence case shown in Fig. 1.

Recently, Kim *et al.*[21] have studied carrier-carrier scattering in an $n$-modulation-doped quantum well with a doping density $\approx 5\times 10^{17}$ cm$^{-3}$. The $n$-modulation-doped quantum well at low temperature ( $>10$ K) is also a degenerate system with Fermi energy $\approx 20$ meV. These authors have found that the dephasing time for electrons with excess energies of $\sim 1$ meV is several ps while the dephasing time for electrons of $\sim 20$ meV drops to $<400$ fs, and the results are consistent with Fermi-liquid theory. It is interesting to compare this experiment to the present one, in light of the fact that the electron densities differ by five orders of magnitude.

We first note that these two systems are very similar as far as Fermi-liquid theory is concerned. To see this, we consider the dimensionless parameter $r_s$, which determines the validity of the random-phase approximation in Fermi-liquid theory,[18,22]

$$r_s = \left[\frac{3}{4\pi n}\right]^{1/3} \frac{m_e^* e^2}{4\pi\varepsilon h^2} \ , \tag{7}$$

where $n$ is the total electron density, $m_e^*$ is the electron

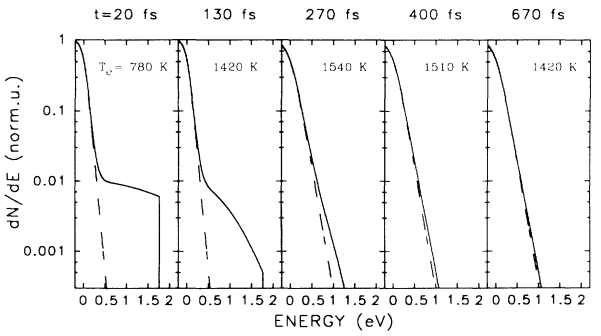


FIG. 4. Model calculations corresponding to the high fluence case shown in Fig. 2.

effective mass, and $\varepsilon$ is the dielectric constant. Physically, $r_s$ is a measure of the screening in the system. The smaller $r_s$, the larger is the screening. For Au, $n=6\times10^{22}$ cm$^{-3}$, $m_0\approx m_e$, and $\varepsilon=5.6$,[19] so $r_s\approx0.54$. For the modulation-doped quantum well, $n\approx5\times10^{17}$ cm$^{-3}$, $m_0=0.067m_e$, and $\varepsilon=13.1$, so $r_s\approx0.76$. Thus, even though their electron densities vary by five orders of magnitude, these two samples are similar systems as far as screening is concerned. We can now proceed to understand the similar relaxation times of the two samples. The relevant parameter in Fermi-liquid theory is the excess energy relative to the Fermi energy, $\delta E/E_F$. Our observed $\approx$700-fs lifetime of 300-meV excess energy ($\delta E/E_F\approx5\%$) electrons in Au is consistent with the observation of dephasing times of several ps for $\delta E\approx1$ meV ($\delta E/E_F\approx5\%$) electrons in $n$-modulation-doped quantum wells.[21]

In summary, we have measured the evolution of the electron energy distribution from the nascent nonthermal distribution to a hot thermalized distribution in an optically excited metal. A thermalization time as long as $\sim1$ ps is reported, and is consistent with Fermi-liquid theory. A model incorporating both electron-electron and electron-phonon scattering is developed to describe the relaxation of the nascent distribution, and gives good agreement with the experimental data.

We thank D. S. Kim and W. H. Knox for helpful discussions. We are grateful to A. A. Grinberg for help in setting up the theoretical model.

[1]M. Kaveh and H. Wiser, Adv. Phys. **4**, 257 (1984).
[2]N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Rinehart and Winston, New York, 1976).
[3]A. H. MacDonald, Phys. Rev. Lett. **44**, 489 (1980); P. Santhanam and D. E. Prober, Phys. Rev. B **29**, 3733 (1984).
[4]W. S. Fann, R. Storz, H. W. K. Tom, and J. Bokor, Phys. Rev. Lett. **68**, 2834 (1992).
[5]G. L. Eesley, Phys. Rev. Lett. **51**, 2140 (1983).
[6]G. L. Easley, Phys. Rev. B **33**, 2144 (1986).
[7]H. E. Elsayed-Ali, T. B. Norris, M. A. Pessot, and G. A. Mourou, Phys. Rev. Lett. **58**, 1212 (1987).
[8]R. W. Schoenlien, W. Z. Lin, J. G. Fujimoto, and G. L. Eesley, Phys. Rev. Lett. **58**, 1680 (1987).
[9]S. D. Brorson, J. G. Fujimoto, and E. P. Ippen, Phys. Rev. Lett. **59**, 1962 (1987).
[10]H. E. Elsayed-Ali, T. Juhasz, G. O. Smith, and W. E. Bron, in *Ultrafast Phenomena VII*, edited by C. B. Harris, E. P. Ippen, G. A. Mourou, and A. H. Zewail (Springer-Verlag, Berlin, 1990).
[11]S. D. Brorson, A. Kazeroonian, J. S. Moodera, D. W. Face, T. K. Cheng, E. P. Ippen, M. S. Dresselhaus, and G. Dresselhaus, Phys. Rev. Lett. **64**, 2172 (1990).
[12]M. I. Kaganov, I. M. Lifshitz, and L. V. Tanatarov, Zh. Eksp. Teor. Fiz. **31**, 2232 (1956) [Sov. Phys. JETP **4**, 173 (1957)].
[13]P. B. Allen, Phys. Rev. Lett. **59**, 1460 (1987).
[14]M. B. Agranat, S. I. Anisimov, and B. I. Makshantsev, Appl. Phys. B **47**, 209 (1988).
[15]A. Lörincz, Z. Bozóki, and A. Miklós, J. Appl. Phys. **70**, 941 (1991).
[16]R. H. M. Groeneveld, R. Sprik, and A. Lagendijk, Phys. Rev. B **45**, 5079 (1992).
[17]S. M. Sze, J. L. Moll, and T. Sugano, Solid-State Electron. **7**, 509 (1964).
[18]D. Pine and P. Nozieres, *The Theory of Quantum Liquids* (Benjamin, New York, 1966).
[19]J. J. Quinn, Appl. Phys. Lett. **2**, 167 (1963).
[20]For this purpose we ignore any time dependence in the chemical potential $\mu\approx E_F$.
[21]D.-S. Kim, J. Shah, J. E. Cunningham, T. C. Damen, S. Schmitt-Rink, and W. Schafer, Phys. Rev. Lett. **68**, 2838 (1992).
[22]R. Groeneveld, Ph. D. thesis, University of Amsterdam, 1992.