Resistivity of a Simple Metal from Room Temperature to 10^6 K

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The resistivity of nearly solid-density Al was measured as a function of temperature over 4 orders of magnitude above ambient by observing the self-reflection of an intense, < 0.5 psec, 308-nm light pulse incident on a planar Al target. As an increasing function of electron temperature, the resistivity is observed initially to increase, reach a maximum which is relatively constant over an extended temperature range, and then decrease at the highest temperatures. The broad maximum is interpreted as "resistivity saturation," a condition in which the mean free path of the conduction electrons reaches a minimum value as a function of temperature, regardless of the extent of any further disorder in the material.

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We report the first experimental study of the electrical resistivity of a solid-density material, in this case a simple Drude metal, over an extended range (4 orders of magnitude) of elevated temperature with *little or no change in its density*. The results show three general regions of the dependence of the resistivity on the temperature: Initially, the resistivity increases with increasing temperatures, reaching a relatively constant value that extends over a wide temperature range, and then it decreases as the temperature is further increased. We argue that these regions reflect differing mechanisms controlling the mean free path of the conduction electrons in different temperature ranges. In particular, the region of maximum resistivity is a result of "resistivity saturation," a condition in which the electron mean free path reaches a minimum value, independent of the degree of material disorder.

The ability to study the resistivity of a well characterized solid-density material over a great range of elevated temperatures was made possible here by the use of ultrashort (< 0.5 psec), relatively high-energy (0-5 mJ) laser pulses. The self-reflection of a laser pulse, focused onto a smooth target at fixed pulsewidth and spot size, was monitored over 4 orders of magnitude in energy. Frequency shifts of the reflected light were also recorded, and as discussed in detail below, these frequency shifts are shown to arise directly from the expansion velocity of the solid-vacuum interface. From the dependence of the interface velocity upon the laser intensity, we were able to determine the electron temperature and degree of interface expansion for each recorded value of the reflectivity. This information is sufficient to determine the resistivity of solid-density Al as a function of temperature up to 10^6 K.

It is important to differentiate between the type of heating-reflectivity experiment reported here and those conducted with high-energy (> 100 mJ), long-duration (> 50 psec) pulses. In the latter, the majority of the energy is absorbed not by the dense target, but rather by the material expanding away from the interface over a scale of many wavelengths. Detailed hydrodynamic calculations are usually required to analyze the data from this type of experiment,[1] and no simple interpretation in terms of the solid-density characteristics of the targets can be given. In sharp contrast, our experiments use such a short pulse length that the interface expansions during the coarse of the reflectivity measurement never exceed $\approx \lambda/15$, even for the highest intensities. Although this amount of expansion is on the order of the low-intensity skin depth and cannot be automatically ignored in the data analysis, we show that it is readily accommodated by a simple model that ties directly to the physical parameters of the solid-density target.

Pulses of light from a synchronously mode-locked oscillator operating at 616 nm and compressed to 400 fsec were amplified to ≈ 0.6 mJ at 10 Hz, doubled in potassium dihydrogen phosphate (KDP) to 308 nm and amplified to two XeCl excimer amplifiers up to 7 mJ/pulse. The overall amplified-spontaneous-emission (ASE) component of the total laser energy output was measured to be less than 1 part in 10^3, a critical consideration, for we found that as little as 1% fractional ASE on the leading edge of the pulse was sufficient to decrease greatly the measured reflectivity, presumably because of the production of long-scale-length vapor or plasma in front of the target in advance of the arrival of the ultrashort heating-reflecting pulse.

The linearly polarized pulses were focused with a 200-mm lens ($f/10$) to a spot area of 10^{-5} cm^2 in vacuum. The targets were mounted at 45° with respect to the beam and the position of the tightest focus was adjusted and monitored with the time-integrated x-ray energy yield from the plasma as a guide. Microscopic analysis of targets confirmed that the tightest focus resulted in the largest x-ray yield for a given laser energy and pulsewidth. Data were taken at 10 Hz with target

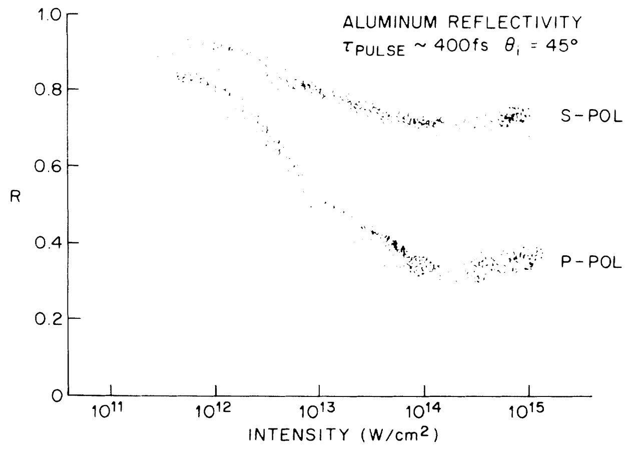

FIG. 1. Self-reflection as a function of intensity of S- and P-polarized light from an aluminum-vacuum interface for a pulse of light near 308 nm with a duration of less than 400 fsec as a function of intensity. Each point is the result of a reflection measurement for a single laser pulse.

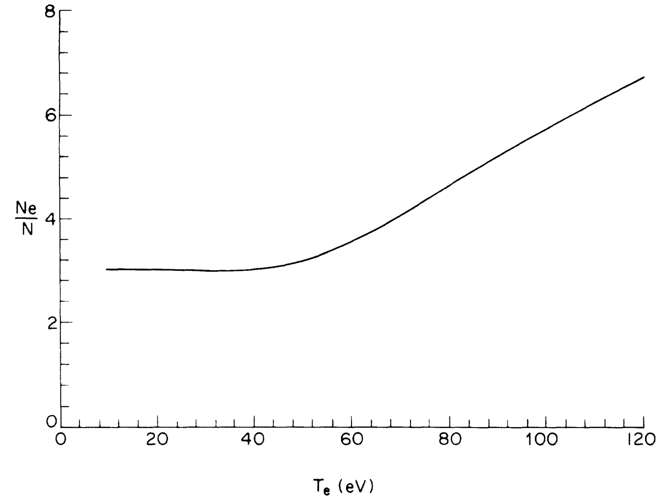

FIG. 2. Calculation of N_e/N for aluminum as a function of electron temperature for $N \approx 6.6\times10^{22}$ cm^{-3} with the assumption of local thermal equilibrium (LTE).

translation such that each pulse encountered a smooth surface. The targets were constructed of vacuum-deposited films of aluminum on glass substrates. The oxides surfaces on the targets played no role because their thickness (≤ 20 Å) is less than 10% of the optical skin depth.[2] The measured reflectivities were found to be independent of the target thickness for thicknesses greater than 400 Å.

Figure 1 shows the reflectivity of S- and P-polarized (**E** vectors perpendicular and parallel to the plane of incidence, respectively) light pulses of a 400 fsec duration and a 400-Å-thick Al film for intensities between 5×10^{11} and 10^{15} W/cm^2. The reflectivity shows three distinct regions as a function of intensity: an initial drop for intensities $\leq 10^{14}$ W/cm^2, a minimum value extending over a factor of 3 in intensity, and an increase in the reflectivity beyond $\approx 3\times10^{14}$ W/cm^2.

In addition to the reflectivity, we recorded the spectrum of the reflected light on a shot-to-shot basis using an optical multichannel analyzer. As a function of increasing laser intensity, the spectrum broadened and *shifted* to higher frequencies. Because of the extreme electron heating rates induced by the laser, there are two possible interpretations of the observed frequency shifts: self-phase modulation[3] arising from the rapidly changing index of refraction, and mass motion normal to the target. The effects of self-phase modulation can be readily calculated by the assumption of Gaussian temporal laser pulses, and reasonable, but not critical, estimates of electron heat capacity, collisionality, and density: The shifts due to self-phase modulation are both opposite in direction to and negligible in size compared to the experimentally observed shifts. In contrast, the observed shifts are entirely consistent, both in magnitude, sign, and broadening, with motion of material away from the target. The average directed velocity of this motion is given[4] by $v_0^2=[2/(\gamma-1)]^2ZkT_e/m_i$ for unsteady flow into vacuum. Here, m_i is the ion mass, Z is the effective ion charge (determined in a self-consistent manner from Fig. 2), and $\gamma=C_p/C_v=1.7$, where $C_{p(v)}$ is the electron heat capacity at constant pressure (volume).[5] Using the frequency-shift data, we were thus able to correlate the electron temperature in the target with the intensity of the laser. This correlation is shown in the abscissa of Fig. 3.

We have taken the ac electrical properties of solid-density Al to be described by the Drude model, an assumption based on the nearly free-electron Fermi surface of Al (Ref. 6) and its lack of interband transitions at 308 nm. In this model, the complex index of refraction, n, is given by $n^2=1+i4\pi\sigma/\omega$, where σ is the complex conductivity such that $\sigma=\sigma_r+i\sigma_i$, with $\sigma_r=(\nu/4\pi)\xi$ and $\sigma_i=(\omega/4\pi)\xi$, with $\xi=\omega_p^2/(\omega^2+\nu^2)$. Here ν is the generalized electron collision frequency, ω is the laser frequency, and ω_p is the plasma frequency ($=4\pi N_e e^2/m_e$). The factor ν is the physical parameter of interest and describes all collisional processes that remove energy from the electromagnetic wave; ν is related to the dc resistivity by

$$\rho_{dc}=\nu m_e/N_e e^2. \qquad (1)$$

Under the conditions of a step function, the Fresnel equations[7] relate the complex index of refraction to the reflectivity of a homogeneous lossy medium, and the parameter ν is obtained from the reflectivity data by standard numerical inversion.

When the vacuum-material interface is not sharp (i.e., when it is not substantially less than the skin depth), the Fresnel equations cannot be used to relate the reflectivity to the material parameters, and the Helmholtz wave

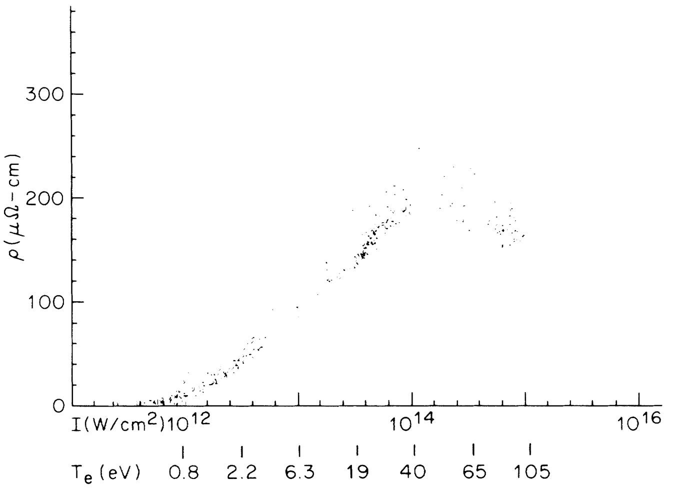

FIG. 3. Resistivity of aluminum vs electron temperature. This result is obtained by iteratively solving the Helmholtz equations [Eqs. (2) and (3) in the text] numerically to obtain the value ν that reproduces the P-polarized reflectivity data in Fig. 1. (Similar results are obtained from the S-polarized reflectivity data.) The temperature axis is determined by Doppler shifts of the reflected light. As discussed in the text, the maximum in the resistivity near $T_e \approx 40$ eV is interpreted as a manifestation of resistivity saturation.

equation must be explicitly solved for each expansion scale length. There exist well known analytic solutions for $\nu/\omega \ll 1$ when the scale of the expansion is greater than several wavelengths,[8] but in our case not only is $\nu/\omega \approx 1$, but the expansion is at most $\lambda/15$. Thus the Fourier-transformed wave equations for S-polarized light,

$$\frac{d^2E}{dx^2} + k^2(n^2 - \sin^2\theta)E = 0\,, \tag{2}$$

and for P-polarized light,

$$\frac{d^2B}{dx^2} + k^2(n^2 - \sin^2\theta)B - i\frac{4\pi}{ckn^2}\frac{d\sigma}{dx}\frac{dB}{dx} = 0\,, \tag{3}$$

must be solved numerically. Here E and B are the complex electric and magnetic field vectors, x is the direction of the material density gradient, θ is the angle of incidence, k is the vacuum wave number, and σ is the complex ac conductivity related to the index of refraction by $n^2 = 1 + i4\pi\sigma/\omega$. We have used solutions to Eqs. (2) and (3) in our analysis.

The final parameter necessary to determine ρ_{dc} is the electron density, N_e, because N_e enters into the analysis not only directly through Eq. (1), but through terms in Eqs. (2) and (3) that depend on ω_p as well. As a function of temperature at solid densities, N_e can be readily modeled within the framework of local thermodynamic equilibrium (LTE)[9] provided all the microscopic ionization and three-body recombination rates are sufficiently high that nearly equilibrium conditions for the electrons and ion excitation states are obtained during the heating-reflecting pulse.[10] Figure 2 shows the results of an LTE calculation in aluminum of N_e/N (N = atomic density of Al) as a function of T_e. The Fermi temperature of Al is nearly 12 eV, with all three of aluminum's valence electrons itinerant; as a result, LTE shows N_e to be a *weak* function of temperature for temperature less than 50 eV. Because of the short pulse duration, the assumption of LTE may not be completely valid; however, because of the relatively smaller ionization rates of the higher ionization stages, any time-dependent calculation will not yield values of N_e larger than those calculated by LTE.

The process of extracting the value of ν from each recorded value of the reflectivity proceeds as follows: (a) For each value of the reflectivity, the input intensity, and hence the average electron temperature, is determined. (b) The effective interface expansion is determined from the Doppler-shift velocity. (c) The Helmholtz equations [Eqs. (2) and (3)] for the appropriate polarization are numerically integrated for the effective expansion length. (d) By iteration the value for ν *within the solid-density aluminum* that correctly gives the experimental value of the reflectivity is determined.[11]

From the P-polarized data of Fig. 1, we have extracted values of ρ vs T_e as shown in Fig. 3. (Equivalent results were obtained for the S-polarization data.) Although the results are presented for the solid-density aluminum at the indicated temperature, they were determined by our taking into account the small, but not insignificant interface expansions.[12] This curve shows clearly the three regions of transport, including a well defined "saturation" in the resistivity near 200 $\mu\Omega$ cm at $T_e > 40$ eV.

The resistivity increases linearly with T_e up to ≈ 5 eV, then sublinearly to nearly 40 eV. For lattice temperatures greater than T_{Debye}, classical electron-lattice scattering predicts a *linear* dependence of ρ upon T_{ion}. It is well known, however, that short-pulse excitation can produce nonequilibrium temperature distributions with $T_e > T_{\mathrm{ion}}$ (Ref. 13); thus the data for 5 eV $< T_e < 40$ eV partially reflect the temperature dependence of the electron-phonon coupling at elevated temperatures.

At the highest temperatures and intensities, our data are consistent with the expectations of a high-temperature plasma; that is, a $T_e^{-3/2}Z$ dependence of the resistivity. This result derived from the Coulomb-scattering cross section averaged over a Maxwellian velocity distribution.[14]

The maximum in the resistivity near 200 $\mu\Omega$ cm over nearly an order of magnitude in laser intensity is interpreted[15] as "resistivity saturation"; that is, there exists a minimum scattering length (or mean free path) of the electrons, regardless of the degree of disorder of the material or the temperature. We can estimate this length by computing $l_{\min} = (\sigma_{\mathrm{dc}} m_e/N_e e^2)v_e$, where v_e [$=(2kT_e/$

$m_e)^{1/2}]$ is the electron velocity at the resistivity maximum. For $T_e = 50$ eV, $l_{\min} \approx 3$ Å. This value is comparable to the interatomic spacing in Al, and suggests the picture that the electron cannot travel less than approximately an interatomic dimension before suffering another scattering event.

This rather remarkable record of the change in the controlling mechanisms of the electrical transport of a simple metal is possible only because the material could be heated to electron temperatures up to and far in excess of its Fermi temperature, without significantly changing the density. Although the approach to resistivity saturation has been observed at much reduced temperatures in some specialized systems,[16] to our knowledge the complete onset and passage through the maximum has not been previously reported.

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[11] For these scale lengths ($\leq \lambda/15$), the results are largely insensitive to both the details of the particular model for N_e as a function of atom density and T_e in the expansion scale length, and the detailed shape of the density profile. Although the measurement represents a Gaussian average over time and space within the plane of the target, the heat conductivity, calculated from the electrical conductivity results by using the Weidemann-Franz relation [J. M. Zimon, *Principles of the Theory of Solids* (Cambridge Univ. Press, Cambridge, United Kingdom, 1965), p. 197] shows that heat diffusion is sufficiently rapid so as to justify the assumption of homogeneous parameters behind the interface.

[12] In the final analysis, we found that the results differed little from a data reduction procedure using Fresnel equations and assuming no interface expansion, presumably a consequence of the large increase in the skin depth under the conditions of high collisionality (i.e., high temperatures).

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