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Electron Transport Phenomena and Dense Plasmas Produced by Ultra-Short Pulse Laser Interaction

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ABSTRACT

Recent experiments with femtosecond lasers provide a test bed for theoretical ideas about electron processes in hot dense plasmas. We briefly review aspects of electron conduction theory likely to prove relevant to femtosecond laser absorption. We show that the Mott-Ioffe-Regel limit implies a maximum inverse bremsstrahlung absorption of about 50% at temperatures near the Fermi temperature. We also propose that sheath inverse bremsstrahlung leads to a minimum absorption of 7-10% at high laser intensity.

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Fsec laser and transport processes

A new generation of high power short-pulse lasers was made possible by the technique of chirped pulse amplification.(1) These lasers generate pulses in the femtosecond range, typically 100 fsec, and can be used to prepare a well-characterized solid-density plasma. Recent femtosecond laser experiments give data concerning electron transport in hot dense matter, data valuable precisely because of the simplicity of the experimental conditions.

Electron transport, which we take to include AC electrical conduction as well as heat conduction, is a key to understanding the laser interaction. The AC conductivity determines the absorption and controls how much energy is deposited in the target. Heat conduction determines how far the energy spreads and this determines the temperature.

Typical parameters for a femtosecond pulse laser are: total laser energy, 0.1 Joule per pulse; pulse length, 100 fsec; power, 1 terawatt; focal spot diameter as low as 10 microns; laser intensity, 10^{12} - 10^{18} Watts/cm^2; wavelength in the visible and repetition rate above one shot per second. Most crucial to control of the target interaction is the contrast between peak power in the pulse and the background laser emission ("prepulse"); for a well-adjusted laser this contrast can be better than 10^4, and after harmonic conversion the contrast improves to 10^8.

As we understand it, the laser interacts with the target over a skin depth of 100 A. The absorbed energy is carried into the target to a depth of ~ 500 A during the laser pulse by an electron thermal wave. At high intensity, the target expands as much as 100 A during the laser pulse, a velocity of 100 km/sec. At low intensity there is virtually no expansion during the heat pulse. The peak electron temperature ranges from 1 eV at low intensities to 1000 eV at the highest intensities. The absorbed energy is transformed into electron thermal energy, ionization energy, ion thermal energy, kinetic energy of hydrodynamic motion, magnetostatic energy and x-rays emission.

Femtosecond laser targets are not in thermal equilibrium. The electron temperature is higher than the ion temperature. The ionization is usually less than equilibrium (LTE) or even than a steady collisional-radiative non-LTE ionization state and the radiation is much less than an equilibrium (black-body) distribution. However the collision rates probably suffice to bring free electrons to a local equilibrium distribution.

Femtosecond laser experiments explore the transition from solid state to plasma and the transport phenomena must be examined from viewpoints traditionally associated with these two distinct phases of matter.

How does solid matter become a plasma?

The ideal solid is translationally invariant and its elementary excitations are labelled by wave-vector k or crystal momentum p. The atomic vibrations are travelling sound waves or phonons. Electron excitations are described in terms of energy bands and Bloch functions. Inner-shell electrons are associated with narrow bands of propagating states, and even electrons far above the Fermi level can be Bragg-reflected and therefore have energy gaps at the Brillouin zone boundaries. Bragg-reflection gives the electrons an effective mass $m^*(k)$ which can be significantly different from the usual electron mass.

The dense plasma is disordered; there is short-range ion correlation but no long-range order. Because of this phonons no longer propagate very well. Free electrons have energies defined in k-space, but bound electrons are localized on specific ions. The ions have definite charge states whose probabilities are determined by a Saha equation. The electrons make transitions corresponding to excitation and ionization processes, but the zero-order Hamiltonian is diagonal in a representation which assigns integer charge to each ion rather than one which assigns a definite momentum to each electron.

How do solid-state energy levels evolve into those of the plasma? The effect of temperature, aside from disordering the ion positions, is to excite electrons to states of higher energies. Quite generally, excited electrons (bound or free) are less able to screen the nuclear charge than they were in states of lower energy. This means the self-consistent electrostatic potential grows stronger as electrons begin to be thermally excited. In turn, this increases the spacing of energy levels and moves lines to shorter wave-lengths. What is the change in the electron band-gaps?

It seems clear that as temperature rises the core bands become very narrow, both as a result of the stronger potential and as a consequence of disorder, which spoils the resonant hopping required for propagating states. However, band gaps at energies above the original Fermi energy evidently decrease with loss of Bragg scattering even if the potential ultimately becomes stronger. This is seen in the optical properties of liquid metals. (2) In particular, conduction electrons in liquid metals (and presumably in hotter plasmas) respond with essentially the free-electron mass.

There remain questions how ionic and covalent materials behave as their temperature is raised. To a certain extent one can make predictions based on the room-temperature band-structure. On the high-temperature side one has the usual plasma picture. In the range between , there are various possibilities of charge transfer, excitonic phases, etc. For example, room temperature glass consists of Si(4+)O(2-) ions. Partway to the plasma state, there will doubtless be some free electrons, i.e., electrons in 3s,3p conduction bands, but we do not know how the core charge states vary.

Electron Conduction Theory

The classical theory of Brownian motion shows the connection between diffusion and frictional drag forces which determine the mobility (u = velocity/force) for an electron. Already this simple theory gives an expression for the AC conductivity of nondegenerate electrons,

$$\sigma(\omega) = ne^2/m < \tau/(1 - i\omega\tau) > \qquad (1)$$

where n = electron number density, e = electron charge (cgs), m = electron mass, and $\tau = m\mu$ = electron collision time. The average is taken over the velocity distribution when τ depends on electron energy.

In the 1930's Sommerfeld and Frank studied electron transport in simple metals using the Boltzmann equation with Fermi-Dirac statistics for the unperturbed distribution function. This theory also gives Eq. (1) for degenerate or nondegenerate electrons (the average is weighted by E df/dE, the derivative of the equilibrium distribution function).

When we use Eq. (1) to calculate absorption, we must solve Maxwell's equations for the evanescent laser wave penetrating into the target over a skin depth $\delta = c/\omega_p$. This calculation gives a useful qualitative formula for the absorbed energy fraction(3):

$$A = 2/\omega_p\tau \qquad (2)$$

Here ω_p is the electron plasma frequency and τ is an average collision time (strictly, $1/\tau = < 1/\tau(E) >$ is the appropriate average). Eq. (2) describes steady-state absorption of a homogeneous laser beam by an idealized homogeneous target, so ω_p and τ do not depend on position, and applies in the high-frequency limit $\omega\tau >> 1$ (this is the case at high target temperatures). The target is assumed to have a sharp interface and to be overdense to the laser ($\omega << \omega_p$). Eq. (2) should be corrected for hydrodynamic expansion of the target, temperature gradients, the spatial beam profile and the time-dependence of laser energy and target temperature. These corrections are straightforward aspects of a numerical simulation.

Another theory of laser absorption is given by the radiative kinetic equation which describes inverse bremsstrahlung transitions (absorption and stimulated emission). It is not difficult to show, using the Kramers cross-section for radiative rates, that the radiative kinetic equation gives the same result as Eq. (1) for conditions $\omega\tau >> 1$ when $\hbar\omega << kT$ (non-degenerate electrons) or $\hbar\omega << E_f$ (degenerate electrons, discussed further below). The agreement with Eq. (1) requires that we use a high-frequency Coulomb logarithm, $\ln \Lambda = \pi/\sqrt{3}$ in the evaluation of Eq. (1). For lower frequencies ($\omega\tau < 1$) one must correct the radiative kinetic equation to include interference in electron-ion multiple-scattering (4), and then one obtains again Eq. (1). At high frequencies ($\hbar\omega >> kT$ or $\hbar\omega >> E_f$), the radiative kinetic equation gives a different answer discussed below.

Starting with a quantum equation of motion for the density matrix, Kohn and Luttinger(5) derived a transport theory for nondegenerate electrons. The equation of motion is expanded with respect to a DC applied electric field (linear response) and also with respect to the perturbation due to randomly located ions. When averaged over ion positions, the diagonal elements of the density matrix correspond to the Boltzmann distribution function and are larger , in a certain sense, than the off-diagonal elements. The diagonal elements are governed by an equation essentially equivalent to the Boltzmann equation.

Kubo recast the density-matrix equation of motion to obtain a general formal expression for the electrical conductivity(6), relating the dissipation produced by Joule heating to fluctuations in the spontaneous currents existing in thermal equilibrium in the absence of the applied field ("Fluctuation-dissipation theorem").

Another systematic theory of conduction phenomena is given by the Green's function perturbation theory (7). This theory reproduces all the previous results and is readily extended to the BCS superconducting state. Disordered materials with strong scattering pose a problem of high-order perturbation theory; one method for treating this case is the coherent potential approximation (8).

In research on heavily doped semiconductors a rule has emerged, known as the Mott-Ioffe-Regel limit, to the effect that the minimum electron mean free path is approximately the atomic spacing(9). With this rule goes a minimum electrical conductivity and an explanation for any smaller values of the conductivity: that would require localized electrons unable to propagate.

Lee and More (10) propose this limit applies to laser plasmas at temperatures of $kT \cong E_f$ because these are the conditions of maximum electron-ion scatttering. (Localized conduction electrons are very unlikely in a high-temperature system.)

The condensed-matter theories mainly discuss electron scattering by screened ion potentials. In plasma physics the Coulomb potential gives a long-range interaction which is traditionally treated by the Fokker-Planck approximation, applied by Spitzer and Harm(11), Rosenbluth et al. (12) and others. For dense plasmas the Coulomb collisions can also be handled by the method of correlation functions, i. e., the Ziman formula.

The Ziman formula shows how interference between scattering on different centers limits the electron mean free path. In particular the Ziman formula gives the small-angle cutoff required for a finite Coulomb logarithm.(13) The simplest version of this formula is

$$\frac{1}{\tau} = n_i v \frac{\pi}{k^4} \int_0^{2k} q^3 \frac{d\sigma}{d\Omega} S(q) dq$$

Here $\hbar k$ = electron momentum, $\hbar q$ = momentum transferred in the collision, $S(q)$ = ion structure factor, $d\sigma/d\Omega$ = electron-ion differential scattering cross-section. The formula is derived with the Born approximation for this cross-section, but the Born approximation is rarely justified except for fully-ionized or low-Z plasmas. When a higher-order cross-section is used one has kept some terms in a multiple-scattering series (and ignored others). For many years it has been known that an expression like Eq. (3) should be used for inverse bremsstrahlung absorption(Ichimaru, ref. 14).

Much of the recent literature on transport in strongly coupled plasmas consists of evaluating the Ziman formula with various approximations for the structure factor S(q), and various treatments of the electron screening of the ion potential. In this context we mention work of Ashcroft and Schaich(15), Hansen et al.(16), DeWitt et al.(17), Ichimaru et al.(18), and Perrot and Dharma-Wardana(19). Because most of these authors are present to describe their own work, we do not need to summarize these important developments further.

Some Points to Discuss

1. Momentum Transfer cross-section

To obtain the collision time τ which appears in transport theory the electron-ion differential cross-section must be weighted by $(1-\cos\theta)$, where θ is the scattering angle. The importance of collisions is determined by the momentum-loss they cause.

The factor $(1 - \cos\theta) = q^2/2k^2$ appears explicitly in the Ziman formula. For Brownian motion this effect emerges from a treatment of correlated random walks. In the Green's function theory, it appears as the difference between the average of a product of Green's functions and the product of the averages(7). Kubo derives this effect from a memory-function in his Colorado lectures(6). All transport theories include this effect, so it is not a difficult question.

2. Landau-Peierls' effect.

The intuitive derivation of the Boltzmann equation seems to fail under conditions where $\hbar/\tau > kT$, because in that case one cannot defend the picture of a small wave-packet traveling between collisions. The inequality is satisfied in low-intensity femtosecond laser interactions for many materials. Theoretical analyses from various viewpoints(e.g., ref. 5) have shown there is no Landau-Peierls' effect, i.e., no quantum correction to Eq. (1) when $(\hbar/\tau > kT)$. The nature of the transport in these circumstances is close to Brownian motion of large electron wave-packets and we began with the observation that Eq. (1) governs this case.

3. Quantum AC transport

The question here concerns high frequencies, $\hbar\omega > kT$. The Kubo formula and the radiative kinetic equation contain a factor $[1-\exp(-\hbar\omega/kT)]$ in which the first term (unity) describes absorption and the second term describes stimulated emission. Eq. (1) has no such factor. Does this mean the Boltzmann equation fails?

For nondegenerate electrons the quantum theories give a conductivity equal to a factor $(kT/\hbar\omega)[1 - \exp(-\hbar\omega/kT)]$ times the result in Eq. (1). When $\hbar\omega << kT$ this factor reduces to unity and we have again Eq. (1).

For degenerate electrons, the quantum theories give a more complicated result, and it is quite remarkable that this agrees with Eq. (1) up to photon energies of the order of the Fermi energy, much larger than kT. It is well-known that Eq. (1) gives a good description of liquid metals interacting with visible light.(2) Thus the quantum correction only affects the ultra-violet or soft x-ray absorption.

4. Electron-electron collisions

The greatest technical difficulties of conduction theory are associated with electron-electron collisions. In the Boltzmann transport theory one has a nonlinear integro-differential equation for the perturbed part of the distribution function. The equation simplifies to a Fokker-Planck approximation, expressed by the Lenard-Balescu collision operator, but still requires a nontrivial numerical calculation.

Fortunately we are mainly interested in target plasmas having high ion charge states -- for example, aluminum has $Z^* = 11$ for intense laser pulses. In this case the relative importance of electron-electron scattering is small, proportional to $1/Z^*$, and we simply neglect electron-electron collisions in view of the uncertainties in other aspects of the modelling.

Is the electron-electron interaction even a $1/Z^*$ correction? In degenerate matter it is well-known that e-e collisions are inhibited by the exclusion principle (two electrons within kT of the Fermi level must find final states also in this narrow energy range).

There is also the subtlety that when electrons carry a given current and a pair of electrons collide, their individual momenta change, and the distribution function f(v) changes, but there is no change in the total current of the electrons. Electron-electron collisions have an indirect effect, for example, moving an energetic electron down to lower energies where its next electron-ion collision will happen sooner. Even this effect is partly cancelled because, while reducing the energy of one electron, the electron-electron collision raises the energy of the collision partner.

Thus the high-energy part of the distribution is affected by electron-electron collisions, and while this plays an important role in the DC conductivity and/or the heat conduction, Eq. (1) shows that the high-frequency AC conductivity is less sensitive to high-energy electrons and this is another reason to omit e-e collisions from a laser absorption model.

5. Minimum Mean Free Path

The range of temperatures near the Fermi energy represents the boundary between solid-state and plasma physics. In this case the electron mean free path must be very short because one has strong lattice disorder and the conduction electrons have enough excitation that they can lose energy by exciting other electrons from the Fermi sea. Even in cold matter, electrons with energies a few eV above E_f have mean free paths no more than a few atomic diameters. At higher temperatures electrons have reduced Coulomb cross-sections but this effect is not yet significant at temperatures near E_f.

It is also evident that despite the strong disorder electrons are not localized or trapped because they are constantly exposed to small energy transfers from other free electrons. Thus we expect the collision time to be essentially that associated with a mean free path of $2\,R_o$ (i.e., one atomic diameter), (10)

$$\tau = 2\,R_o/v_F \qquad (4)$$

If we combine this approximation with Eq. (2) for the absorption, we find a maximum inverse bremsstrahlung absorption,

$$A_{max} = 1.407\,(a_o n)^{1/6}\,(Z^*)^{-1/3} \qquad (5)$$

where $a_o = .529\ 10^{-8}$ cm is the Bohr radius, and the numerical value for aluminum is $A_{max} = 53.3\%$ (assuming a valence of 3).

The weak dependence on electron density is the most striking feature of Eq. (5). If there is no other absorption mechanism, Eq. (5) predicts a maximum absorption of 50% at temperatures near the Fermi temperature for all target materials. (It is assumed that insulators break down to a metallic state with a well-defined electron density and Fermi temperature.) At higher and lower temperatures the mean free path is longer and the absorption is lower.

Eq. (5) is not a rigorous limit on the inverse bremsstrahlung absorption because of the corrections discussed in connection with Eq. (2). However any material showing absorption significantly above Eq. (5) is probably showing an additional absorption mechanism (such as line absorption).

6. Nonlinear phenomena

Whereas the Kubo formula claims to give the exact linear response conductivity, when we think about laser plasmas we rapidly leave the linear regime. The AC electric field of the laser is of the order of a billion volts per centimeter and the temperature gradient is more than a kilovolt per micron or 10^{11} Kelvin per centimeter. Even referred to voltage or temperature changes over one mean free path these numbers are large enough to cast doubt on the application of linear-response theory.

One specific nonlinear-response phenomenon is the Silin effect: at high laser intensity, the electron quiver velocity is larger than the thermal velocity and should replace it in the collision cross-sections (leading to a smaller collision rate). There is every reason to believe this effect occurs in high-power interaction with low-density plasmas. For femtosecond laser interactions, however, the incident laser field is partly cancelled by a reflected wave leading to a smaller electric field, and smaller quiver velocity, and the high target density implies a high collision rate, so the quiver energy is thermalized and the thermal velocity keeps up with the quiver velocity.

The Fourier law for heat conduction breaks down in very high temperature gradients, another nonlinear response phenomenon. It is replaced by a nonlocal conduction process which usually implies a maximum heat current, $q = f\,n\,v\,kT$, where n = electron density, v = electron thermal velocity, T = electron temperature and f is a constant which should be 0.6 on the Knudsen model (free streaming electrons). There is an extensive literature based on modeling laser-plasma experiments with inhibited heat conduction, meaning values of f as low as 0.05. For the moment the femtosecond laser experiments seem to be compatible with the theoretical value, f = 0.6.

A related issue is the possible existence of non-Maxwellian electron distributions. These can be caused by atomic processes (depletion of energetic electrons by ionization) as well as by the laser absorption itself or by escape of the most energetic electrons. Because the Coulomb cross-section depends strongly on electron energy, a depletion of the high-energy portion of the distribution would strongly affect the heat transport coefficients. However in femtosecond laser plasmas, the laser absorption pushes the distribution out of equilibrium only over the skin depth while thermalization can occur throughout the plasma heated by the electron thermal wave.

Finally we mention instability effects, for example, a mechanism by which electrons generate ion sound waves when they carry a large enough current to have a supersonic drift velocity. Such effects have a threshhold and are not included in the linear-response formalism.

7. Geometric Effects

It has been suggested that the anomalous skin effect could occur in femtosecond laser plasmas if the electron mean free path were to exceed the skin depth. This could, hypothetically, occur at high laser intensities where the target temperature reaches 1 keV in aluminum. However the criterion for the anomalous skin effect is two-fold: the ratio of mean free path to skin depth must exceed unity but also must exceed $\omega\tau$. This second condition is very hard to satisfy in a femtosecond laser-heated target, and for this reason we are skeptical that the anomalous skin effect occurs in these plasmas.

The author has proposed a different surface-related absorption mechanism, Sheath Inverse Bremsstrahlung, which generalizes the surface-assisted absorption mechanism of Holstein. This mechanism applies for very high temperatures (1 keV) on targets which still have a sharp interface with the vacuum, and which may again correspond to conditions difficult to achieve. For the ideal sharp interface, the theory gives an extra absorption

$$A_{sib} = 1.6\, v/c \qquad (6)$$

Here $v = (kT/m)^{1/2}$ is the average electron thermal velocity. The formula predicts as much as 10% absorption at keV temperatures.

Finally, it should be mentioned that the laser focal spot is almost certainly surrounded by a large toroidal magnetic field. While preliminary estimates show this field does not greatly alter the conductivity responsible for absorption of the laser, more thorough study of magnetic effects is certainly required.

Practical Theory of Conduction Phenomena

The comprehensive conductivity model of Lee and More (10) combines a semi-empirical solid/liquid conductivity, the high-temperature Spitzer result, and a minimum conductivity obtained from Eq. (4). It is in general agreement with several laser reflectivity experiments, when combined with appropriate modelling of the hydrodynamics.(21) It is essential that the AC conductivity be calculated from the DC model by Eq. (1) as written to obtain good agreement with experiment. The new experiments with shorter laser pulses and higher intensities will give much more conclusive tests of all these ideas about conduction phenomena, just because of the reduced importance of the hydrodynamic corrections to the raw data.

It remains to mention several other comprehensive practical models for conduction coefficients developed by Rinker(22), Drska and Vondrasek(23), and Kalitkin and Ermakov(24).

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