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Plasmon Excitation in the Scattering of Electromagnetic Waves From Good Conductors

by
Clifford R. Schumacher

DTRC–89/025 Plasmon Excitation in the Scattering of Electromagnetic Waves From Good Conductors

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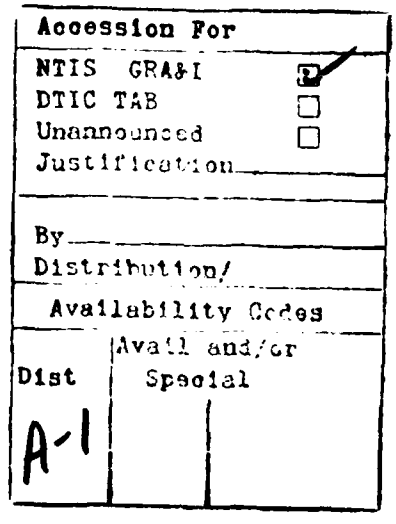

ABBREVIATIONS AND SYMBOLS

c	Velocity of light
$\mathbf{D}$	Electric displacement vector
DTRC	David Taylor Research Center
$\mathbf{E}$	Electric field vector
e	Electron charge
f_o	Frequency (in THz) at which the real part of the dielectric function vanishes
$\hbar$	Planck's constant divided by 2π
ε	Dielectric constant
IR	Infrared
$\mathbf{j}$	Electron current density vector
λ	Wavelength
m	Electron mass
n	Electron density
ω	Angular frequency
ω_0	Frequency at which the real part of the dielectric function vanishes
ω_p	Electron plasma frequency
ω_τ	Electron damping frequency
R	Reflection coefficient for power
ρ	Resistivity
σ	Conductivity
τ	Mean collision time
THz	Terahertz
$\mathbf{v}$	Mean electron velocity vector

ABSTRACT

Ordal et al. report excellent Drude model fits to their Kramers-Kronig analysis of measurements of the optical constants of 15 metals—Ti, Fe, Co, Ni, Pt, V, Pd, W, Pb, Cu, Mo, Ta, Ag, Au, and Al. Their resulting values of plasma and damping frequencies not only provide excellent fits to the measured values of the conductivity but also were shown to predict the frequency dependence of the dielectric constant. All static values of the dielectric constant of these 15 metals were negative and very large, but increased monotonically with increasing frequency, passing through zero in the optical range between wavelengths of 1,000 and 6,000 Å. This behavior was interpreted as plasmon excitation, and very sharply peaked loss factors were calculated. Furthermore, the novel behavior of the dielectric constant caused a large impedance discontinuity that kept the reflection coefficient very nearly equal to 1 until it dropped very rapidly at the plasma resonance frequency to values approaching zero as photon energies reached the electron volt range.

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INTRODUCTION

While many measurements have been made of the dielectric constant of poorly conducting materials, the dielectric constant of good conductors is usually regarded as unknown. This situation results from the total current density producing magnetic fields according to the Maxwell equations being composed of two parts: the conduction current associated with free electrons and the displacement or polarization current associated with bound charges.

Both currents are proportional to the electric field vector **E**. The conduction component is also proportional to the conductivity σ, while in harmonic fields the displacement component is also proportional to the dielectric constant ε and the angular frequency ω. Since values of σ for good conductors are such that σ does not become comparable with expected values of $\omega\varepsilon$ until frequencies in the visible part of the spectrum are reached, Slater and Frank[1] felt that the optical properties of metals can be described by the conduction current alone through the radio frequency, microwave, and infrared (IR) parts of the spectrum and that dielectric effects only begin to be important in the visible region.

Slater and Frank[1] also stated that at low frequencies the optical properties of a conductor are determined entirely by σ, not by ε, and that there is *no* experimental way of finding the ε of a good conductor at low frequencies.

Recently, Ordal et al.[2–4] used an improved method to calculate the surface impedance of metals from the measured transmission of a nonresonant cavity. They also reported excellent Drude model fits to the Kramers-Kronig analysis of their data

combined with that of others at higher frequencies. Their resulting values of plasma and damping frequencies not only provide excellent fits to the measured values of σ but also can be used to calculate ε as a function of frequency.

The values and behavior of ε as a function of frequency are found to be quite different from those of poorly conducting materials. These results are interpreted as plasmon excitation, very sharply peaked loss factors are calculated, and significant effects on the scattering of electromagnetic waves are exhibited.

DRUDE MODEL FITS TO THE OPTICAL PROPERTIES OF METALS

Many measurements of the optical constants of metals have been made, primarily at near IR, visible, and ultraviolet wavelengths. Although the extremely high reflectivity of metals at submillimeter wavelengths generally precludes direct measurement of the reflectance or absorptance as is customary at shorter wavelengths, Ordal et al.[2–4] reported improved measurements of 15 metals—Ti, Fe, Co, Ni, Pt, V, Pd, W, Pb, Cu, Mo, Ta, Ag, Au, and Al— using a nonresonant cavity. Their method was used to obtain $r(\omega)$ [the real part of the normalized surface impedance $z(\omega) = r(\omega) + ix(\omega)$] from the measured transmission of the nonresonant cavity between $1/\lambda = 30$ and 300 cm^{-1}. They then used optical constants from the literature to calculate $r(\omega)$ at shorter λ. The resulting $r(\omega)$ spectrum was Kramers-Kronig analyzed to obtain $x(\omega)$.

By thus combining their far-IR measurements with others at shorter λ, Ordal et al.[2–4] obtained the real and imaginary parts of the normalized surface impedance or, equivalently, the real and imaginary parts of the dielectric function $\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$ in terms of two adjustable parameters, thus providing a fit to ε and σ useful for photon energies ranging from zero to the electron volt range, where the Drude model loses its validity because of interband optical transitions.

The Drude model was proposed in 1904 and is the earliest realistic model for a metal.[5,7] Drude assumes the electrons form a gas of free particles, which could respond to electromagnetic fields by producing current. Assuming each electron to have a mean collision time τ, then the mean electron velocity vector $\mathbf{v}$ is given by Newton's law as

$$\frac{\partial \mathbf{v}}{\partial t} = \frac{e}{m}\mathbf{E} - \frac{1}{\tau}\mathbf{v} \quad , \tag{1}$$

where $\mathbf{E}$ is the applied electric field vector. If $\mathbf{E}$ depends on time through a factor $e^{i\omega t}$, it follows that

$$\mathbf{v} = \frac{e\mathbf{E}}{m\{i\omega + (1/\tau)\}} \quad , \tag{2}$$

Since $\mathbf{j} = ne\mathbf{v}$, where n is the electron density, the current density due to all electrons is

$$\mathbf{j} = \frac{\sigma_o}{1 + i\omega\tau}\mathbf{E} \quad , \tag{3}$$

where $\sigma_0 = ne^2\tau/m$ is the static conductivity. Rationalizing the denominator,

$$\mathbf{j} = \frac{\sigma_o}{1+\omega^2\tau^2}(1 - i\omega\tau)\mathbf{E} = \sigma(\omega)\left(\mathbf{E} - \tau\frac{\partial \mathbf{E}}{\partial t}\right) \quad , \tag{4}$$

where the component of **j** proportional to **E** is the conduction current, with the coefficient of proportionality being the effective conductivity at frequency ω:

$$\sigma(\omega) = \frac{\sigma_o}{1+\omega^2\tau^2} \quad . \tag{5}$$

The component of **j** proportional to the time rate of change of **E** (and, therefore, out of phase with it) is physically equivalent to the displacement current arising from ε.

These relations show that, as long as $\omega\tau << 1$, **j** is almost entirely in phase with **E**, and for most purposes ε is unimportant. When $\omega\tau >> 1$, most of the current is out of phase with **E**, and the properties of waves traveling through a medium are then mainly influenced by ε.

Considerable simplification arises if Ordal's IR spectroscopic notation[2–4] is used, where all "frequencies" are expressed in cm^{-1} and are thus equal to the physicist's $1/\lambda$. The Drude model contains the effects of both conduction and displacement currents, and its **j** may be written in terms of either of the complex analytic functions $\sigma(\omega)$ or $\varepsilon(\omega)$. Choosing $\varepsilon(\omega)$, Ordal[4] wrote the real and imaginary parts of the dielectric function as

$$\varepsilon_1 = 1 - \frac{{\omega_p}^2}{\omega^2 + {\omega_\tau}^2} \quad , \tag{6}$$

and

$$\varepsilon_2 = \frac{\omega_\tau {\omega_p}^2}{\omega(\omega^2 + {\omega_\tau}^2)} \quad , \tag{7}$$

which is completely equivalent to the description in terms of ε and σ, recognizing that Eqs. 6 and 7 provide Drude model predictions for the dependence of ε and σ on photon frequency.

In Eqs. 6 and 7, the damping frequency ω_τ is

$$\omega_\tau(\mathrm{cm}^{-1}) = \frac{1}{2\pi c\tau} \quad , \tag{8}$$

where τ is the electron lifetime in seconds. The plasma frequency $\omega_p(cm^{-1})$ is defined correspondingly by dividing the conventional plasma frequency in hertz by $2\pi c$.

Using Eq. 7, the static value of the conductivity may be written in terms of ω_p and ω_τ, Ordal[4] referred to this as σ_{opt}, the "high or optical frequency" conductivity:

$$\sigma_{opt} = \frac{{\omega_p}^2}{4\pi\omega_\tau} \quad , \tag{9}$$

where σ_{opt} has the units cm^{-1}. The value of σ_{opt} should be equal to the standard static conductivity σ_o, which can be expressed in terms of measured values of the static resistivity ρ_O given in the *American Institute of Physics Handbook*[6]:

$$\sigma_o(\mathrm{cm}^{-1}) = \frac{1}{2\pi c[\varrho_o(\mathrm{s})]} = \frac{9 \times 10^{11}}{2\pi c[\varrho_o(\Omega\mathrm{cm})]} \quad . \tag{10}$$

These two conductivities are not exactly equal, as can be seen from the results of fits to the dielectric function of 15 metals (Table 1), where ω_τ, ω_p, and ρ_{opt} are taken from Ordal et al.[2–4] However, Table 1 shows that most values of ρ_O/ρ_{opt} are very close to unity.

Table 1. Results of a Drude model fit to the dielectric function of 15 metals.

Metals	ω_τ (cm^{-1})	ω_p (cm^{-1})	ρ_o[a] ($\mu\Omega$cm)	ρ_{opt} ($\mu\Omega$cm)	$\frac{\rho_o}{\rho_{opt}}$	$\varepsilon_1(0)$
Pb[b]	1175.0	59,400	21.0	19.98	1.051	–2,555
Ta[c]	704.0	66,200	13.1	9.64	1.359	–8,841
Pt[d]	358.0	41,500	10.42	19.44	0.536	–5,530
V[d]	489.0	41,600	19.9	16.95	1.174	–7,236
W[d]	487.0	51,700	5.33	10.93	0.488	–11,270
Al[c]	424.0	95,400	2.74	2.80	0.980	–50,624
Mo[d]	412.0	60,200	5.33	6.82	0.781	–21,350
Ti[d]	382.0	20,300	43.1	55.62	0.775	–2,823
Co[d]	295.0	32,000	5.80	17.29	0.336	–11,770
Au[d]	215.0	72,800	2.20	2.43	0.904	–114,650
Ni[b]	178.0	39,400	7.04	6.88	1.023	–48,990
Fe[c]	156.0	29,500	9.80	10.76	0.911	–35,760
Ag[d]	145.0	72,700	1.61	1.65	0.978	–251,380
Pd[d]	124.0	44,000	10.55	3.84	2.745	–125,910
Cu[d]	73.2	59,600	1.70	1.24	1.375	–662,900

[a]Ref. 6, pp. 9-39, 9-40.
[b]Ref. 2, p. 747.
[c]Ref. 3, pp. 1205-1207.
[d]Ref. 4, p. 4494.

The values of ω_τ and ω_p in Table 1 not only provide excellent fits to the measured values of ρ_o but also can be used to calculate values of ε as a function of frequency. The value $\varepsilon_1(0)$, the static value of the real part of the dielectric function $\varepsilon(\omega)$, is obtained from Eq. 6 :

$$\varepsilon_1(0) = 1 - \frac{\omega_p{}^2}{\omega_\tau{}^2} \quad , \tag{11}$$

and is also listed in Table 1 for each of the 15 metals.

As Eq. 6 shows, the function $\varepsilon_1(\omega)$ increases monotonically from the large negative static values displayed in Table 1 to the value 1 at infinite frequency, passing through zero in the vicinity of the optical range. On the other hand, Eqs. 5 and 9 show $\sigma(\omega)$ decreases monotonically with increasing frequency. This behavior is shown in Fig. 1 and is

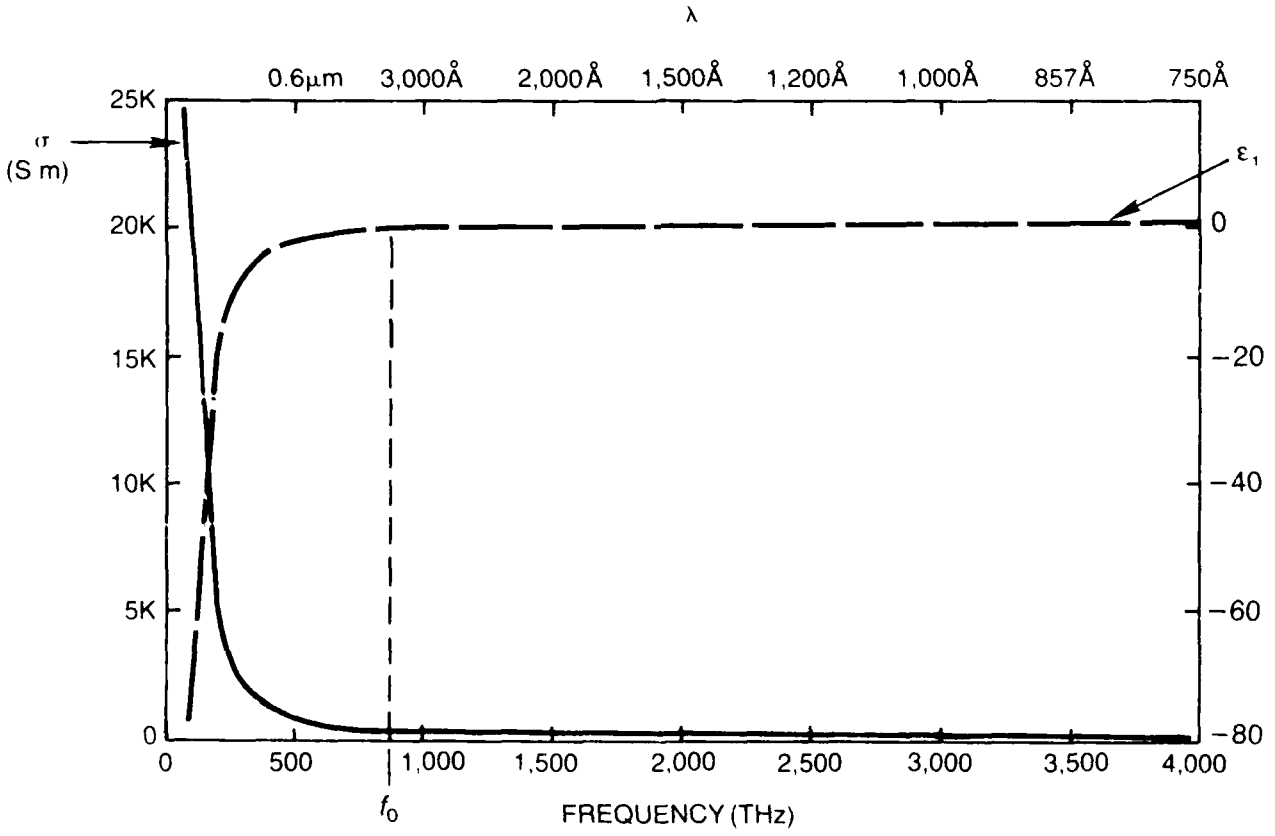

Fig. 1. Values of σ and ε_1 for *Fe* as functions of λ and frequency (Thz). f_o is the frequency at which ε_1 vanishes.

characteristic of plasma or collective oscillations in a solid, resulting from the high density of electrons present in a metal and the fact that they can act cooperatively due to the Coulomb interaction among them.

The energy of these oscillations is large compared to single-particle energies because a plasma oscillation at long wavelengths involves the correlated motion of a very large number of electrons. No single electron is greatly perturbed but, because large numbers of electrons are moving together in a coherent fashion, the resultant energy of the collective mode is quite substantial.

ELECTRONS, PLASMONS, AND PHOTONS IN SOLIDS

Drude completely neglected the ionic cores present in solids and the fact that electrons obey Fermi statistics. Pines[5] provided a much more complete treatment, including the effects of the periodic lattice, phonons, electron-electron and electron-phonon interactions, and plasmons. A plasmon is the quantum of plasma oscillation energy. Electron interactions in solids differ from those in a free electron gas, because changes in screening behavior due to the periodic ion potential influence the plasmon spectra. Plasmons are bosons, and their distribution function has the characteristic boson form at finite temperatures.

Under certain conditions plasma oscillations represent normal modes of the entire system, meaning that once such oscillations are excited, they do not decay with time. Since they maintain themselves, an internal electric field $\mathbf{E}(\omega_p)$ would exist in the solid at the plasma frequency ω_p due to oscillations in the electron density, without the presence of an external field $\mathbf{D}(\omega_p)$. By the constitutive relation, since

$$\mathbf{D}(\omega_p) = \varepsilon(\omega_p)\ \mathbf{E}(\omega_p) = 0 \quad , \tag{12}$$

and $\mathbf{E}(\omega_p) \neq 0$, the condition for plasma oscillations is,

$$\varepsilon(\omega_p) = \varepsilon_1(\omega_p) + i\varepsilon_2(\omega_p) = 0 \quad . \tag{13}$$

Thus, plasma oscillations should exist as normal modes of the system only if both real and imaginary parts of the dielectric constant vanish at ω_p. In practice, however, plasma oscillations will exist if $\varepsilon_2(\omega_p) < 1$ when $\varepsilon_1(\omega_p) = 0$. Since ε_2 represents the damping of the plasma resonance, the condition $\varepsilon_2(\omega_p) < 1$ implies damping should be small.

More precisely, when plasma oscillations are damped, the frequency at which $\varepsilon_1(\omega) = 0$ does not quite provide the plasmon energy. By Eq. 6, the real part of the dielectric function vanishes at the frequency ω_o, where

$$\omega_o{}^2 = \omega_p{}^2 - \omega_\tau{}^2 \quad . \tag{14}$$

Because $\omega_\tau/\omega_p << 1$, ω_o is very nearly equal to ω_p. Values of ω_o in cm^{-1} are given in Table 2, along with the equivalent frequency f_o in THz and the corresponding photon energy in eV for all 15 metals. Table 2 also gives $\varepsilon_{20} = \varepsilon_2(\omega_o)$, the value of the imaginary

part of the dielectric function at ω_o, which is seen to be much less than 1 for all 15 metals.

Table 2. Imaginary part of the dielectric function ε_{20}, loss factor, and half-width $\Delta\omega_o$ at the plasmon resonance frequency ω_o.

Metals	ω_O (cm^{-1})	f_o (THz)	$\hbar\omega_o$ (eV)	ε_{20}	Loss Factor	$\Delta\omega_o$ (cm^{-1})
Ti	20,296.4	609	2.52	0.0188	53.1	382.0
Fe	29,499.6	885	3.66	0.0053	189.1	156.0
Co	31,998.6	960	3.97	0.0092	108.5	295.0
Ni	39,399.6	1,182	4.89	0.0045	221.3	178.0
Pt	41,496.2	1,245	5.15	0.0134	74.4	558.0
V	41,597.1	1,248	5.16	0.0118	85.1	489.0
Pd	43,999.8	1,320	5.46	0.0028	354.8	124.0
W	51,697.7	1,551	6.41	0.0094	106.2	487.0
Pb	59,388.4	1,782	7.37	0.0198	50.5	1,175.0
Cu	59,600.0	1,788	7.39	0.0012	814.2	73.2
Mo	60,198.6	1,806	7.47	0.0068	146.1	412.0
Ta	66,196.3	1,986	8.21	0.0106	94.0	704.0
Ag	72,699.9	2,181	9.02	0.0020	501.4	145.0
Au	72,799.7	2,184	9.03	0.0030	338.6	215.0
Al	95,399.1	2,862	11.84	0.0044	225.0	424.0

The principal experimental evidence for the existence of plasmons as a well-defined excitation mode of the valence electrons in solids comes from characteristic energy-loss experiments, where the energy spectrum of kilovolt electrons is observed, either as they emerge from a thin solid film or after they are reflected from a solid surface.[5] In fact, the most familiar method of determining ω_p utilizes measurement of these characteristic electron energy losses, which are proportional to $Im(1/\varepsilon(\omega))$, a function of width $\Delta\omega_O = \omega_\tau$ that is very sharply peaked about ω_O when the conditions for plasma oscillations are fulfilled.[8] When the angular distribution of the inelastically scattered electrons is measured, $Im(1/\varepsilon(\omega))$ is directly measured for the electrons in the solid.

Whereas fast electron scattering is a longitudinal probe of the solid, in which the electron gas responds to a time-varying longitudinal field, measurements of the optical reflectivity of a solid constitute a transverse probe of the solid, because the electromagnetic wave couples directly to the transverse current-density fluctuations of the electrons. Therefore, the dielectric constant is a tensorial quantity because, just as the response of the electron gas to a time-varying longitudinal field defines a longitudinal dielectric constant, the system response to an external electromagnetic field defines a transverse dielectric constant.

Pines[5] presented a comparison of the optical values of plasmon energies for the alkali metals, based on reflection experiments, with the values measured in electron energy-loss experiments. He remarked that agreement between the two methods is quite good. Detailed experiments have been made to compare the electron energy-loss function with that calculated from known optical constants. General agreement has been found in the detailed structure of the two loss functions,[8] so no experimental evidence exists to date for a difference between the two dielectric constants.

Values of $Im(1/\varepsilon(\omega))$, calculated from the Table 1 values of ω_τ and ω_p, are given in Table 2 and exhibited for all 15 metals as a function of f_o and λ in Fig. 2.

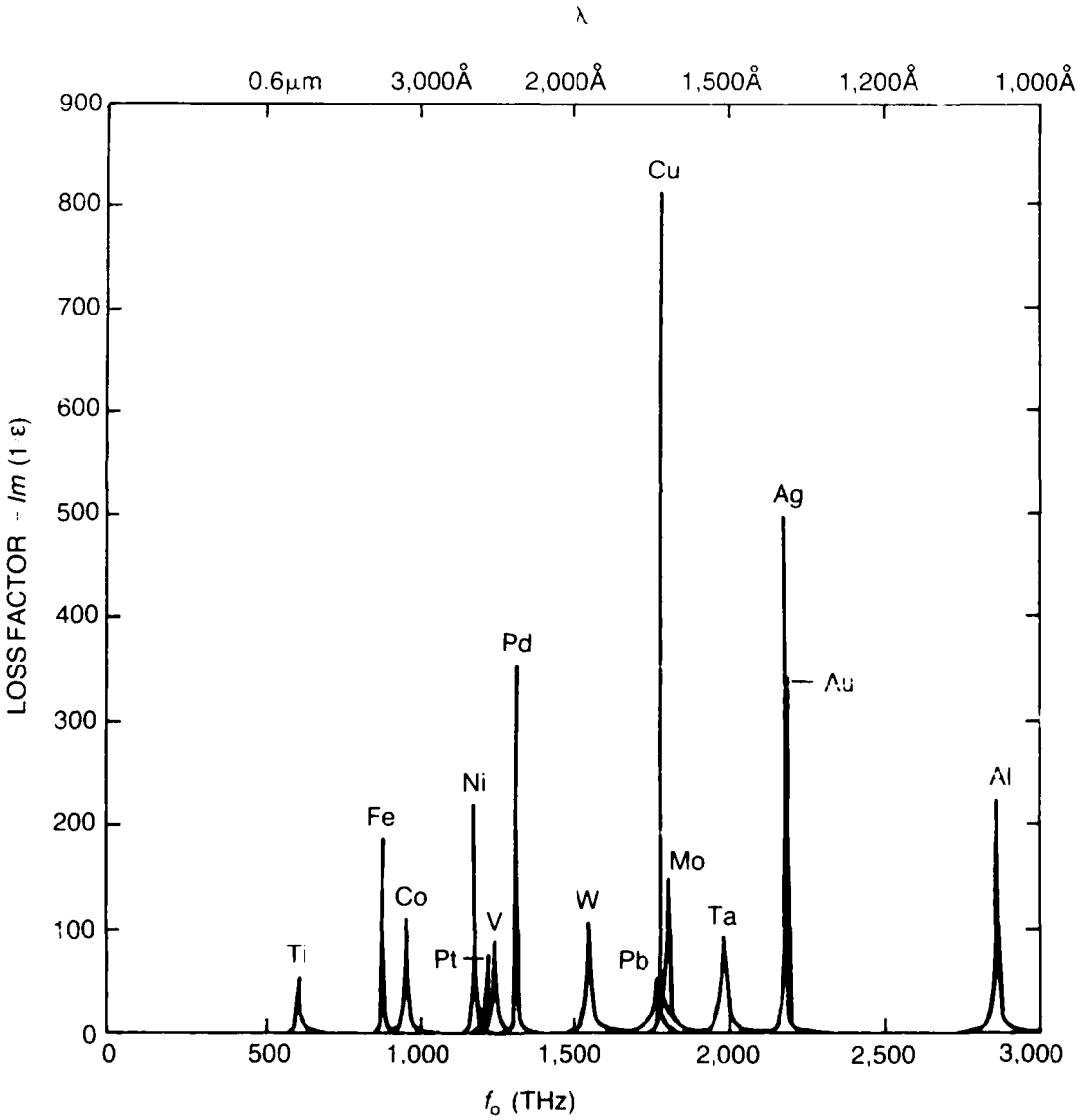

Fig. 2. Values of the loss factor $= -Im\,(1/\varepsilon)$ for 15 metals as a function of f_o and λ.

This novel behavior of the dielectric constant in a region of appreciable conductivity has a profound effect on the scattering of electromagnetic waves from metals. If one considers scattering from a simple model of a solid with $\varepsilon \equiv 1$ and σ equal to its constant static value, the reflection coefficient gradually decreases from 1 with increasing frequency (Fig. 3), in accordance with the familiar Hagen-Rubens relation discussed by Born and Wolf.[9] The author has provided an explanation of the scaling laws that shows σ must increase with increasing frequency for the reflection coefficient to remain constant.[10]

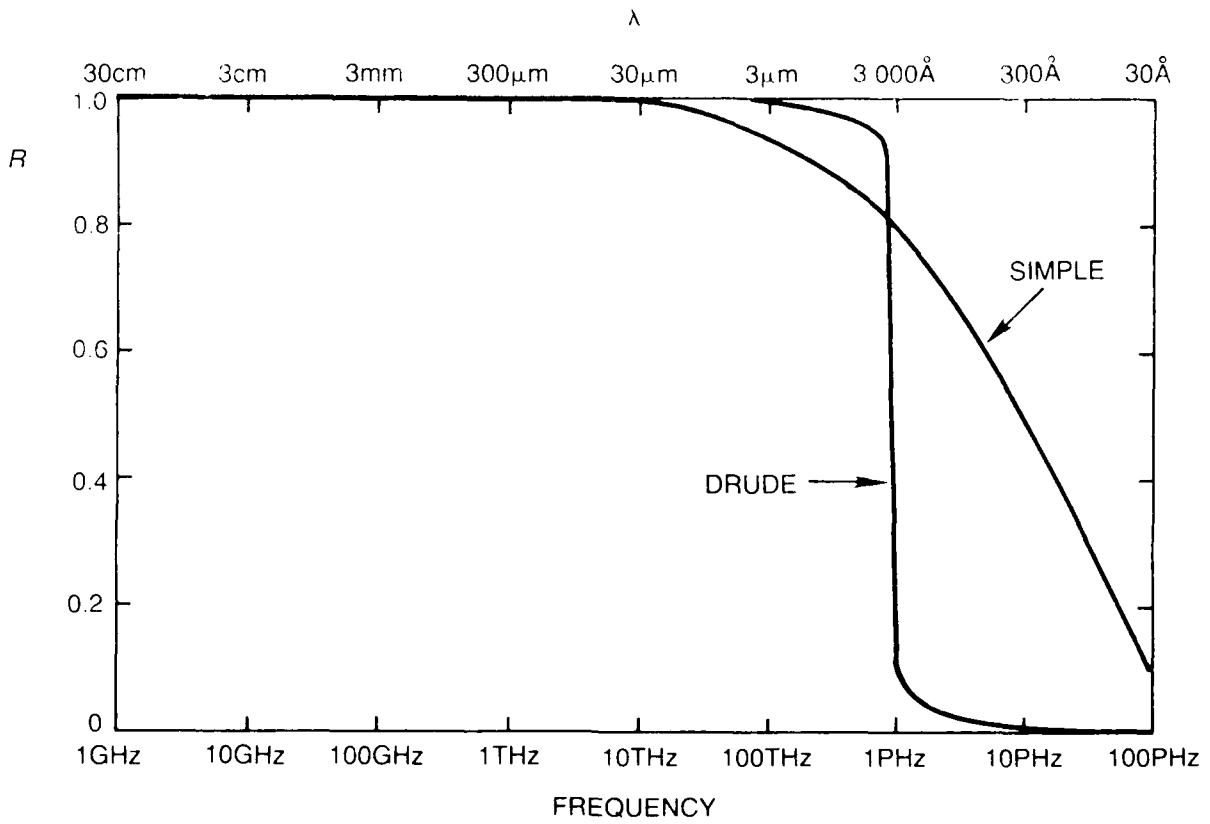

Fig. 3. Values of *R*, the reflection coefficient for power, for *Fe* as a function of frequency and λ for the Drude theory and for a simple solid model with the static value of σ and $\varepsilon \equiv 1$.

Even though the Drude σ decreases with increasing frequency in accordance with Fig. 1, which by itself would lead to a reflection coefficient decreasing from 1 with increasing frequency more rapidly than the simple solid model shown in Fig. 3, the strongly negative Drude ε causes a large impedance discontinuity that keeps the reflection coefficient very nearly equal to 1 until it drops very rapidly at the plasma resonance frequency to values far below those of the simple solid model.

This finding has significant practical consequences. Usually, infinite σ is assumed for the metals used in radar calculations, even though eventual decreases are expected in the reflection coefficient of physical scale models as the measurement frequency in-

creases, caused by the polarization not being able to keep up with exciting electromagnetic fields. In fact, scaling laws require that σ increase to large unphysical values in the IR range for the reflection coefficient to remain constant.[10]

However, this treatment of plasmon excitation in good conductors yields the same reflection effect as a σ actually increasing with frequency in the physical scale modeling range, but finally decreasing to zero as photon energies reach the electron volt range, in accordance with Fig. 3. Thus, not only is the assumption of infinite σ fully justified in physical scale model calculations, but also there is a new understanding of the reflection properties of metallic scale models.

The previous results also have implications for radar-absorbing materials. An ideal absorber is one that allows incident electromagnetic energy to enter without reflection and then to rapidly attenuate in a short distance. However, it does not appear possible to satisfy these two conditions simultaneously. Undoubtedly, attenuation is rapid if both electric and magnetic losses are high. However, these losses are related to imaginary components of the dielectric and magnetic polarization, both of which result in reflection.

CONCLUSIONS

For the first time, static values of the dielectric constant, ε, of metals have been obtained from experimental data. All these static values are shown to be negative and very large. Higher frequency values of ε were shown to increase monotonically with increasing frequency, passing through zero in the optical range between wavelengths of 1,000 and6,000Å. This frequency dependence of ε is obtained from values of plasma and damping frequencies reported by Ordal et al.,[2–4] which were derived from excellent Drude model fits to the Kramers-Kronig analysis of a combination of their far-IR measurements of the optical constants of 15 metals and other previous higher frequency measurements.

The authors' far-IR measurements[2–4] also predict values of the conductivity, σ, that decrease monotonically with increasing frequency but are in excellent agreement with measured static values. This behavior is characteristic of plasma or collective oscillations in a solid and is interpreted as plasmon excitation. Very sharply peaked loss factors are calculated for all 15 metals. The novel behavior of ε causes a large impedance discontinuity, which keeps the reflection coefficient very nearly equal to 1 until it drops very rapidly at the plasma resonance frequency to values approaching zero as photon energies reach the electron volt range. This behavior is quite different from the scattering from a simple model of a solid with the static value of σ and $\varepsilon \equiv 1$, which yields a reflection coefficient decreasing more slowly with increasing frequency, in accordance with the familiar Hagen-Rubens relation.

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