# Dynamics and Transport of Electronic Carriers in Thin Gold Films

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Thin films of gold are optically excited with ~120 fs laser pulses. The resulting nonequilibrium particle dynamics is accounted for through interaction among quasiparticles and phonons, and through particle transport as treated within Fermi liquid theory. Agreement between experiment and theory is very good.

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The advent of ultrashort duration laser pulses, particularly those in the femtosecond time domain, has engendered experiments on the dynamics of particles excited well out of thermal equilibrium. Conditions in these experiments are in stark contrast with those in the presence of longer laser pulse duration, the distinction being that at longer pulse duration (>1–10 ps) excited particles and their surroundings have had sufficient time to approach thermal equilibrium. On the other hand, at temporal resolutions of <500 fs it is possible to resolve the dynamics of nonequilibrium excited carriers and phonons, as well as particle transport.

The general field of particle dynamics and transport in metals has produced a large literature, a part of which is listed in Refs. [1] and [2]. More contributions continue to appear [3–5] attesting to the vitality of this area of condensed matter research. The major reason for conducting the present experiments is the realization that almost all modern high speed electronic contacts are thin gold films deposited by a variety of techniques. With the approach of the optical computer, the switching time will approach the femtosecond time domain and, therefore, nonequilibrium particle transport in the contacts. Thus nonequilibrium dynamics and transport define the limiting properties of the contacts.

The experimental technique applied in this investigation involves pulsed laser excitations of carriers at one (front) surface of a metallic thin film and detection of carrier evolution by a second, appropriately delayed, train of laser pulses incident at the "back" surface. The presence of excitations produces changes in the dielectric constants which, in turn, are detected by changes in the optical reflectivity $\Delta R$ at the (back) detection surface. Historically, the starting point of the analysis of experimental results (with but a few exceptions [4]) has been a set of coupled differential equations frequently referred to as the "two temperature model" (TTM) [6,7]. This formulation tracks the temporal evolution of thermal energy contained in the electron and phonon distributions starting from the time of the exciting laser pulse and in the additional presence of spatial thermally diffusive transport. In a recent critical measurement of electron emission from gold films by Fann *et al.* [8,9] it was demonstrated that excited electrons do not achieve a Fermi-Dirac distribution until after 600 fs. In a recent paper [5] we report on single crystalline and polycrystalline thin gold films optically excited with a ~100 fs "front-back" laser configuration. Thus the excitation of particles by these means must also be well out of thermal equilibrium. (For simplicity we restrict further discussion to single crystalline films.)

Figure 1 displays changes in the fractional reflectivity $\Delta R/R$ as a function of the delay time as measured at the back surface of single crystalline Au films of various thickness. The inset to Fig. 1 displays the time to traverse the film, $t_T$, as a function of film thickness for the signal to reach 15% of the maximum. We define this as the "onset" time. To a first approximation the (inverse) slope yields a constant transport velocity $v_T \sim 10^6$ m/s, which would imply unhindered, i.e., possibly ballistic transport of excited particles. All other components of the transport

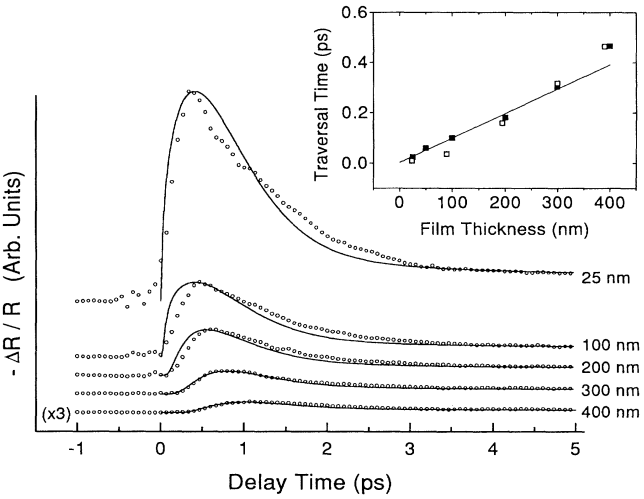


FIG. 1. Transient reflectivity $\Delta R/R$ in single crystalline gold film as a function of the delay time and for films of various thicknesses. The solid lines represent an analytic basis for the experimental results, and the open circles represent the experimental results. Note the ×3 scaling for the 400 nm sample. Inset: Traversal time of signal onsets. Solid symbols are experimental values. The solid line is a fit to experimental values and yields a transport velocity of $1.02 \times 10^6$ m/s. The open symbols are the onsets as obtained from theory.

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arrive at a later time, which implies that interactive scattering has occurred. On the other hand, $t_T$ does not vary as the second power of the film thickness as would be expected from random-walk thermal diffusion as required in the TTM. We shall test these ideas in terms of the theoretical analyses presented below.

We now demonstrate that particle-particle interactions inherent in the Fermi-liquid theory (FLT) [10], within the relaxation time approximation (RTA) [11], do account for the entire range of experimental observations. FLT treats an excitation as a "quasiparticle," that is, as an excited electron "dressed" in its own exchange hole. Under this approximation, it is the interaction among excited particles themselves, and with phonons, which determines the dynamics of the system. (In what follows the terms quasiparticles, excited particles, or just particles are used interchangeably.) The results of this approach are not only found to be more consistent with experiment but can also be readily calculated numerically. Moreover, with the single exception of one parameter, $S$, the values of parameters contained in the FLT are either obtained from the literature, the experiment, or are derived within the theory (see Table I). Even the single unknown factor is itself found to be independent of the strength of the laser excitation, the thickness of the gold film, and transport time. Agreement between theory and experiment is remarkably good.

Further experimental details have been published elsewhere [5,12] and will be only briefly discussed here. The experiment begins when a 120 fs laser pump pulse is incident on a gold film surface. The pump pulse, which possesses a wavelength of 620 nm, excites a distribution of $p$-band electrons from the Fermi edge to states up to 2 eV above the Fermi energy $E_F$. This also leaves a corresponding distribution of holes below the Fermi energy. The pump excitation is strictly intraband since the next interband state is roughly 3 eV above $E_F$ [13]. Afterwards a weak probe pulse at the same photon energy monitors the change in the reflectivity induced by transitions from the $d$-band edge to holes near $E_F$ [1]. The excitation generates quasiparticles at the front surface of the film within a 15 $\mu$m spot size and a 15 nm skin depth.

The nonthermal particle-particle interactions, outlined above, are readily handled in terms of FLT in the RTA. In this formalism, an ultrashort laser pulse produces, at time equal zero, nonequilibrium particles with an initial distribution $n_e(E, t = 0)$. The evolution of the number of excited particles, $n_e$, for $t > 0$ is governed by particle-particle ($ee$), particle-phonon ($ep$), scattering and quasiparticle generation (QG). Thus

$$\frac{dn_e(E,t)}{dt} = \left(\frac{dn_e(E,t)}{dt}\right)_{ee} + \left(\frac{dn_e(E,t)}{dt}\right)_{ep} + \left(\frac{dn_e(E,t)}{dt}\right)_{\mathrm{QG}}. \quad (1)$$

Within the random phase approximation, the first term in Eq. (1) is

$$\left(\frac{dn_e(E,t)}{dt}\right)_{ee} = -\frac{n_e(E,t)}{\tau_{ee}(E)} = -\frac{n_e(E,t)}{\tau_0}\left(\frac{E}{E_F}\right)^2, \quad (1a)$$

where FLT yields the relaxation time $\tau_{ee}(E) = \tau_0(E_F/E)^2$ with $\tau_0 = 128/\pi^2\sqrt{3}\,\omega_p$ [10]. In gold, $\omega_p = 1.25 \times 10^{16}$ rad/s and $\tau_0 = 0.6$ fs. $E$ is the excitation energy measured relative to the Fermi sea. The particle-particle interaction time $\tau_{ee}$ is a rapidly decreasing function of the excess energy $E$, and the average excess energy (as defined below) is a rapidly decreasing function of time. Therefore, the average particle-particle relaxation time $\langle\tau_{ee}\rangle \equiv \tau_0[E_F/\langle E(t)\rangle]^2$ is a rapidly increasing function of the delay time as is illustrated in Fig. 2. In other words, $\langle\tau_{ee}\rangle$ is an important contributor to the particle dynamics but only as long as $E$ is large; i.e., for only short delay

TABLE I. Optical parameters of gold used in the present analysis.

| Parameter | Value | Source |
|---|---|---|
| Optical skin depth | ~15 nm | Ref. [18] |
| Spot size | ~15 $\mu$m | [a] |
| Pump laser irradiance | 0.31 mJ/cm$^2$ | [a] |
| Laser wavelength | 620 nm (2 eV) | [a] |
| Transport velocity $v_T$ | $(1.02 \pm 0.06) \times 10^6$ m/s | Ref. [5] |
| Plasma frequency $\omega_p$ | $1.25 \times 10^{16}$ rad/s | Ref. [18] |
| Characteristic scaling time $\tau_0$ | 0.6 fs | Ref. [10] |
| Fermi energy $E_F$ | 5.52 eV | Ref. [11] |
| Average phonon energy $\hbar\overline{\Omega}$ | 7.24 meV | Ref. [19] |
| McMillan factor $\lambda\langle\omega^2\rangle$ | $23 \pm 4$ meV$^2$ | Ref. [16] |
| Quasiparticle-phonon scattering rate $\dot{q}$ | $(7.919 \pm 1.377) \times 10^{10}$ eV/s | [a] |
| Phonon-quasiparticle energy exchange factor $S$ | $(2.87 \pm 0.3) \times 10^{-3}$ | [b] |
| Average quasiparticle energy per particle $\langle E(t)\rangle$ | $aE_F(\tau_0/t)^b$ $a = 0.65$, $b = 0.59$ | [a] |
| Quasiparticle-quasiparticle scattering time $\langle\tau_{ee}(t)\rangle$ | $\tau_0[E_F/\langle E(t)\rangle]^2$ | Ref. [10] |
| Quasiparticle-phonon scattering time $\langle\tau_{ep}(t)\rangle$ | $\dot{q}/\langle E(t)\rangle$ | [a] |
| Phonon-quasiparticle scattering time $\langle\tau_{pe}\rangle$ | (31.8 $\pm$ 6.5 ps) | [a] |

[a]Present experiment.
[b]Fitted parameter.

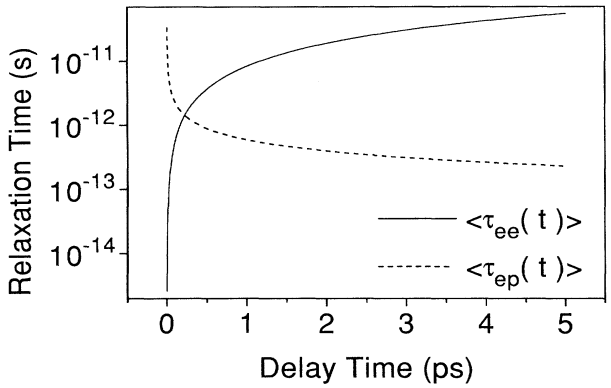


FIG. 2. Calculated $\langle \tau_{ee}(t) \rangle$ (solid line) and $\langle \tau_{ep}(t) \rangle$ (dashed line).

times. The second term in Eq. (1), for the dynamics of the particle-phonon interaction, is written as

$$\left(\frac{dn_e(E,t)}{dt}\right)_{ep} = \frac{-\dot{q} n_e(E,t)}{\langle E(t) \rangle} + \frac{S\dot{q} n_p(E,t)}{\hbar\overline{\Omega}_p}, \quad (1b)$$

in which $\dot{q}$ is the rate of energy transfer from a particle to the lattice [4,14], $\langle E(t) \rangle$ is the average particle energy [4], $\hbar\overline{\Omega}$ is the average acoustic phonon energy in gold, and $n_p(E,t)$ is the phonon number. The first term in Eq. (1b) is the rate of energy transfer from the quasiparticles to the phonon field, and the second term is the rate of energy transfer from phonons to the quasiparticles. Here $\langle E(t) \rangle$ is calculated numerically in the absence of $e$-$p$ coupling by $\langle E(t) \rangle = \int E n_e(E,t)\, dE / \int n_e(E,t)\, dE$. The phonon number is calculated using $n_p(E,t) = \sum_{t' < t}^{t} \dot{q} n_e(E,t')\, dt / \hbar\overline{\Omega}$. $S$ is a phonon-quasiparticle energy exchange factor which is observed to be independent of particle energy, time, and film thickness. It is a dimensionless parameter which, when multiplied by $\dot{q}$, represents the rate of energy returned from the phonons to the particles. The factor $S$ is the sole unknown parameter in this derivation and is determined by fitting Eq. (1) to experimental data near 5 ps. It follows from Eq. (1b) and the RTA that the first term defines an average particle-to-phonon relaxation rate given by $1/\langle \tau_{ep} \rangle = \dot{q}/\langle E(t) \rangle$. Similarly the second term defines an average phonon-to-quasiparticle relaxation rate which may be written as $1/\langle \tau_{pe} \rangle = S/\langle E(t) \rangle / \tau_{ep} \hbar\overline{\Omega}$. Thus $\langle \tau_{pe} \rangle$ can be calculated once $S$ is known. At this point we adopt [14,15] McMillan's electron-phonon energy transfer coefficient $\lambda\langle \omega^2 \rangle$ to calculate $\dot{q} = \pi\hbar\lambda\langle \omega^2 \rangle / 2\ell n 2$ [4]. For gold $\lambda\langle \omega^2 \rangle = 23 \pm 4$ meV$^2$ [16] and $\dot{q} = 7.919 \times 10^{10}$ eV/s.

The last term in Eq. (1) describes the quasiparticle generation rate in which an excited particle of momentum $\vec{p}$ is scattered, via the screened Coulomb potential, against ground state electrons, creating a new excitation $\vec{p} + \vec{q}$ and an additional particle-hole pair of momentum $-\vec{k} - \vec{q}$ and $-\vec{k}$ [10]. Thus three particles are produced per scattering event. The probability that a new quasiparticle lies in an interval between $E'$ and $E' + dE'$ is given by $P(E', E) = 2(E - E')\, dE/E^2$ [17]. Multiplying this by Eq. (1a) and summing over all energies yields the rate of quasiparticle creation

$$\left(\frac{dn_e(E,t)}{dt}\right)_{QG} = 6 \int_E^{\infty} dE' \frac{n_e(E',t)}{\tau_0} \frac{E' - E}{E_F^2}. \quad (1c)$$

In the derivation it is assumed that the number of excited holes below $E_F$ equal that of the quasiparticle number with $E > E_F$.

At this point it is useful to imagine a cylinder which extends through the thickness of the film with end faces of 15 $\mu$m diameter and length equal to the thickness of the film. Particle-particle interactions occur within the cylinder and, simultaneously, particles are transported down the concentration gradient that exists between the excited and unexcited volumes of the film. We assume that the excited particles are equally likely to travel in all directions between scattering events and, noting that the areas of the ends of the cylinder are nearly $10^3$ times greater than the area of its sides, then the flux of particles leaving the sides of the cylinder is much smaller than the flux that travels roughly parallel to the axis of the cylinder. Thus to a first approximation, the transport is one dimensional. Thus we introduce as an ansatz the expression for spatial transport to be proportional to a linear gradient of $n_e$ with $z$

$$\frac{dn_e(z,t)}{dt} = v_T \frac{\Delta_k n_e(z,t)}{\Delta z}. \quad (2)$$

In order to obtain a numerical description of the particle transport, the thickness of the film is divided into a series of slices along $z$ from the front to the back surface. The position $z$ is defined for the $k$th slice to be $z = k\Delta z$ and $\Delta_k n_e(z,t)$ is the net change in the number of excited particles in the $k$th slice due to the gain of particles from the $k - 1$ slice and the loss of particles into the $k + 1$ slice. Equations (1) and (2) are then solved iteratively in each slice of the film using $S$ as the only fitting parameter. Each iteration of Eq. (2) then determines the transport through all the slices, including the back surface. The density of carriers is then calculated down to one skin depth from the back surface. The results of this calculation are shown as solid curves in Fig. 1. The value of $S$ averaged over the various films is found to be $2.87 \times 10^{-3}$ and $\tau_{pe} = 31.8$ ps. The slow phonon-particle decay implies that although a large number of phonons are produced through decay of excited particles only a small fraction of these return energy to the particles. Note the excellent agreement between the experimental data and the theoretical basis. A further result of the numerical solution of Eqs. (1) and (2) is the values of various parameters of FLT and the RTA as listed in Table I. Moreover, the following functional relationships are recoverable from the calculations, namely, $n_e(E,t)$, $n_p(E,t)$, $\langle \tau_{ee}(t) \rangle$, $\langle \tau_{ep}(t) \rangle$, $\langle \tau_{pe} \rangle$, and $\langle E(t) \rangle$.

The major components of the particle dynamics and transport, as displayed in Fig. 1, can now be identified as

follows: (i) The variation in the onset of the signal reflects the time delay required by a particle to traverse, essentially unhindered, the various film thicknesses. (ii) Excited particles lose energy by interactions with ground state particles and with phonons, (iii) and the steeply rising part of the signal is governed by the creation of excited particles. (iv) The decreasing part of the signal reflects the dominance of the particle-phonon interaction, including the inverse effect of the phonons on the particles. (v) The very slowly decreasing tail of the signal arises from the very long scattering time $\langle \tau_{ep} \rangle$ at these delay times. (vi) Equation (2) describes the particle transport as simply driven by a one-dimensional, particle concentration gradient.

We return to a discussion of the transport mechanism under which it is assumed that at least a part of the particles travel unhindered across the film. In order for this to happen, the average time between scattering events would need to be long compared to the traversal time $t_T$. The pertinent information is contained in Fig. 2. Approximately one femtosecond after the laser's excitation we find $\langle \tau_{ee} \rangle \sim 2.6$ fs and $\langle \tau_{ep} \rangle \sim 33.5$ ps. Thus strong $e$-$e$ scattering occurs during this interval. Randomization of the particle momentum may also occur during this time. For the thinnest samples (25 nm), for which the traversal time is about 0.02 ps, we find $\langle \tau_{ee} \rangle$ is $\sim$0.08 ps and $\langle \tau_{ep} \rangle$ $\sim$5.8 ps. We compare this to the thickest samples (400 nm) where $t_T \sim 0.43$ ps, $\langle \tau_{ee} \rangle \sim 3.1$ ps, and $\langle \tau_{ep} \rangle \sim 1$ ps. We conclude that $e$-$e$ interactions are the dominant scattering mechanism very soon after laser excitation. However, this interaction is diminished after 0.02 ps, whereas $e$-$p$ interactions do not become prominent until about 0.43 ps.

We have demonstrated that FLT, within the RTA, together with simple 1D transport, accounts for the experimental results. The theoretical basis predicts that throughout the period of observation, nonequilibrium, nonthermal, particle-particle interaction dominates the dynamics and transport. Thus we conclude, at least in the femtosecond domain, that at no time during the present experiment is the system of excited particles at equilibrium with a thermal bath, nor do its components follow thermal diffusive transport.

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