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THE OPTICS OF METALS: AN INTRODUCTION

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ABSTRACT

The Optics of Metals is an interdisciplinary subject, of interest for courses of Electromagnetic Theory, Optics, and Solid State Physics. The review is meant for presentation at the undergraduate level. The subject matter is chosen for its simplicity and includes the following topics: the optical constants, the Drude model, inhomogeneous waves, propagation and refraction (including the skin effect), reflectivity (including plasma effects), bulk plasmons, and surface plasmon-polaritons.

I. INTRODUCTION

The Optics of Metals is an interdisciplinary subject, and numerous textbooks of Electromagnetic Theory(1), Optics(2), and Solid State Physics(3) include sections or even a chapter devoted to it. A book has been also published on this theme(4). Most aforementioned texts are at the graduate level.

This review is meant for presentation at the undergraduate level. Hence the subject matter was chosen for its simplicity, from both the physical and mathematical point of view. At the same time the interdisciplinary nature of Metal Optics is stressed.

The material properties of metals may be described in terms of their "optical constants". Various descriptions are in use and will be given in Sec. 2. The Drude model[5] (Sec. 3), in spite of its antiquity, is alive and well and most useful when it comes to specific results for physical understanding of metallic behavior. In Sec. 4 we show that electromagnetic waves in metals are not simple plane waves: for a finite angle of incidence of the light the planes of constant phase and the planes of constant amplitude are not parallel to each other. Such waves are called "inhomogeneous". They exhibit some interesting propagation characteristics (Sec. 5) such as the skin effect and the dependence of the phase velocity on the angle of incidence. Sec. 6 deals with metallic reflectivity, the plasma edge, the critical angle, and the Brewster angle. The last two themes to be treated usually belong to Solid State Physics, however they fit in naturally into a presentation based on electromagnetic theory. They are plasmons (Sec. 7) and surface plasmons (Sec. 8). We shall derive the various modes which may propagate in the bulk of a metal and at its surface and also discuss optical techniques for their excitation.

With the exception of the last section, which deals with relatively new developments, this review is limited to semiinfinite metallic media (Fig. 1). A tutorial article by Nestell and Christy[6] is available on the subject of the optics of thin metallic films. A thorough treatment of the classical theory of optical dispersion was given by Christy[7]. Recent advances, including experimental data on optical properties of many metals and alloys are reviewed in a monograph by Nilsson[8].

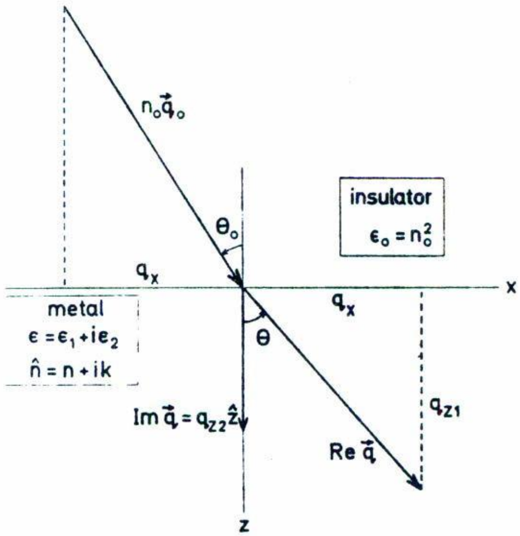

Fig. 1 Refraction of a plane wave from an insulator into a metal. The metal is characterized by either its complex dielectric constant ε or its complex index of refraction N. The wave in the metal is inhomogeneous: the real and imaginary parts of the wavevector $\vec{q}$ point in different directions for $\theta_0 \neq 0$. Note that the case $\theta > \theta_0$ may be realized for $n < 1$.

2. THE OPTICAL CONSTANTS ε, σ, AND N

One of Maxwell's equations,

$$\vec{\nabla} \times \vec{H} = \frac{4\pi}{c}\vec{J} + \frac{1}{c}\frac{\partial \vec{D}}{\partial t} , \tag{1a}$$

is frequently expressed in one of two alternative forms. In the first, the conduction current $\vec{J}$ is "absorbed" in an effective displacement vector $\vec{D}_{ef}$:

$$\vec{\nabla} \times \vec{H} = \frac{1}{c}\frac{\partial \vec{D}_{ef}}{\partial t} . \tag{1b}$$

In the second description, that part of the displacement current which is associated with material (non-vacuum) properties is "absorbed" in an effective current $\vec{J}_{ef}$:

$$\vec{\nabla} \times \vec{H} = \frac{1}{c}\frac{\partial \vec{E}}{\partial t} + \frac{4\pi}{c}\vec{J}_{ef} . \qquad (1c)$$

The formulations (1a), (1b), and (1c) are completely equivalent and the choice is a matter of convenience.

The linear response corresponding to Eq. (1b) is given by

$$\vec{D}_{ef} = \tilde{\varepsilon} \cdot \vec{E} , \quad \tilde{\varepsilon} = \tilde{\varepsilon}_1 + i\,\tilde{\varepsilon}_2 \qquad (2)$$

where $\tilde{\varepsilon}$ is the complex dielectric tensor. This description is appropriate to anisotropic crystals. Alternatively, the linear response appropriate to Eq. (1c) is

$$\vec{J}_{ef} = \tilde{\sigma} \cdot \vec{E} , \quad \tilde{\sigma} = \tilde{\sigma}_1 + i\,\tilde{\sigma}_2 \qquad (3)$$

where $\tilde{\sigma}$ is the complex conductivity tensor. Our review will be limited to an harmonic time dependence, i.e. all fields and sources are proportional to exp-(iωt). Then substituting Eq. (2) in Eq. (1b), and Eq. (3) in Eq. (1c), and comparing the results we find that

$$\varepsilon_{ij} = \delta_{ij} + (4\pi i/\omega)\,\sigma_{ij} . \qquad (4)$$

A cubic crystal is characterized by isotropy, i.e. $\vec{D}_{ef} \,||\, \vec{J}_{ef} \,||\, \vec{E}$ for an arbitrary direction of the electric field. Then the off-diagonal elements of $\tilde{\varepsilon}$ vanish and the diagonal elements are equal. Eq. (4) is replaced by

$$\varepsilon = 1 + (4\pi i/\omega)\sigma ; \qquad (5)$$

σ is often called the "optical" conductivity. Separating Eq. (5) into real and imaginary parts we get

$$\varepsilon_1 = 1 - (4\pi/\omega)\sigma_2 \ , \quad \varepsilon_2 = (4\pi/\omega)\sigma_1 \ . \tag{6}$$

The complex index of refraction, N, provides a third description of the optical constants of an isotropic dielectric medium. It is defined by the relation

$$\varepsilon = N^2 = (n + ik)^2 \ . \tag{7}$$

The real and imaginary part of ε are given by

$$\varepsilon_1 = n^2 - k^2 \ , \quad \varepsilon_2 = 2nk \ . \tag{8}$$

Conversely, the real and imaginary part of N may be found from Eqs. (8) in terms of the real and imaginary part of ε. They are given by the equations

$$2n^2 = (\varepsilon_1^2 + \varepsilon_2^2)^{1/2} + \varepsilon_1 \tag{9a}$$

$$2k^2 = (\varepsilon_1^2 + \varepsilon_2^2)^{1/2} - \varepsilon_1 \ . \tag{9b}$$

3. THE DRUDE MODEL

The Drude model[5] has gone a long way in explaining transport and optical properties of conductors. The dielectric function has all the merits of simplicity:

$$\varepsilon = 1 - \frac{\omega_p^2}{\omega(\omega+i\nu)} \ . \tag{10}$$

The plasma frequency ω_p is given by $\omega_p = (4\pi ne^2/m^*)^{1/2}$ where n is the density of the charge carriers and m^* is their effective mass. The other parameter of the model is the collision frequency (or relaxation frequency) ν . Its reciprocal, $\tau = 1/\nu$, is the collision time. It is a phenomenological way to describe the average time between collisions due to complex microscopic processes such as electron-phonon and electron-electron interaction. Note that, for $\nu=0$, the Drude dielectric function vanishes at the frequency ω_p, i.e. $E(\omega_p)=0$.

By using Eq. (5) we can find the expression for the conductivity that corresponds to Eq. (10). It is given by

$$\sigma(\omega) = \frac{\sigma_0}{1 - i\omega\tau} \quad , \qquad \sigma_0 = \frac{ne^2\tau}{m^*} \quad ; \qquad (11)$$

σ_0 is the D.C. conductivity. This result may be derived from a simple equation for the total force acting on an electron:

$$m^* \frac{d\vec{v}}{dt} = -e\vec{E} - m^* \frac{\vec{v}}{\tau} \quad . \qquad (12)$$

The second term is a damping force on an electron moving with the drift velocity $\vec{v}$. The current is given by $\sigma\vec{E} = -ne\vec{v}$ and Eq. (11) readily follows.

The real and imaginary part of the Drude dielectric function (10) are plotted in Fig. 2. The large negative values of ε_1 and its change of sign at $\omega \approx \omega_p$ are characteristic of metallic behavior. The collision frequency is chosen to be $\nu=0.01\ \omega_p$ (or $\omega_p\tau=100$), which is a reasonable value for a pure metal at room temperature. This frequency is usually taken as a dividing line between the low-frequency region $\omega \ll \nu$, and the high-frequency region $\omega \gg \nu$. However, a real metal does not often satisfy the Drude model in a frequency range as wide as shown in Fig. 2. A frequent test of the validity of the model for $\omega \gg \nu$ is plotting of experimental values of ε_1 as a function of λ^2 (where λ is the wavelength), and of ε_2 as a function of λ^3. In the case of "Drudelike" behavior both graphs should be straight lines; this follows from Eq. (10) because

$$\varepsilon_1 \cong 1 - \omega_p^2/\omega^2 \ , \quad \varepsilon_2 \cong \omega_p^2\nu/\omega^3 \qquad (\nu \ll \omega) \qquad (13)$$

The optical constants n and k for the Drude model are gotten by substituting eq. (10) in eqs. (9). The result is shown in Fig. 3. Various useful aproximations are summarized in Table I. For this purpose the spectrum is divided into five frequency regions.

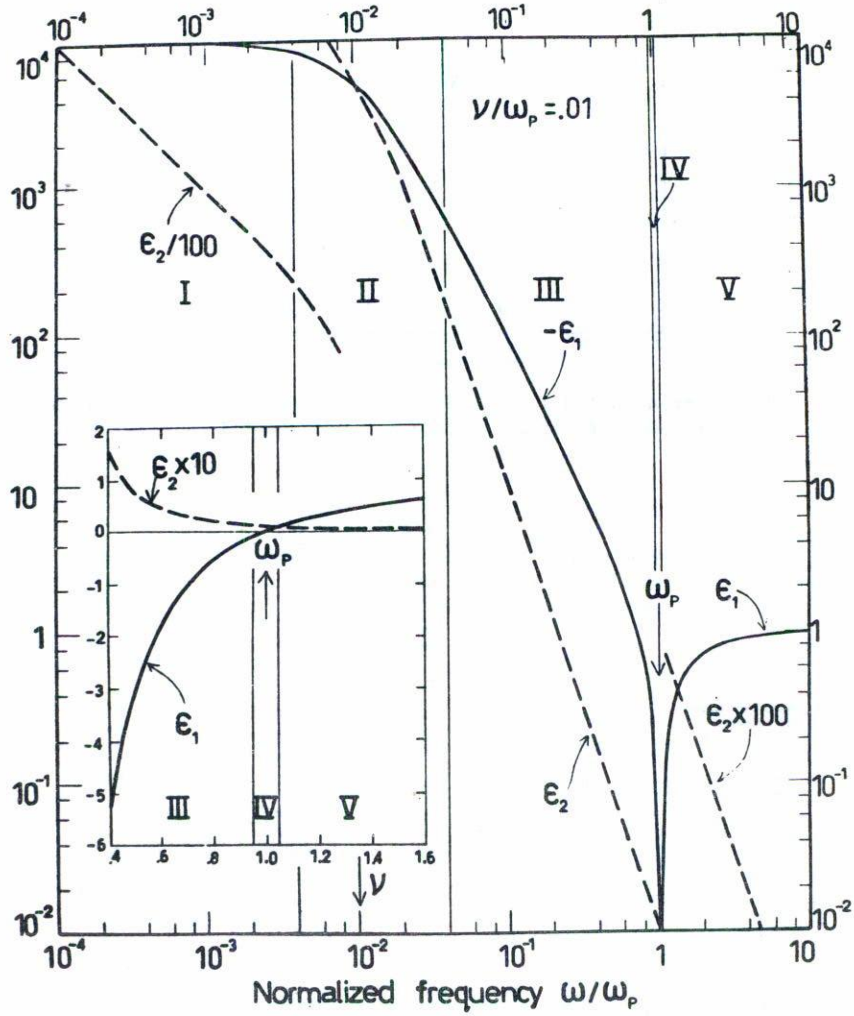

Fig. 2 Real part ε_1 (solid line) and imaginary part ε_2 (broken line) of the Drude dielectric function. Note the changes of scale and the change in sign of ε_1 from negative to positive in the vicinity of the plasma frequency. This is shown in more detail in the inset. The regions I-V are defined in Table I.

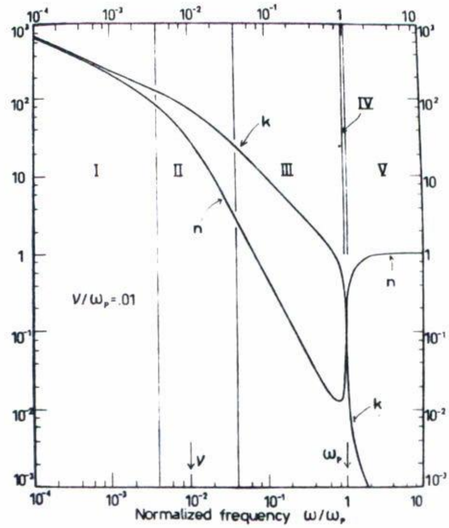

Fig. 3 Real part n (solid line) and imaginary part k (broken line) of the complex index of refraction for the Drude model of a metal.

Table 1. The optical constants of a typical metal classified according to frequency regions.

Region	Drude Model			Relation between ϵ_1 and ϵ_2	n	k	Relation between n and k
	Frequency range	ϵ_1	ϵ_2				
I	$\omega \ll \nu$ radio, microwave, far infrared	$-\frac{\omega_p^2}{\nu^2} < 0$	$\frac{\omega_p^2}{\omega\nu}$	$\epsilon_2 \gg \lvert\epsilon_1\rvert \gg 1$	$\sqrt{\epsilon_2/2}$	$\sqrt{\epsilon_2/2}$	$k \cong n \gg 1$
II	$\omega \sim \nu$ infrared (at room temperature)	$-\frac{\omega_p^2}{\omega^2+\nu^2} < 0$		$\epsilon_2 \sim \lvert\epsilon_1\rvert \gg 1$			$k > n \gg 1$
III	$\nu \ll \omega < \omega_p$ visible (not very near to ω_p)	$1 - \frac{\omega_p^2}{\omega^2} < 0$	$\frac{\omega_p^2\nu}{\omega^3}$	$\epsilon_2 \ll \lvert\epsilon_1\rvert$	$\frac{\epsilon_2}{2\sqrt{\lvert\epsilon_1\rvert}}$	$\sqrt{\lvert\epsilon_1\rvert}$	$k \gg n$
IV	$\omega \simeq \omega_p$ visible or ultraviolet		$\frac{\nu}{\omega_p}$	$\lvert\epsilon_1\rvert \ll 1$ $\lvert\epsilon_2\rvert \ll 1$			$k \sim n \ll 1$
V	$\omega > \omega_p$ ultraviolet (not very near to ω_p).	$1 - \frac{\omega_p^2}{\omega^2} > 0$	$\frac{\omega_p^2\nu}{\omega^3}$	$\epsilon_2 \ll \epsilon_1 < 1$	$\sqrt{\epsilon_1}$	$\frac{\epsilon_2}{2\sqrt{\epsilon_1}}$	$k \ll n < 1$

The most important limitations of the Drude model are the following: 1) It fails to describe adequately complex scattering processes, which cannot be lumped into a single, frequency-independent scattering frequency ν. 2) It ignores effects of spatial dispersion[(8),(9)] (or non-local effects) which can be described only by a dielectric function which depends explicitly on the wavevector $\vec{q}$, in addition to the frequency. These effects are characterized by a more general response than Eq.(2), namely

$$\vec{D}_{ef}(\vec{r}) = \int \varepsilon\,(\vec{r},\vec{r}\,')\,\vec{E}\,(\vec{r}\,')\,d^3r'$$

in the isotropic case. By "nonlocal" it is meant that $\vec{D}$ at a point $\vec{r}$ is dependent on $\vec{E}$ at other points $\vec{r}\,'$. If $\varepsilon\,(\vec{r},\vec{r}\,') = \varepsilon(\omega)\;\delta(\vec{r}-\vec{r}\,')$, where δ is the Dirac-delta function, then the "local" response $\vec{D}(\vec{r}) = \varepsilon\,(\omega)\;\vec{E}\,(\vec{r})$ is recovered. In this case the Fourier transform of $\varepsilon(\vec{r},\vec{r}\,')$ is independent of $\vec{q}$ and is given by $\varepsilon(\omega)$. "Nonlocal" effects are important when $q\ell/|1-i\omega\tau| \gtrsim 1$, where $\ell = v_F\tau$ is the mean free path of the charge carriers and v_F is the Fermi velocity. In the low-frequency region $\omega\tau<<1$ this condition reduces to $q\ell \gtrsim 1$, which means that the mean free path is of the order of, or larger than, the wavelength $2\pi/q$. The mean free path usually increases as the temperature is lowered. Therefore effects of spatial dispersion may be very important at low temperatures; the case of the anomalous skin effect is such an example.[(3)] In the high-frequency region $\omega\tau>>1$ nonlocal effects are pronounced for $qv_F/\omega \gtrsim 1$, i.e. when the phase velocity ω/q is of the order of, or smaller than, the Fermi velocity v_F. 3) Quantum mechanical effects such as band structure and the shape of the Fermi surface are ignored. The inclusion of these requires much more sophisticated theories. 4) A particular effect of band structure are interband transitions, which become extremely important at sufficiently high frequencies, usually in the visible range of the spectrum.[(7),(8)] Interband transitions may be powerful enough to cause a drastic deviation of the frequency for which $\varepsilon_1(\omega)$ vanishes from the value ω_p. They may also contribute a "tail" which reaches as far as the infrared region. Clearly the Drude model is limited to intraband transitions.

The limits of the Drude model are often pushed further by means of various generalizations. a) It is sometimes assumed that the collision frequency ν depends on the frequency ω. A dependence of the form $\nu = \nu_0 + \beta\omega^2$ is predicted on the basis of a theory of electron-electron scattering[10] and is helpful in accounting for certain observations for the noble metals.[11] b) Various groups of charge carriers, each one characterized by its plasma frequency ω_i and collision frequency ν_i may contribute to observed properties. Then Eq. (10) is replaced by

$$\varepsilon(\omega) = 1 - \sum_i \frac{\omega_i^{\,2}}{\omega(\omega + i\nu_i)} . \qquad (14)$$

c) If the ionic cores are polarizable then $\lim_{\omega\to\infty} \varepsilon(\omega)$ is not equal to 1 as predicted by Eq. (10). This shortcoming may be remedied by multiplying Eq.(10) by the factor ε_∞, the high-frequency dielectric constant. In this case the definition of the plasma frequency must be modified, to read $\omega_p = (4\pi ne^2/m^*\varepsilon_\infty)^{1/2}$. This procedure is also valid for highly doped polar semiconductors (such as InSb or GaAs) in a frequency region not very near to the transverse phonon frequency ω_T. In the latter case ε_∞ can be as large as 16.

The alkali metals, the noble metals, and a number of transition metals satisfy quite well the Drude model for frequencies sufficiently below the onset of interband transitions. Thus Eq. (10) is usually an adequate description of metals for infrared and lower frequencies. Moreover, the model may hold for very high frequencies (e.g. in the visible and the ultraviolet range), provided that the frequency of interest does not lie too near to an interband transition threshold.

4. INHOMOGENEOUS WAVES

Assuming that our medium is homogeneous, isotropic, linear and sourceless an arbitrary component ϕ of the electromagnetic fields satisfies the wave equation,

$$\nabla^2\phi - \frac{\varepsilon}{c^2}\frac{\partial^2\phi}{\partial t^2} = 0 . \qquad (15)$$

The simplest solution of this equation is a "plane wave",[12]

$$\phi = \phi_0 e^{i(\vec{q}.\vec{r} - \omega t)} \tag{16}$$

where the wavevector $\vec{q}$ is related to the frequency by means of the "dispersion relation"

$$q^2 = q_o^2 \varepsilon(\omega), \qquad q_o = \omega/c \,. \tag{17}$$

Here q_o is the vacuum wavevector. By Eq. (17), in the insulating medium (with dielectric constant ε_o) the wavevector is $q_o\sqrt{\varepsilon_o}$, and its component along the interface is (see Fig. 1)

$$q_x = q_o\sqrt{\varepsilon_o}\sin\theta_o \quad . \tag{18}$$

This component is continuous across the inteface; otherwise no boundary condition could be satisfied. We assume that the plane of incidence is the x-z plane, thus $q_y = 0$, without restriction of the generality. Then, by Eqs. (17) and (18), the normal component of the wavevector inside the conductor is given by

$$q_z^2 = q^2 - q_x^2 = q_o^2\,(\varepsilon - \varepsilon_o \sin^2\theta_o)\,. \tag{19}$$

Because ε is a complex quantity, so must be q_z. Its real and imaginary part are the solutions of the equations

$$q_{z_1}^{\,2} - q_{z_2}^{\,2} = q_o^2\,(\varepsilon_1 - \varepsilon_o \sin^2\theta_o) \tag{20a}$$

$$2q_{z_1}\, q_{z_2} = q_o^2\, \varepsilon_2 \,. \tag{20b}$$

We find

$$2(q_{z_{1,2}}/q_o)^2 = \left((\varepsilon_1 - \varepsilon_o \sin^2\theta_o)^2 + \varepsilon_2^2\right)^{1/2} \pm (\varepsilon_1 - \varepsilon_o \sin^2\theta_o) \tag{21}$$

Thus the wavevector components q_x and $q_z = q_{z_1} + iq_{z_2}$ are completely determined by the optical constants of the two media ($\varepsilon_o, \varepsilon_1$, and ε_2), the vacuum wavevector q_o, and the angle of incidence θ_o.

The meaning of a complex wavevector is readily understood by writing out Eq. (16) explicitly:

$$\phi = \phi_o e^{i(q_x x + q_{z_1} z - \omega t)} e^{-q_{z_2} z} = (\phi_o e^{-q_{z_2} z}) e^{i(\mathrm{Re}\vec{q}\cdot\vec{r} - \omega t)} , \qquad (22)$$

where

$$\mathrm{Re}\vec{q} = q_x \hat{x} + q_{z_1} \hat{z} \qquad (23)$$

is the real part of the wavevector. The imaginary part of $\vec{q}$ has only a normal component (q_{z_2}), thus

$$\vec{q} = \mathrm{Re}\vec{q} + i q_{z_2} \hat{z} \quad . \qquad (24)$$

We have a picture of a wave propagating in the direction of $\mathrm{Re}\vec{q}$ (which is determined by the ratio q_{z_1}/q_x) and attenuated in the direction $\hat{z}$. These two directions coincide only in the special case of normal incidence, $\theta_o = 0$. The planes of constant phase are perpendicular to $\mathrm{Re}\vec{q}$ and the planes of constant amplitude are perpendicular to the normal to the interface, $\hat{z}$. For $\theta_o \neq 0$, these sets of planes are <u>not</u> parallel to each other. A wave possessing this property is called "inhomogeneous": at a given instant t, different points on the wavefront have different amplitudes, because the attenuation depends on the depth z of material traversed.

For <u>insulating</u> materials usually $\varepsilon_2 \ll \varepsilon_1$ and therefore, $q_{z_2} \ll q_{z_1}$ for arbitrary θ_o. For this reason the problem of inhomogeneousness of the wave, although existent in principle, is not very important. On the other hand, the optical properties of conductors cannot be described in terms of ordinary or "homogeneous" plane waves.

5. PROPAGATION AND ATTENUATION IN THE CONDUCTOR

We shall first deal with the simple case of normal incidence, $\theta_o = 0$. Then $q_x = 0$ and Eqs. (21) reduce to

$$2(q_{z_{1,2}}/q_o)^2 = (\varepsilon_1^2 + \varepsilon_2^2)^{1/2} \pm \varepsilon_1 . \qquad (25)$$

By Eqs. (9) we get

$$q_{z_1} = q_o n \qquad \text{and} \qquad q_{z_2} = q_o k \ . \qquad (26)$$

In terms of the complex index of refraction we may write

$$q = q_z = q_o N \ . \qquad (27)$$

The wavelength is

$$\lambda = \frac{2\pi}{q_{z_1}} = \frac{2\pi}{q_o n} = \frac{\lambda_p}{n\omega/\omega_p} \ , \qquad (28)$$

where $\lambda_p = 2\pi c/\omega_p$ is the "plasmon wavelength". By Eq. (22) the amplitude of the wave falls off to 1/e of its value at the interface (z=0) after penetrating into the metal to a depth $z = 1/q_{z_2}$. This quantity is the "skin depth" δ ; for normal incidence,

$$\delta = \frac{1}{q_{z_2}} = \frac{1}{q_o k} = \frac{\lambda_p}{2\pi k\omega/\omega_p} \ . \qquad (29)$$

The normalized wavelength, λ/λ_p, and the normalized skin depth δ/λ_p are plotted in Fig. 4 for a Drude metal, with n and k given in Fig. 3. In the regions I-III we have $\lambda \gg \delta$ and, therefore, the wave is damped out before penetrating into the metal to a distance of the order of one wavelength. These regions are dominated by the "skin effect":[13] strong attenuation of the wave, rather than propagation into the interior of the conductor. On the other hand, in region V, $\lambda<<\delta$, meaning that the wave can complete many oscillations before being damped out. This is the propagation region and damping effects are quite small. Thus, above the plasma frequency, a metal becomes transparent. In the context of Solid State Physics this phenomenon is called the "ultraviolet transparency of metals".[3] It is caused by the fact that the real part of the dielectric function becomes positive and, therefore, the index of refraction ($N\simeq n\simeq\sqrt{\varepsilon_1}$) is basically real: the metal then behaves not unlike an insulator, as far as electromagnetic phenomena are concerned. We shall have to say more on the ultraviolet transparency of metals in Secs. 6 and 7.

Now we return to the case of an arbitrary angle of incidence θ_o. The wavelength is found from Eq. (23):

$$\lambda = \frac{2\pi}{|\mathrm{Re}\ \vec{q}\ |} = \frac{2\pi}{(q_x{}^2 + q_{z_1}{}^2)^{1/2}} \qquad (30)$$

where q_x and q_z are given by Eqs. (18) y (21), respectively. One may also define an index n', which gives the ratio of the speed of light to the phase velocity of the wave:

$$n' = c|\mathrm{Re}\ \vec{q}|/\omega = (q_x^2 + q_{z_1}^2)^{1/2} \ / \ q_o \qquad (31)$$

$$= \left[\varepsilon_1 + \varepsilon_o\sin^2\theta_o + \left((\varepsilon_1 - \varepsilon_o\sin^2\theta_o)^2 + \varepsilon_2^2\right)^{1/2}\right]^{1/2}\Big/\sqrt{2} \ . \qquad (32)$$

It is interesting that n', and therefore the phase velocity in the conductor, depend on the dielectric constant ε_o of the insulator, as well as on the dielectric constant $\varepsilon_1+i\varepsilon_2$ of the metal. In addition, the phase velocity depends on θ_o. This has been recently stressed by Ciddor.[14] However the effect is most pronounced under skin-effect conditions, when $q_{z_1} \lesssim q_{z_2}$.

The angle of refraction may be found from Fig. 1:

$$\theta = \tan^{-1}(q_x/q_{z_1}) \ . \qquad (33)$$

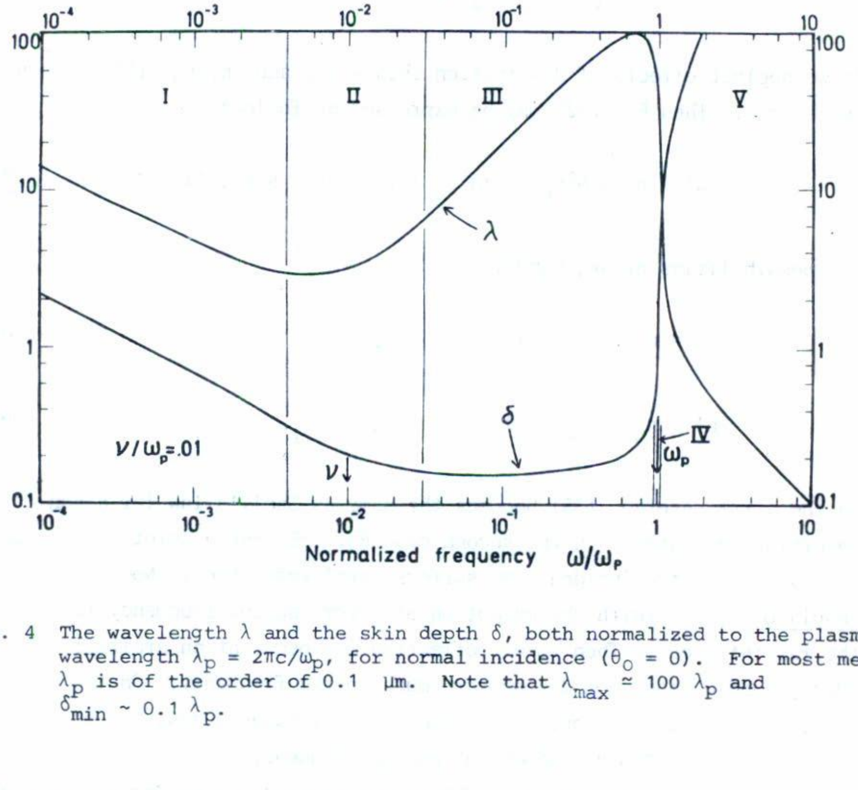

Fig. 4 The wavelength λ and the skin depth δ, both normalized to the plasmon wavelength $\lambda_p = 2\pi c/\omega_p$, for normal incidence ($\theta_o = 0$). For most metals λ_p is of the order of 0.1 µm. Note that $\lambda_{max} \simeq 100\ \lambda_p$ and $\delta_{min} \sim 0.1\ \lambda_p$.

By the continuity of q_x across the interface we must have

$$q_x = |\text{Re}\ \vec{q}|\ \sin\theta = q_o n_o \sin\ \theta_o\ . \qquad (34)$$

Hence Eq. (31) leads to

$$n' \sin \theta = n_o \sin \theta_o . \tag{35}$$

This is the generalization of Snell's law of refraction, valid for an absorptive on conductive medium. Note that n' reduces to n only under certain restrictions. We define the "critical angle", θ_c, by equation

$$\sin \theta_c \equiv \sqrt{\varepsilon_1/\varepsilon_o} \quad , \qquad \varepsilon_1 > 0 \; . \tag{36}$$

If we neglect effects of dissipation then k = 0 and, by Eq. (8), $\varepsilon_1 = n^2$ and $\varepsilon_2 = 0$. Then Eq. (32) may be expressed as follows:

$$n' = n_o(\sin^2\theta_c + \sin^2\theta_o + |\sin^2\theta_c - \sin^2\theta_o|)^{1/2}/\sqrt{2} \; . \tag{37}$$

Two possibilities arise, namely

$$n' = n \qquad \text{if} \qquad \theta_o \leq \theta_c \tag{38}$$

$$n' = n_o \sin \theta_o \quad \text{if} \qquad \theta_o \geq \theta_c \; . \tag{39}$$

In the first case Eq. (35) becomes the same as Snell's law for a non-absorbing insulator. In the second case Eq. (35) has a solution only for $\theta = 90^o$. Thus the incident ray suffers total reflection. Now, in an insulator $\varepsilon_1 > 0$ (with the exception of a very narrow frequency region in the far infrared). Then total reflection may occur at an arbitrary frequency, and θ_c does not depend very much on the frequency. On the other hand, in a metal $\varepsilon_1 > 0$ only above the plasma frequency and, by Eqs. (36) and (13), θ_c is strongly dependent on the frequency.

The reader may be puzzled by the fact that, in an insulator, total reflection occurs only when the ray is incident on an optically rarer medium, i.e. $n < n_o$. Isn't a metal optically denser than an insulator? The answer is that above the plasma frequency a metal is optically rarer even than vacuum! In fact, for $\nu \ll \omega_p$ we get from Eqs. (8) and (13),

$$n^2 \cong 1 - \omega_p^2/\omega^2 < 1 \quad \text{for} \quad \omega > \omega_p \ . \tag{40}$$

In this case, and $\varepsilon_o = 1$ (vacuum), Eq. (36) becomes

$$\sin \theta_c = \sqrt{1 - \omega_p^2/\omega^2} \ , \qquad \omega > \omega_p \ . \tag{41}$$

In practice, dissipative effects do exist. Because of this, as well as practical experimental considerations, the effect of total reflection is not easy to observe in metals.

Turning to the low-frequency regions I-III we find that the expression for the skin depth, Eq. (29) is also valid for a finite θ_o, provided that $\sin^2\theta_o << |\varepsilon_1|/\varepsilon_o$. In region I we get from Table I and Eqs. (29), (8) and (6)

$$\delta = \frac{1}{q_o\sqrt{\varepsilon_2/2}} = \frac{c}{\sqrt{2\pi\sigma_1\omega}} \ . \tag{42}$$

This is the well known expression for the "classical" skin depth.[3] In region I the optical conductivity is very nearly independent of ω and $\sigma_1 \simeq \sigma_o$, see Eq. (11). Therefore, in this region the skin depth is inversely proportional to the square root of the frequency. This behavior is well known for radio and microwave frequencies in metals at room temperatures. At very low temperatures, however, a drastic departure from the simple dependence $\delta \propto \omega^{-1/2}$ is observed. Then the anomalous skin effect[3],[15] is operative, and is caused by the fact that the mean free path of the electrons becomes of the same order or larger than the wavelength. As mentioned in Sec. 3, the Drude theory must be replaced by a more sophisticated, so-called non-local theory of conductivity.

Table II gives some useful expressions valid under restricted specified conditions. The Drude model, however, has not been invoked in the derivation.

Table II. Basic relations for refraction at a metallic surface*

Region	Assumptions	q_{z_1}/q_0	q_{z_2}/q_0	Relation between q_{z_1} and q_{z_2}	n'	θ
I, II, III	$\lvert\varepsilon_1\rvert >> \varepsilon_0 \sin^2\theta_0$	n	k	$q_{z_1} \lesssim q_{z_2}$	$(n^2+n_0^2\sin^2\theta_0)^{1/2}$	$\tan^{-1}\left(\frac{n_0}{n}\sin\theta_0\right)$
$\theta_0 > \theta_c$	$\eta^2 >> \varepsilon_2$	nk/η	η	$q_{z_1} << q_{z_2}$ $q_{z_1} << q_0$	$n_0 \sin\theta_0$	90°
$\theta_0 \cong \theta_c$	$\eta^2 << \varepsilon_2 << \varepsilon_0 \sin^2\theta_0$	$\sqrt{nk}$	$\sqrt{nk}$	$q_{z_1} \cong q_{z_2} << q_0$	$n_0 \sin\theta_0$	90°
$\theta_0 < \theta_c$	$\eta^2 >> \varepsilon_2$	n	nk/η	$q_{z_1} >> q_{z_2}$	n	$\sin^{-1}\left(\frac{n_0}{n}\sin\theta_0\right)$

* $\eta = \lvert\varepsilon_1 - \varepsilon_0 \sin^2\theta_0\rvert^{1/2}$

6. REFLECTIVITY

Formulas for the reflectivity, for the case described in Fig. 1, are derived in textbooks of electromagnetism and optics. These so-called Fresnel formulas are gotten by a straightforward application of Maxwell's equations and the usual boundary conditions at the interface. We shall quote the formulas for s- and p-polarized incidence: in the first case the electric field of the wave is parallel to the interface, while in the second case it is the magnetic field which is parallel to it. The results for the reflectivity are

$$R_s = \left| \frac{\sqrt{\varepsilon_0}\cos\theta_0 - \sqrt{\varepsilon - \varepsilon_0 \sin^2\theta_0}}{\sqrt{\varepsilon_0}\cos\theta_0 + \sqrt{\varepsilon - \varepsilon_0 \sin^2\theta_0}} \right|^2 \tag{43}$$

$$R_p = \left| \frac{\varepsilon \cos \theta_o - \sqrt{\varepsilon_o \varepsilon - \varepsilon_o^2 \sin^2\theta_o}}{\varepsilon \cos \theta_o + \sqrt{\varepsilon_o \varepsilon - \varepsilon_o^2 \sin^2\theta_o}} \right|^2 . \tag{44}$$

For normal incidence ($\theta_o = 0$), both formulas reduce to

$$R = \left| \frac{\sqrt{\varepsilon_o} - \sqrt{\varepsilon}}{\sqrt{\varepsilon_o} + \sqrt{\varepsilon}} \right|^2 = \left| \frac{n_o - N}{n_o + N} \right|^2 . \tag{45}$$

In particular, in region I (see Table I) we have $k \cong n \gg 1$, and Eq. (45) may be approximated by

$$R \cong 1 - \frac{2n_o}{n} \cong 1 - n_o \left(\frac{2\omega}{\pi\sigma_1} \right)^{1/2} . \tag{46}$$

The last expression was gotten by using Eqs. (6) and (8). Eq. (46) explains why metals are such good reflectors of light at low frequencies. Because $n \gg n_o$ the reflectivity deviates very little from one, the deviation being proportional to $(\omega/\sigma_1)^{1/2}$. This behavior is seen in Fig.5. The plasma edge at ω_p marks the onset of the "ultraviolet transparency" of metals. At very high frequencies $\varepsilon \to 1$ and, therefore, $R \to 0$.

It is evident from Fig. 5 that, at oblique incidence, the plasma edge is displaced to a higher frequency. We may derive this effect from Eqs. (43) or (44), assuming that ε_2 is negligible. If the second terms in both the numerators and denominators of these equations are imaginary, then the reflectivites are equal to one. For $\varepsilon_o = 1$ the condition is

$$\varepsilon_1 - \varepsilon_o \sin^2\theta_o \cong 1 - \frac{\omega_p^2}{\omega^2} - \sin^2\theta_o = \cos^2\theta_o - \frac{\omega_p^2}{\omega^2} < 0 . \tag{47}$$

We conclude that, for $\omega \leq \omega_p/\cos\theta_o$, an ideal metallic surface is perfectly reflecting ($R_s = R_p = 1$).

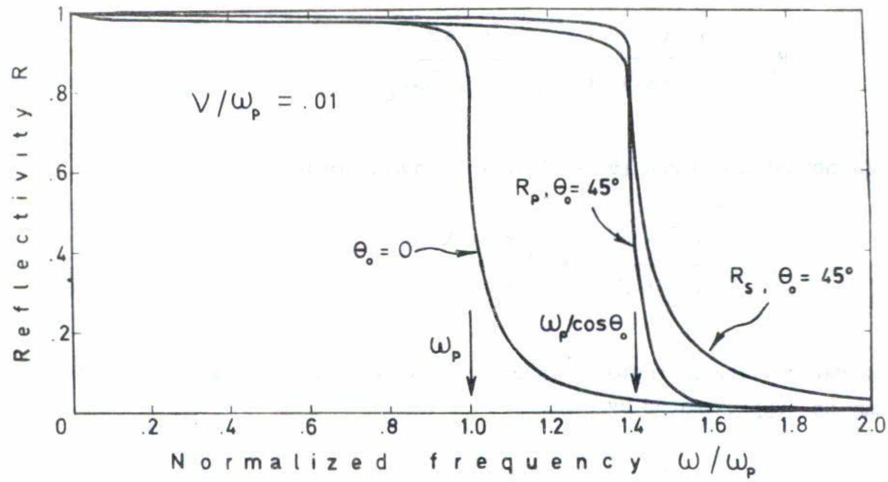

Fig.5 Reflectivity of a metallic surface for normal incidence ($\theta_0 = 0$) and oblique incidence ($\theta_0 = 45°$). In the latter case results are shown for s-polarized (R_s) and p-polarized (R_p) light. Note that, at oblique incidence, the plasma edge is displaced to a higher frequency, given by $\omega_p/\cos\theta_0$. These graphs correspond to an experiment with frequency scan.

The plasma edge also comes into play in the case of angular scan (Fig.6): the frequency is kept at a constant value, greater than ω_p, and the angle θ_0 is varied. Neglecting again ε_2, and using Eq. (36), Eq.(43) may be rewritten in the form

$$R_s = \left| \frac{\cos\theta_0 - \sqrt{\sin^2\theta_c - \sin^2\theta_0}}{\cos\theta_0 + \sqrt{\sin^2\theta_c - \sin^2\theta_0}} \right|^2 . \qquad (48)$$

For any frequency, above ω_p, the critical angle θ_c is a real angle, given by Eq. (36). Then, by Eq. (48), if $\theta_0 \geq \theta_c$ we have $R_s = 1$. (The same is also true for R_p). Of course, this is just what we would expect from the definition of the critical angle. (see discussion following Eq. (36)). The interesting point is that, for $\varepsilon_0 = 1$, the condition $\theta_0 \geq \theta_c$ is identical with the condition $\omega \leq \omega_p/\cos\theta_0$ (or Eq. (47)). Thus we are led to the understanding that, for a finite angle θ_0, a metal is perfectly reflecting (if ε_2 is neglected) between the frequencies ω_p and $\omega_p/\cos\theta_0$ because total reflection takes place. Of course, it is also perfectly reflecting below ω_p, although for a different reason. In this frequency region $\varepsilon < 0$ and no electromagnetic fields can penetrate into the interior of the metal due to the screening effect of the free electrons at the surface.

We also note that R_p, Eq. (44), vanishes for an angle given by $\tan\theta_o = (\varepsilon/\varepsilon_o)^{1/2}$. The RHS of the last equation defines the well known Brewster angle ($\tan\theta_B = n/n_o$) of optics. Clearly, $\theta_B < \theta_c$. While the minimum in R_p is too small to be notable on the scale of Fig.6(a) (with $\omega = 2\omega_p$), it is very important in the case of Fig. 6(b) (with $\omega = 1.1\,\omega_p$).

7. PLASMONS

In the foregoing sections we were discussing waves which may propagate in a metallic medium when an external wave is incident at the surface of this medium. Therefore the waves in the conductor correspond to forced oscillations, driven by the incident wave. In the absence of damping effects these waves are transverse, i.e. the fields $\vec{E}$ and $\vec{H}$ are both perpendicular to $\vec{q}$. Now consider an obliquely incident and p-polarized wave. Tne normal component fo the displacement vector is continuous across the interface and therefore $D_{zo} = \varepsilon(\omega)\,E_z$. This equation may be satisfied with $D_{zo} = 0$, $\varepsilon(\omega) = 0$, and $E_z \neq 0$. In words: with no external excitation whatsoever the medium supports an oscillating electric field at a frequency such that the dielectric function vanishes. According to Eq. (13), in the absence of damping, this happens at the plasma frequency, $\omega = \omega_p$. This is then a natural frequency of the system, and the corresponding "plasma oscillations" are normal modes. If $\varepsilon(\omega)$ is given by Eq. (10) then the equation $\varepsilon(\omega) = 0$ has the approximate root $\omega = \omega_p - i\nu/2$. This means that the plasma oscillations decay with a time constant $2/\nu = 2\tau$.

It is not difficult to see that the plasma oscillations -unlike the electromagnetic waves studied in the previous sections- are longitudinal, i.e. the electric field oscillates in the direction of propagation. Indeed, it follows from Eq. (1b) that

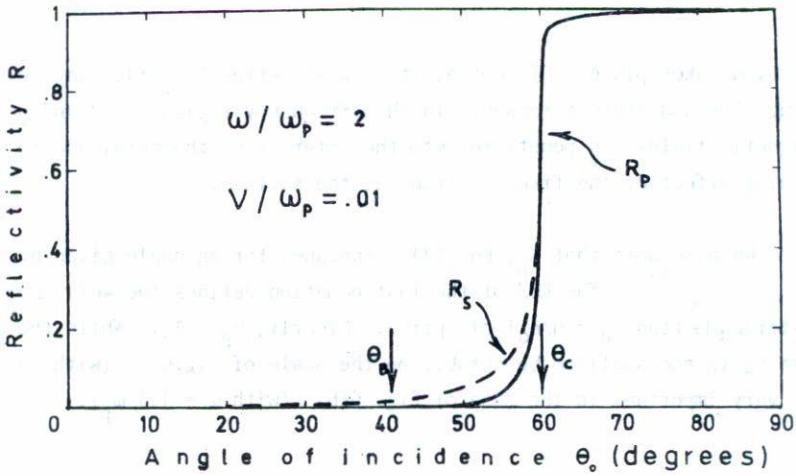

Fig.6.a Reflectivity of a metallic surface at a constant frequency $\omega = 2\omega_p$: angular scan for p-polarized (solid line) and s-polarized (dashed line) ligth. Note that the results for R_p and R_S differ appreciably only in a limited frequency region. Almost all the light is reflected for angles larger than the critical angle θ_C. There is a minimum in R_p at the Brewster angle θ_B which, however, is not notable on the scale of the figure.

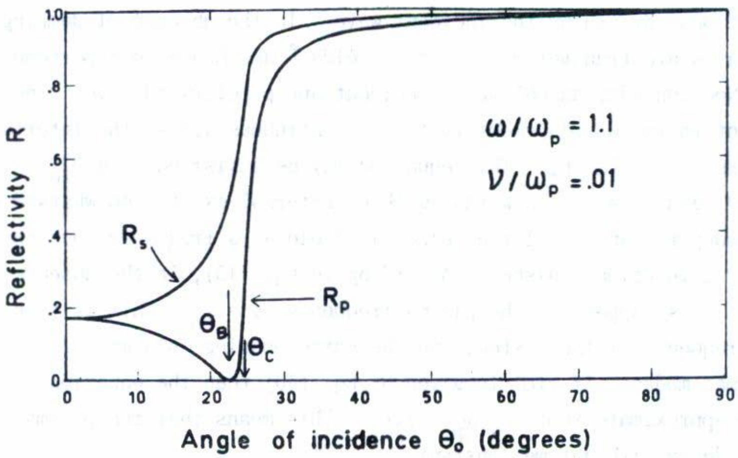

Fig.6.b Same as (a) for $\omega = 1.1\omega_p$. The Brewster minimum causes R_p to deviate qualitatively from R_S.

$$\vec{\nabla} \cdot \vec{D}_{ef} = 0 = \varepsilon(\omega)\ i\vec{q}\cdot\vec{E}\ . \tag{49}$$

When $\varepsilon(\omega) = 0$ we may have $\vec{q}.\vec{E} \neq 0$, i.e. Eq. (49) is consistent with an electric field component along the wavevector. The formal proof follows from the following considerations. If $\varepsilon(\omega) = 0$ then the RHS of Eq. (1b) vanishes. Therefore we have $\vec{\nabla}\times\vec{H} = 0$ and $\vec{\nabla}\cdot\vec{H} = 0$, as well. These two equations imply a spatially constant, or vanishing, magnetic field. We are not interested in the first possibility, so we shall take $H = 0$. Then it follows from another Maxwell's equation, that

$$\vec{\nabla} \times \vec{E} = -\frac{1}{c}\frac{\partial\vec{H}}{\partial t} = 0\ . \tag{50}$$

Therefore, $\vec{q}\times\vec{E} = 0$ i.e. $\vec{E}||\vec{q}$. It is interesting that, unlike in other electromagnetic wave phenomena, the oscillations of $\vec{E}$ are not accompanied by oscillations of $\vec{H}$. Thus the plasma oscillations are an electric phenomenon.

The dispersion relation, ω versus q, of the plasma waves is shown in Fig. 7 (labeled L). The horizontal line exhibits "dispersionless" behavior, i.e. ω = const. However, this simple result is shortcoming of our local theory (see Sec.3). In a nonlocal theory ε also depends on the wavevector, and the equation $\varepsilon(\omega,q) = 0$ has a q-dependent solution for ω. A small-q expansion gives the following result:(3),(8),(15)

$$\omega^2 = \omega_p^2 + (3/5)\ q^2 V_F^2\ . \tag{51}$$

It is obvious that plasma modes may be excited in a metal only by p-polarized light, which possesses a normal component, E_z, of the electric field. Melnyk and Harrison(9) have developed a theory of plasmon excitation in metals. They suggest that the effect is amplified in thin metallic films; in this case nonlocal effects play an important role. Lindau and Nilsson(9) have successfully carried out the corresponding experiment.

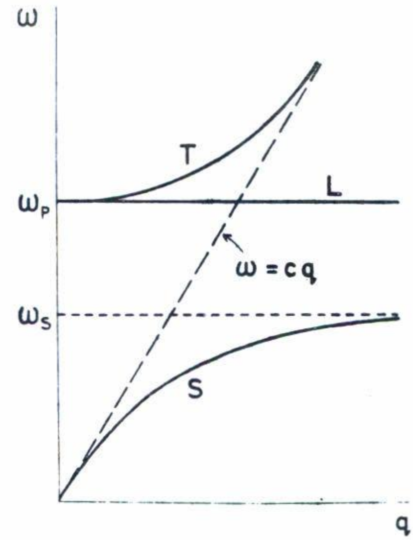

Fig. 7 The fundamental electromagnetic modes of a simple metal (schematic): transverse bulk (T), longitudinal bulk (L), and surface (S) mode.

The dispersion relation of the transverse waves is given by Eq.(17). Upon substituting Eq. (13) and rearranging, Eq. (17) may be written in the form

$$\omega^2 = \omega_p^2 + c^2q^2 \tag{52}$$

The last result makes it particularly clear that propagation of electromagnetic waves (with real q) in a metal is possible only for $\omega > \omega_p$. This fits neatly into the picture of the plasma edge and the ultraviolet transparency. The transverse solutions (52) are shown in Fig. 7 (labeled T). In the limit of very high frequencies the free electrons cannot follow, any more, the oscillations of the electric field. Then the propagation in the metal becomes like propagation in vacuum, and the dispersion relation approaches the "light line".

8. SURFACE PLASMON-POLARITONS

In the preceding section we limited the discussion to bulk modes of the metallic medium. This means that the boundary and the adjacent insulating medium had no effect at all. We shall now show that the interface between a metallic and an insulating medium supports certain normal modes of the electromagnetic field. They are known as "surface plasmon-polaritons". (Several review articles are listed in Ref. 16).

We rewrite Eq. (17) in the form

$$q_x^2 + q_z^2 = q_o^2 \varepsilon . \tag{53}$$

The corresponding equation for the insulator reads

$$q_x^2 + q_{zo}^2 = q_o^2 \varepsilon_o . \tag{54}$$

We have used the fact that q_x, unlike q_z, is continuous across the interface. Next we take the x-component of Eq. (1b):

$$q_z H_y = - q_o \varepsilon E_x . \tag{55}$$

Because E_x and H_y are continuous, for the insulating medium we write

$$q_{zo} H_y = - q_o \varepsilon_o E_x . \tag{56}$$

Dividing Eqs. (55) and (56) we find

$$\frac{q_z}{q_{zo}} = - \frac{\varepsilon}{\varepsilon_o} . \tag{57}$$

Eqs. (53), (54), and (57) may be solved for the three unknowns q_x, q_z, and q_{zo}. The results are

$$q_x = q_o \left(\frac{\varepsilon\ \varepsilon_o}{\varepsilon + \varepsilon_o}\right)^{1/2} \tag{58}$$

$$q_z = q_o \ \frac{\varepsilon}{(\varepsilon + \varepsilon o)^{1/2}} \tag{59}$$

$$q_z = q_o \ \frac{\varepsilon_o}{(\varepsilon + \varepsilon_o)^{1/2}} \ . \tag{60}$$

We shall neglect dissipation, assuming that ε is real. Of course, ε_o is real and positive. Then we can see from Eqs. (58)-(60) that, in a frequency region such that $\varepsilon(\omega) < - \varepsilon_o$, both q_z and q_{zo} are imaginary, however q_x is a real quantity. Such a state of affairs has the following interpretation. We have an electromagnetic mode which propagates in the x-direction (in the plane of the interface) with a wavevector given by Eq. (58). Its amplitude is maximal at the interface and falls off exponentially away from it, with decay constants given by Eqs. (59) and (60) in the respective media. It should be stressed that this decay is not caused by dissipation of energy. Indeed, we have "switched off" the phenomenological damping ($\nu = 0$), by assuming that ε is real. Thus the exponential decay is an intrinsic property of the mode, in fact just the property that makes it a surface mode.

If we substitute Eq. (10) in Eq. (58) and solve for ω versus q_x we get a dispersion relation for surface plasmon-polaritons. It is drawn schematically in Fig.7 (labeled S). The limiting frequency, ω_s, of these excitations is gotten from the condition that the denominator of the square root in Eq. (58) vanishes. Neglecting dissipation we have

$$\varepsilon + \varepsilon_o = 1 - \omega_p^2 /\omega^2 + \varepsilon_o = 0 \tag{61}$$

or

$$\omega_s = \omega_p/(1 + \varepsilon_o)^{1/2} \ . \tag{62}$$

If the metal is bounded by vacuum the RHS reduces to $\omega_p/\sqrt{2}$. This is the well known surface plasmon frequency. It is a natural frequency of oscillation of the plasma at the surface, just as ω_p is a natural frequency of oscillation of the bulk plasma. Both the electrons and the electric field oscillate in the x-z plane. The tip of the vector $\vec{E}$ traces an ellipse in this plane, at any point as a function of time (or, at any instant, as a function of x).

At this point the reader may wonder whether surface plasmon-polaritons may be excited at a metallic surface by direct incidence of light. The answer, at least for a smoooth surface, is "no". We may find the reason by suitably rewriting Eq. (58):

$$q_x = q_o\sqrt{\varepsilon_o}\left(\frac{|\varepsilon|}{|\varepsilon| - \varepsilon_o}\right)^{1/2} > q_o\sqrt{\varepsilon_o} \quad . \tag{63}$$

For simplicity we assume that our metal is bounded by vacuum. Then Eq.(63) becomes $q_x > q_o$, which reads: the wavevector of the surface plasmon-polariton must be larger than the wavevector in vacuum. On the oter hand, for an obliquely incident wave, $q_x = q_o \sin\theta_o < q_o$. We conclude that direct excitation is not possible because it would violate the law of conservation of momentum (of the light).

The missing momentum may be supplied in a more sophisticated, Attenuated Total Reflection (ATR), experiment. It involves three media: a prism, a thin metallic film, and air (Fig.8). The prism boosts the wavevector of the incident light to a value $q_o n_p$, where n_p is the refractive index of the prism (usually between 1.5 and 4). Then $q_x = q_o n_p \sin\theta_o$. This may be made larger thar q_o, as required by Eq. (63), provided that $n_p \sin\theta_o > 1$. Therefore, the surface polaritons may be excited only for angles of incidence which are greater than the critical angle between the prism and the air.

In the geometry of Fig.8 the incident wave actually tunnels through the thin film and excites a plasmon-polariton at the free metallic surface. An optimum choice of the film thickness leads to experimental dispersion relations which usually fit the theoretical one (for two media), Eq. (58), very well. Of course, the light must be p-polarized, because the excitation involves the component E_z, as well as E_x. The configuration of Fig.8 is known as the "Kretschmann[17] geometry"; it has been suggested by Simon[18] for an experiment by undergraduate students. An alternative configuration

-the "Otto[19] geometry"- involves an air gap between a prism and a massive metallic film. An experiment for students in this geometry was described by Barker[20].

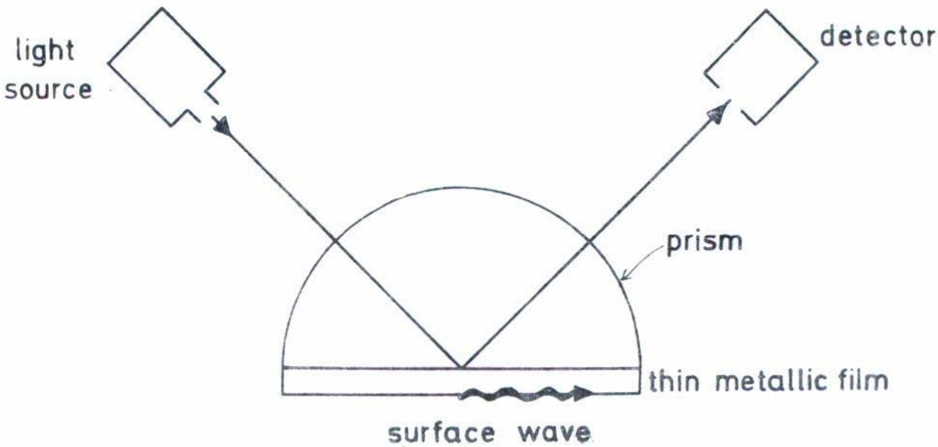

Fig. 8 Schematic representation of the excitation and detection of surface electromagnetic waves (surface polaritons).

Fig.9 shows the result of theoretical simulation of an optical experiment in the geometry of Fig.8.[21] Note the dramatic minimum in the reflectivity for p-polarized light, which is completely absent for s-polarization. Indeed, the minimum occurs at an angle greater than the critical angle between the prism and the air which bound the thin metallic film. Roughly speaking, the energy corresponding to the difference between R_s and R_p at the minimum has gone into the excitation of a surface plasmon-polariton. In Fig.9 the frequency ω is kept constant; for this frequency the wavevector of the excitation is gotten from the equation

$$q_x = q_o n_p \sin \theta_{min} \tag{64}$$

Repeating the same procedure for other values of ω one may trace "experimentally" the entire dispersion curve ω versus q_x. This technique, and the resulting curve are shown in Fig. 10. The result is quite similar to the curve

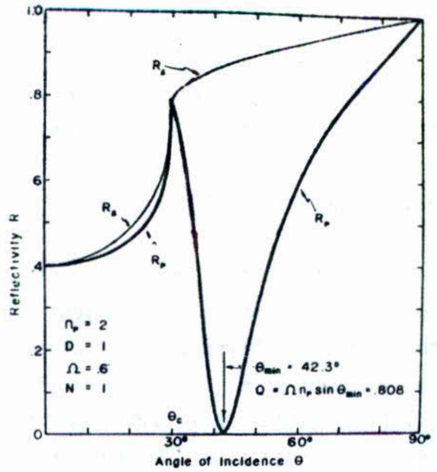

Fig.9 The reflectivities of s- and p-polarized light as a function of the angle of incidence (at constant frequency) in the experimental configuration of Fig.8[21]. The deep minimum in the R_p curve is a result of surface plasmon excitation. Definitions: $D = d\omega_p/c$, where d is the film thickness, $\Omega = \omega/\omega_p$, $N = \nu/\omega_p$, $Q = q_x c/\omega_p$.

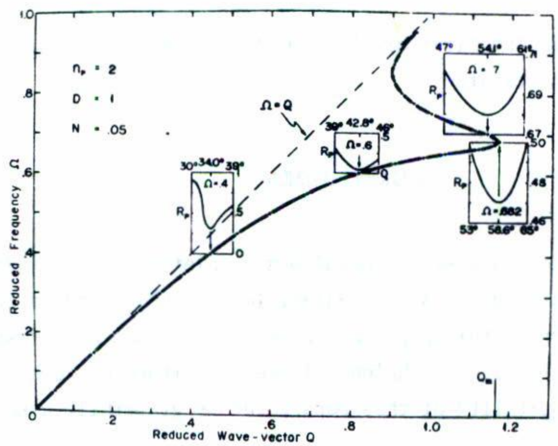

Fig.10 The dispersion relation for surface plasmons determined by simulating an optical experiment in the geometry of Fig.8. The arrows in the four insets at points along the dispersion curve show that these have been determined from the minima of the R_p versus Q curves (at constant Ω). The scales of the Q variables of the insets are identical with that of the main plot. Note the backbending at $Q = Q_m = 1.165$. The "light-line" $\Omega = Q$ (broken line) demonstrates that the entire dispersion curve lies in the "non-radiative region". (From Ref.21).

labeled S in Fig.7. However, as $q_x \to \infty$ the limit $\omega \to \omega_s$ is not attained. Rather, the dispersion curve bends back at a limiting value of the wavevector and a frequency slightly lower than ω_s. The reason for this effect lies in the damping of surface plasmon-polaritons[22] (in Figs. 9 and 10 finite values of ν/ω_p were chosen).

We conclude with an interesting point, attention to which was called by Cardona[23]. The surface polariton is an undriven oscillation of the electromagnetic fields at the surface. Therefore it corresponds formally to a reflectivity problem with no incident wave, i.e. to an infinite reflectivity. Then the denominator of Eq. (44) should vanish under conditions which give rise to the existence of a surface polariton. We use Eqs. (53), (54), and (18) to prove that this is, indeed, true. The denominator of Eq. (44) may be then expressed as follows:

$$\varepsilon \cos\theta_o + \sqrt{\varepsilon_o\varepsilon - \varepsilon_o^2 \sin^2\theta_o} = (\varepsilon q_{zo} + \varepsilon_o q_z)/(q_o\sqrt{\varepsilon_o})$$

The expression in the first brackets on the RHS vanishes when Eq. (57) is satisfied, as anticipated.

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