SURFACE ELECTRODYNAMICS II:
COLLECTIVE AND SINGLE PARTICLE MODES IN METALS

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ABSTRACT

Some aspects of Surface Electrodynamics that include non-local effects are presented in a simple form. In particular, surface polaritons and plasmons and the effect coming out from the excitation of electron-hole pairs are introduced and discussed. Longitudinal and transverse response functions are considered in a simple way. Collective and single excitations are illustrated with some calculations. The non-local effect is divided in two terms: a pure surface effect and a surface-induced bulk contribution.

RESUMEN

Se presentan, en términos sencillos, algunos aspectos de la Electrodinámica de Superficies que incluye efectos no locales. En particular, se introducen y discuten los polaritones de superficie y los plasmones y el efecto resultante de la excitación de pares electrón-agujero. Se consideran en forma simple las funciones respuesta longitudinal y transversal. Las excitaciones individuales y colectivas se ilustran con algunos cálculos. El efecto no local se divide en dos términos: un efecto puramente superficial y una contribución del bulito inducida por la presencia de la superficie.
I. INTRODUCTION

Recent progresses in Surface Science have made possible an accurate description of the interaction between an external electromagnetic (EM) field and the bulk taking into account the selvedge. Although the experimental settlement is very difficult, different techniques have been developed to the point of discerning between very fine effects. The source of the field can be a laser, a gun of monoenergetic electrons or any other source of energy and momentum collimated particles. As a matter of fact, the interaction takes place through the surface of the solid. Therefore, both a proper description and a careful preparation of the surface region are of paramount importance to investigate bulk-selvedge-probe coupling. The sample has to be preserved from contamination under very specific conditions. Particular efforts have been invested during the last decade to study the EM excitation of metals. One knows from these studies that the surface region, i.e., the region through which an external field penetrating the metal heals to its bulk value, extends several angstroms inside the metal, although it is difficult to define it unambiguously. This region can be an order of magnitude larger than the surface density profile where the surface induced electronic density piles up, as has been discussed in a previous paper /1/ herein referred to as I.

In spite of practical difficulties to carry out the experiments, measurements of, e.g., photocurrents /2/ and EEL (electron energy losses) /3/ on metals reveal how important the inclusion of different non-local effects is to account for the surface presence properly. They show also that one requires very often of a self-consistent treatment of the surface electronic structure to predict results in agreement with the experimental findings. Self-consistent calculations have been carried out using the simplest minded picture, i.e., the jellium model, to represent the metal. The most realistic calculations have used for a long time a random phase approximation (RPA) /4,5/. Simple models give results differing sometimes by orders of magnitude from the experimental data /4/.

Non-local optical and electronic properties of metal surfaces are not only important in fundamental studies. They are of interest in understanding a number of related phenomena when characterizing clean and adsorbate covered surfaces and interfaces. These can be analyzed with different probes, such as external charges, EM radiation or neutral atoms. Spectroscopic techniques using incident electrons, such as low-energy electron diffraction (LEED) /6/, electron-energy loss spectroscopy (EELS) /7/, or high-resolution electron loss spectroscopy (HREELS) /8/, are well known in the literature. Incident EM radiation can be used to study the standard optical properties. Some of these tools are differential reflectance /9/, ellipsometry /10/, plasmon dispersion /11/, surface-enhanced spectroscopies /12/, ultraviolet photoemission /13/, etc (cf. references in I). In all these phenomena one must understand the role of normal-mode excitations. Plasmon modes have received a great deal of attention, however we will see that single-particle modes need a more detailed study. Physi- and chemisorption processes on metals is a subject of very active research /14/. Their effects on optical properties are also of major interest (cf. Paper V of this series).

Despite the importance of their response to external probes, our present understanding of the dynamic behavior of realistic systems like finite size samples, transition metals, rough surfaces, etc., is far from complete. One even finds, e.g., that the analysis of modest experiments as the reflection of light by a smooth semi-infinite free-electron (FE) like metal, has not been carried out in
a satisfactory way. More generally, the long-standing problem of the subtle relationship between bulk and surface effects during the electronic excitation of metals is a subject where comparatively small progress has been achieved.

The purpose of this paper is to discuss the basic physics of the normal-mode excitations of a surface system. External charges, that produce longitudinal excitations, and EM radiation, that can produce both longitudinal and transverse modes, are the prototypes of the external stimuli to which the system responds and exhibits its normal-mode spectrum. This structure is contained in the response functions. It is of worth to discuss in a simple understandable manner how to obtain some of these. In particular the reflection coefficient, the longitudinal and transverse dielectric functions and the surface impedance are examined. It will be seen that they can be simplified without losing the essential physical features of interest for the surface problem.

Explicit results for the low-energy absorption due to electron-hole (e-h) pair excitation in simple metals are presented. The pure surface effect arises in the strong variations of the field within the very surface region of the solid. Its effect is analogous to the atomic case where a rapid variation of the field within the extension of the atom makes the dipole approximation invalid and gives rise to extra absorption /15/. On top of all of this one must consider interband transitions as direct bulk excitations. These can take place whenever the incident EM field provides enough energy and momentum to allow the transition from one band to another or into vacuum. A proper treatment of this type of transitions is rather complicate. It is not considered in this report. The intraband volume term, on the other hand, is usually neglected. It will be seen that it is manageable within reasonably simple physical models. Its contribution to absorption originates on the excitation of e-h pairs in the interior of the metal due to the surface presence. This contribution extends previous formalisms and presents an illustrative derivation of the surface induced absorption in FE like metals, whether perturbed by photons or electrons. We plan to carry out further applications taking into account both effects and their interference.

Bulk effects are dominant at low frequencies, as self-consistent calculations reveal /4/. To emphasize this feature is one of the purposes of the present contribution. The interaction with a bulk medium is studied first, raising the question of simplifying the dielectric functions and the surface problem, with emphasis on the effects of non-local response or spatial dispersion. The essential on the physics of the normal-mode excitations can be discussed in terms of the semiclassical infinite-barrier (SCIB) model, which is used in some sections.

The paper is divided in the following way. In section 2 a qualitative picture of single and collective modes is given. The section starts with a brief historical preview. Section 3 contains an introduction to some response functions of major use in surface electrodynamics. In section 4 the problem of absorption and reflection when non-local effects are present is focused. Section 5 is addressed to surface collective modes. The paper ends with a discussion of the results and further comments.

2. BULK AND SURFACE EFFECTS ON SINGLE AND COLLECTIVE MODES

a. Historical Background

To discern between bulk and surface effects when light interacts with metals has
been an attractive question since very early. The qualitative understanding has been based on the FE model of a semi-infinite metal provided with a discontinuous (step-function) potential. In the analysis of photoemission data, Tamm and Schubin are the first persons to our knowledge pointing out the importance of the separation between bulk and surface /16/. They attribute the surface effect to the rapid changes of the potential energy at the surface of the metal and the volume contribution to the internal variations of the potential. In a later work, Mitchell gives a more proper description of the surface photoeffect /17/. He assumes that the bulk contribution is small near the threshold. After these early works /18/ it has been explicitly assumed that photoemission from a FE metal is a pure surface effect, although it is clear that in general one must distinguish between the surface contribution and the volume photoelectric effect which takes place directly (interband transitions) or indirectly /19/. A noticeable exception will be considered later in this paper /20/.

Regarding the optical process of absorption and reflection, first attempts were addressed the other way around. Reuter and Sondheimer (RS) tried to incorporate in a bulk-like absorption mechanism the intricate relationship between the fields and the induced current /21/. The connection between absorption and reflection as a function of frequency was elucidated to a great extent in terms of a penetration length and a skin region. In their study, remarkable progress was achieved for the understanding of the anomalous absorption (AA) that appears at frequencies very well below the plasmon mode. Their analysis for s-polarized light has even a quantitative meaning, though their conclusions regarding p-polarization are not valid /22/. A surface pure effect due to the induced electronic density brings additional absorption mechanisms. Further work /23-26/ have tried to incorporate other effects within the SCIB model. This point is further examined in this contribution.

There is another subject in which the separation between bulk and surface effects has deserved interest since very early, viz electron spectroscopy /27-35/. First efforts led to the discovery of plasma waves excited by fast electrons passing through a metal. In this process the quantum character of the oscillation plays an essential role /29/. Experiments on the reflection of electrons by metal surfaces were carried out in the earliest thirties by Rudberg /27/. Some years later Rutheman and Lang /28/ performed careful measurements on energetic electrons fired through a thin metallic film. They measured both the energy of the incident and of the emerging electrons. What was surprising was the sharp energy transfer to the metallic electrons in certain metals. These quanta of plasma were predicted by Pines and Bohm in 1951 /29/. As Mott pointed out /30/, due to the high frequency of the excitation the electrons do not pay attention to the periodic ion potential. In other words, at those high energies they do not distinguish between the ions in a periodic array and a uniform positive background. In this collective motion inside the solid there is no qualitative distinction between metals, insulators and semiconductors. The extension to surface plasmons associated with the waves of charge bound at a vacuum-solid interface was done by Ritchie /31/. Subsequent experiments provided ample evidence for the existence of these collective modes as well /32/.

The coupling between slow electrons and the metal is greatly facilitated by the longitudinal character of the electric field coming out from the external electron /3,33/. When the energy of the incident electrons is very low (1 eV or less), the energy-loss spectra are explained in terms of e-h pair excitation /34/. In these cases the effect of a smooth surface is of paramount importance. One could expect
that the charge fluctuations within the density profile region would provide the most efficient mechanisms for energy dissipation. This is not necessarily the case, as recent experiments suggest /3/.

b. Surface EM Waves and Surface Plasmons

We consider here an idealized situation in which the ions are fixed and the electrons are excited individually (e-h pairs) or collectively. These are the normal modes of the electron gas. Single-particle excitations have a pure quantum origin. Collective modes, on the other hand, may be considered classically. Nevertheless, non-local effects are very important in surface studies. This paper is limited to non-interacting or weakly-interacting electrons. However, it should be noticed that in a collective mode the electrons do not behave as independent particles: their mutual interaction leads them to screen out any charge disturbance. Some elementary excitations, as magnons, phonons and excitons, are outside the scope of this contribution.

The motion of a particle in a plasma is accompanied by a cloud of other particles. They move along with it in such a way as to screen out the electric field produced by its charge. This is a microscopic screening. In addition, the motion of electrons can carry out long wave-length, high-frequency oscillations, which involve the coherent motion of many thousands of particles. The Coulomb interaction is efficient in suppressing the density fluctuations of charge due to the availability of many highly mobile free electrons. This is the macroscopic screening in an electron plasma. Those and other responses of the system constitute the normal-mode excitations of the plasma.

When low-energy electrons or photons interact with the electron plasma transferring momentum below a certain cut-off value, collective oscillations of the plasma can be excited. For incident electrons, these oscillations propagate as longitudinal charge-density fluctuations through the volume (bulk plasmons) or along the surface (surface plasmons) of the metal. Light can produce longitudinal and transverse excitations.

An exciting EM wave travelling through a polarizable medium is modified by the polarization it induces and becomes coupled to it. For an electron plasma the resulting excitation has both a photon and a plasmon content. This coupling mode is called a surface-plasmon polariton. It is associated with some form of guiding surface and may be completely bound in the sense that their associated fields decay exponentially in the normal direction (local mode), or unbound, in which case it would be maintained by balancing the energy radiating away from the surface with energy radiating to the surface.

In Sec.5 we study two extreme regimes, namely the very strong photon and the very strong plasmon content. The familiar plasmon mode is a bulk polariton. The surface polariton in the same state corresponds to surface plasma oscillation. It is indeed a coupling between light and the surface. Should a photon couple to the lattice vibrations in the crystal, a phonon-polariton would be produced. Excitation of surface phonons by electrons in plane surfaces has been studied recently /34/. Light in the far infrared can couple to the surface ions in rough surfaces /35,36/. (cf. paper III)

The surface EM modes may have in principle TE (s) and TM (p) modes. Although the TE solution is the only surface mode for magnon systems, it is not an acceptable solution for non-magnetic materials. These systems do accept indeed a TM mode.
However if the momentum transfer exceeds the value of $q$, there are no more collective modes of the plasma excited. The plasma decays into $e-h$ pairs, and the final result are single-particle excitations. The latter cannot be described in a classical scheme, as it has been discussed in I and will be illustrated here.

As can be seen, coupling between the salvedge and the bulk is very important in the study of collective excitations. The importance of the surface EM waves and plasmon excitations arises in that they serve as a sensitive probe of the structure of material surfaces, since such coupled modes provide information about the characteristic quantities used to describe the medium. One could extract more valuable information regarding surface properties if one were able to treat the interaction between these excitations in a more proper way. In particular, a knowledge of the different contributions from bulk and surface is of paramount importance.

c. Electron-hole Pairs Bulk Production and the Anomalous Skin Effect.

We consider also in this paper AA induced by photons and electrons on FE like metals. It is a very-well known fact that excitation of free electrons is not possible without violating energy and momentum conservation principles. Therefore absorption is regarded as taking place, e.g., at the nuclei positions within the bulk during the EM wave propagation as a result of phonon excitations. Impurities, dislocations and other imperfections serve as scattering sources during the electronic motion. This is the ordinary Joule heating. It is the only one persisting at high temperatures. In the absence of collisions, all radiation is reflected at frequencies below the plasma frequency. For frequencies above it, the material is perfectly transparent to radiation. The introduction of a periodic structure breaks the translational symmetry and interband transitions are made possible in metals.

There is another excitation mechanism that comes about due to a broken symmetry. In a half-space medium the component of linear momentum perpendicular to the surface is not a good quantum number any longer. As a result, when using the FE model to describe a semi-infinite metal, the surface presence gives rise to new absorption processes in a homogeneous electron gas. Therefore it is convenient to distinguish between two different effects on the response of a metal bounded by a surface. There is a surface-induced electronic density whose details make strongly model dependent some surface sensitive properties. This is called in this paper the pure surface effect. The other one is solely due to the very presence of the surface. It extends tens of angstroms into the metal and is referred here as a bulk effect, or more explicitly, as a surface-induced volume effect. In other words, the nature of the surface effects are understood further if one divides the surface in two regions, a region I with an oscillating electron density and a region II with a uniform density profile. Spatial variations of the field in region II give rise to additional absorption in the bulk. Since in a quantum mechanical model it is only the amplitude of the wave functions that can be added, both effects must be expected to interfere, although there are frequency regions and momentum transfer ranges where the wavefunction overlap may be neglected.

e-h pair excitation in the bulk of a metal, made possible due to the breaking of the translational symmetry, has been afforded in two recent studies. Whereas one is addressed to the theory of photoemission /20/, the other qualifies surface and bulk effects in the outcome of the electron energy-loss (EEL) experiment /36/.
by imposing the condition that the EM field vanishes along an infinite extension through the metal /20/, Meyers and Feuchtwang find that the photocurrent is increased up to almost a factor of two compared to the standard treatment. The latter is characterized by a constant field through space. As a conclusion, additional photocurrent comes out from the spatial variation of the electronic surface potential (dV/dz). The conclusion from their study is that photoemission cannot be seen as a pure surface effect. They show that a consistent model must account for the inherent spatial decay of the far field. It should be noticed that this contribution is different to the direct volume effect mentioned above.

In the other study, Persson et al. /36/ consider the excitation of e-h pairs due to the incident electrons for very low q and ω. They use a Golden-rule formulation for the excitation of e-h pairs. As a bulk contribution they analyze the contribution to the potential associated to the evanescent field that penetrates the metal. This excitation mechanism as well is found to contribute even more to the energy loss of the scattering electrons than the common Joule heating. Adding together the pure surface effect, the bulk contribution and the interference, remarkable agreement is found between theory and experiment /36/.

Being photoyield and EEL a consequence of the same basic mechanism, namely a surface induced e-h pair excitation, it is worthwhile to envisage this common feature within the same scheme. While EEL is due exclusively to a longitudinal field, the bulk photoyield comes out also from a transverse optical excitation. A touching point of both experiments may be found in the outstanding anomalous skin effect (ASE). Common studies of ASE are carried out looking upon scattering mechanisms of the conduction electrons taking place within the surface region /37/. As it was pointed out by RS, local models cannot give a proper description of ASE since the latter is a typical non-local effect. In their quantitative study, RS show that AA is essentially a low-temperature phenomenon, relatively largest in the infra-red.

RS worked out their calculations in the SCIB model that they introduced by the first time. Their expressions for the fields involve rather complicated integrals, mostly when the electrons are diffusely reflected at the surface. Subsequent work by Dingle /23/ simplify their expressions and make them applicable to diffuse electron reflection. He finds a strong dependence of the AA with the specular parameter. In a series of papers, Fuchs and co-workers /24,25/ consider also p-polarization, generalize it to the diffuse scattering theory and obtain self-consistent-field dielectric functions. All these works try to evaluate what we have called the surface induced bulk effect, as will be discussed further. One should notice on passing that their procedure requires a lengthy algebra. In some respects, introduction of diffuseness is equivalent to include in phenomenological models the non-local effect that arises in the density profile fluctuations. It should be noticed that also photoemission has been treated oftenly using SCIB /25,26/. Because of limitations in the model, those calculations cannot be very realistic in the whole frequency range. A microscopic generalization of SCIB connecting it to the infinite barrier model (IBH) has been proposed recently /38/. Computations that employ an IBH are still very heavy for our purposes. In fact, combined with the perturbative approach introduced by Bagchi et al. /39-41/ and improved recently by Mochán et al. /42,43/, a SCIB is a departure point to account for surface sensitive properties. It is one of the conclusions of the present work that this is appropriate to describe what we call the bulk contribution to the anomalous skin effect.
The formalism developed by Feibelman is general in principle, though he specializes to first order contributions in an expansion in terms of $q/\omega$, where $q$ is the perpendicular component of momentum in the material. This approximation amounts to include only the pure surface effect. It is not valid at frequencies well below the plasma frequency of which the ASE is a classical example. A more general procedure has been given by Mukhopadhyay and Lundqvist (HL) /44/, who do not use any expansion in $q/\omega$. They formally solve for the fields in three regions, namely bulk, surface and vacuum, and arrive at an integral equation for the fields in the surface region. Their formalism is difficult to apply directly.

Persson et al. /34/ develop a linear response function which can be easily applicable. There are however some limitations in their scheme. It has been seen that subsequent approximations in their work is equivalent to an approximation to SCIB /49/.

For our purposes we have found very convenient a formalism developed by Bagchi et al. /39/ and extended by Hochan et al. /42/. A perturbative approach allows one to use the surface impedance found in the unperturbed problem to evaluate it for a more realistic situation. This procedure can be combined with the general scheme proposed by HL. The latter has been used by Apell /45/ to obtain the reflection coefficient in terms of the surface length parameters (response functions), therefore including the pure surface effect in a convenient way, as was discussed in I. Some calculations employing this ideas are under current development.

3. LINEAR RESPONSE THEORY

a. Longitudinal Excitations

Let us consider a charge $Q$ placed at a distance $d$ outside a metal surface. The surface is supposed to be translationally invariant in the surface plane (Fig.1). This leads to conservation of parallel momentum and it affords to work with all quantities of interest Fourier transform according to:

\[ V(k_x, z, \omega) = \int d^2 r e^{i(wt-k_x \cdot \vec{r})} V(\vec{r}, z, t) \]  

(1)

$\vec{r}$ being a vector in the surface plane. Since time is uniform, one can also Fourier transform this variable and work in $\omega$-space. Since $k_x$ and $\omega$ are conserved quantities the problem is basically one-dimensional.

Outside the metal, $z<0$, Poisson's equation takes the form

\[ \frac{d^2}{dz^2} V(k_x, z, \omega) = 4\pi \rho_{\text{ext}}(k_x, z\omega) \]  

(2)

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where
\[ p_{\text{ext}}(k_z,z,\omega) = Q(\omega) \delta(z-d) \]  \hspace{1cm} \text{(3)}

is the Fourier transformed charge distribution. \( Q = Q(\omega) \) can describe any possible time-dependence of \( Q \).

In the region \(-d<z<0\), one solution to Eq. (2) is
\[ V(k_z,z,\omega) = V_0 [e^{-k_z z} - \rho(k_z,\omega)e^{k_z z}] \]  \hspace{1cm} \text{(4)}

with
\[ V_0 = \frac{2\pi Q(\omega)}{k_z} e^{-kd} \]  \hspace{1cm} \text{(5)}

the strength of the test-particle potential. The second term in Eq. (4) is a solution to the homogeneous equation in (2). This is of course the potential acting back from the metal when it responds to the charge \( Q \) setting up a screening charge \( \rho_{\text{ind}}(k_z,z,\omega) \) in the surface region. The as yet undetermined function \( \rho(k_z,\omega) \) in Eq. (4) plays the role of a reflection coefficient analogous to the case considered in I, where one shines light on a substrate. The screening charge \( \rho_{\text{ind}} \) either comes from a grounded metal or from the other side of a finite sample. One can therefore solve Poisson's equation finding the potential corresponding to \( \rho_{\text{ind}} \).

It coincides with Eq. (4), as it must, when \( z<0 \) since \( V \) vanishes outside the metal. One gets
\[ V(k_z,z,\omega) = \frac{2\pi}{k_z} \int_{-d}^{0} dz' e^{-k_z (z-z')} [p_{\text{ext}}(k_z,z',\omega) + \rho_{\text{ind}}(k_z,z',\omega)] \]  \hspace{1cm} \text{(6)}

which for \(-d<z<0\) takes the following form:
\[ V(k_z,z,\omega) = V_0 [e^{-k_z z} + \int_{-d}^{0} e^{-k_z z'} \rho_{\text{ind}}(k_z,z',\omega)/Qe^{-kd} e^{k_z z}] \]  \hspace{1cm} \text{(7)}

Comparing Eqs. (4) and (7) one can identify an explicit expression for the metal feed-back:
\[ \rho(k_z,\omega) = -\frac{2\pi}{k_z} \int_{-d}^{0} e^{-k_z z'} \rho_{\text{ind}}(k_z,z',\omega)/V_0 \]  \hspace{1cm} \text{(8)}

In other words, the reflection coefficient that yields the strength of the image potential felt by an electron outside a solid surface is a weighted average of the induced screening charge. The weight is related to the kernel in Eq. (6) which is the two-dimensional Fourier transform of the Coulomb interaction. In comparison with the invested amount of work, Eq. (8) is indeed a remarkable result. However to proceed further one needs to calculate \( \rho_{\text{ind}} \). Obviously the induced charge is set up in response to the external perturbation. One therefore requires a function, calculated from a specific model of the metal response relating \( \rho_{\text{ind}} \) to \( p_{\text{ext}} \), viz.:
\[ \rho_{\text{ind}}(k_z,z,\omega) = \int_{-d}^{0} dz'H(k_z,z,z',\omega) v_{\text{ext}}(z',\omega) \]  \hspace{1cm} \text{(9)}
H is called the density-density response function /29,46,47/. It is easy to see that the reflection coefficient can be written as:

$$\rho(\mathbf{\kappa}_s, \omega) = \int d\mathbf{z} e^{-\mathbf{k}_s \cdot \mathbf{z}} H(\mathbf{\kappa}_s, \mathbf{z}, \omega) e^{-\mathbf{k}_s \cdot \mathbf{z}'}.$$  \hspace{1cm} (10)

It should be noticed that \(\rho\), an average over the microscopic response function, is independent of the external potential.

A discussion of the transverse and longitudinal response for a semi-infinite homogeneous medium has been done in I. There we limited to the SCIS model. Employing it we found explicit expressions for the surface impedance in this approach. In order to do a more general discussion of these results, it is convenient at this point to outline some formulae from the theory of linear response /29,33,47/. An external potential \(V_{\text{ext}}(\mathbf{x}, t)\) produces an induced potential \(V_{\text{tot}}(\mathbf{x}, t)\) so that the total potential is

$$V_{\text{tot}}(\mathbf{x}, t) = V_{\text{ext}}(\mathbf{x}, t) + V_{\text{int}}(\mathbf{x}, t).$$  \hspace{1cm} (11)

Omitting for brevity the \(t\)-dependence, the dielectric response function \(\epsilon(\mathbf{x}, \mathbf{x}')\) is defined by

$$V_{\text{tot}}(\mathbf{x}) = \int \epsilon^{-1}(\mathbf{x}, \mathbf{x}') V_{\text{ext}}(\mathbf{x}') d^3\mathbf{x}'.$$  \hspace{1cm} (12)

or

$$V_{\text{ext}}(\mathbf{x}) = \int d^3\mathbf{x}' \epsilon(\mathbf{x}, \mathbf{x}') V_{\text{ext}}(\mathbf{x}').$$  \hspace{1cm} (13)

One can define a susceptibility function \(\chi\) such that

$$\epsilon_{\text{p, ind}}(\mathbf{x}) = \int d^3\mathbf{x}' \chi(\mathbf{x}, \mathbf{x}') V_{\text{ext}}(\mathbf{x}').$$  \hspace{1cm} (14)

This is an exact definition. From these equations it follows that

$$\epsilon^{-1}(\mathbf{x}, \mathbf{x}') = \delta(\mathbf{x} - \mathbf{x}') + \int d^3\mathbf{x}'' \frac{1}{|\mathbf{x} - \mathbf{x}''|} \chi(\mathbf{x}, \mathbf{x}'').$$  \hspace{1cm} (15)

Here the RPA in which the states \(|\psi_n\rangle\) are Slater determinants of single-particle states is employed. The medium is described as responding to the field \(V_{\text{tot}}\) with an approximate susceptibility \(\chi''\) defined by /46/

$$\epsilon_{\text{p, ind}}(\mathbf{x}) = \int d^3\mathbf{x}' \chi''(\mathbf{x}, \mathbf{x}') V_{\text{tot}}(\mathbf{x}')$$  \hspace{1cm} (16)

whence

$$\epsilon_{\text{RPA}}(\mathbf{x}, \mathbf{x}') = \delta(\mathbf{x} - \mathbf{x}') - \int d^3\mathbf{x}'' \frac{1}{|\mathbf{x} - \mathbf{x}''|} \chi''(\mathbf{x}'', \mathbf{x}').$$  \hspace{1cm} (17)

or, in Fourier transform,

$$\epsilon_{\text{RPA}}(\mathbf{k}, \omega) = 1 - \frac{4\pi}{k^2} \chi''(k, \omega).$$  \hspace{1cm} (18)

Following Hedin and Lundqvist /47/, a formula for \(\chi\) in terms of exact eigenstates of the interacting electron gas can be written as
\[ \chi(x, x'; \omega) = \frac{e^2}{\hbar} \sum_{n} \frac{\langle \phi_n | \delta p(x) | \delta p(x') \rangle | \phi_n \rangle}{\omega + \omega_n + i\eta} - \frac{\langle \phi_n | \delta p(x') \rangle | \phi_n \rangle \langle \phi_n | \delta p(x) \rangle | \phi_n \rangle}{\omega + \omega_n + i\eta} \]  

where \( \eta \) is an infinitesimal that tends to zero.

In RPA the excitation frequencies are simply

\[ \omega(p, k) = (\hbar/m)(k^2 + k \cdot p)/2. \]  

For fixed \( k \) the allowed momenta take quantized values \( p_n \), and the excitation frequency \( \omega_n \) of the general case (19) is just the value of (21) for \( p = p_n \).

An illustrative exam of the discrete spectra as done by Flores and Garcia-Molinier (FG) /33/ follows. Figure 2a shows the qualitative dependence of \( \epsilon_{\text{RPA}} (k, \omega) \) on \( \omega \) for fixed small \( k \). One finds zeros and poles whose meaning can be understood in the following way: let us define \( \delta V/\delta \psi \) as a response function. The normal modes of the system are then the poles of this function. In the RPA we have, in Fourier space:

\[ V_{\text{ind}} = \frac{4 \pi}{k^2} \psi_{\text{ind}} \psi_{\text{tot}}, \quad V_{\text{tot}} = (1 - 4 \pi \hat{k}^2)^{-1} V_{\text{ext}}, \]  

whence the response function in this approximation is

\[ (\delta V_{\text{ind}}/\delta V_{\text{ext}})_{\text{RPA}} = 1/\epsilon_{\text{RPA}} - 1 \]  

dressed modes

with \( \epsilon_{\text{RPA}} \) given by Eq. (18). In a lower approximation, \( V_{\text{ind}} \) is also obtained by using \( \chi \) but is assumed to be induced only by the bare external potential \( V_{\text{ext}} \). This is the Hartree-Fock approximation, in which the electron-electron interaction is neglected in calculating the induced potential. We then have

\[ V_{\text{ind}} = \frac{4 \pi}{k^2} V_{\text{ext}}. \]  

This amounts to

\[ V_{\text{tot}} = (1 + 4 \pi \hat{k}^2)^{-1} V_{\text{ext}}, \quad (1/\epsilon_{\text{HF}} = 1 + 4 \pi \hat{k}^2) \]  

and the response function is

\[ (\delta V_{\text{ind}}/\delta V_{\text{ext}})_{\text{HF}} = \epsilon_{\text{RPA}} - 1 \]  

bare modes

It is useful to compare Eqs. (22) and (25). The bare normal modes are the poles of \( \epsilon_{\text{RPA}} \). The dressed normal modes are the zeros of \( \epsilon_{\text{RPA}} \). These normal modes appear at the values of \( \omega \) indicated in Fig. 2a. We start out with the bare excitation frequencies \( \omega_n \), at which \( \epsilon_{\text{RPA}} (k, \omega) \) has a pole. When the electron-electron interaction is switched on, the normal-mode frequencies are shifted to the right, towards the renormalized values corresponding to the dressed modes. The total number of modes does not change. If there are \( N \) bare single-particle (or e-h) modes for \( \omega = \omega_n \), the electron-electron interaction changes this into \( N + 1 \) dressed e-h modes and one collective mode, the plasmon excitation at \( \omega = \omega_p (k) \). For small \( k \), this is the distant zero, far out to the right in Fig. 2a. The normal excitation of the system (both single-particle and collective modes) accounts for the contribution to the loss function \( \text{Im}(-1/\epsilon) \) already discussed in I.
The above analysis can serve as a guide-line when looking for some simplifications of $e_{RPA}$ which is sufficient for the survey intended here. We recall the well known sum rules /29/:

\[ \int_0^\infty \text{Im}(\varepsilon(k, \omega)) = (1/2) \omega_k^2 \]

and

\[ \int_0^\infty \text{Im}(1/\varepsilon(k, \omega)) = -(1/2) \omega_k^2 . \]

(26) (27)

Applied to $e_{RPA}$ and in the light of Fig.2a the meaning of Eq.(26) is that the total number of bare modes is $N$, whereas Eq.(27) means that the total number of dressed modes is $N$. Both are equivalent to the $f$-sum rule, which states that the sum of all the normalized oscillator strengths is equal to unity.

With the above prescription and the physical picture given by the preceding discussion one can construct simple dielectric functions that improve the classical scheme. The crudest simplification that includes a $k$-dependence is:

\[ \varepsilon(k, \omega) = 1 - \frac{\omega_k^2}{\omega_k^2 - \omega_k^2(k)} . \]

(28)

This is qualitatively shown in Fig.2b. All the spectra of a bare mode is condensed in just one bare $e$-$h$ mode $\omega = \omega_k(k)$. The renormalized spectrum consists of only one collective mode, the plasmon given by $\omega_k(k) = \omega_{ph} + \omega(k)$. Further approximations are involved in deciding what $\omega_k(k)$ is. To lowest order it is of the form $\omega_k(k) = \beta_k k$. If $\beta_k = \sqrt{3}/5V_F$, one has the hydrodynamic approximation, which results by assuming a small $k$ in (21) and replacing $k \cdot \vec{p}$ by $k \cdot \vec{p}$, where the average is taken over the Fermi distribution. A more accurate description of the $e$-$h$ pair structure has been proposed by FG and coworkers:

\[ \omega_k^2(k) = \beta_k^2 k^2 + (k^2/4m^2)k^4 \]

(29)

which yields the hydrodynamic approximation for low $k$ and renders a good approximation to (20) for high $k$. This is the so-called plasmon pole approximation with $\eta = 0$ /44,47/. It should be noticed that: a) $\omega$ must be understood as $\lim(\omega + i\epsilon)$, so that Eq.(28) satisfies the two sum rules irrespective of the form of $\omega_k(k)$, and b) Eq.(28) contains only one mode, so that there is no vestige of $e$-$h$ pair structure in the renormalized spectrum.

Single-particle modes play an important role in surface electrodynamics and this cannot come out of any simplification like Eq.(28). This suggest the next step /33/, shown in Fig.2c. One starts with two bare $e$-$h$ modes, $\omega_{ph}(k)$ and $\omega_k(k)$. It means that $\varepsilon(k, \omega)$ has two zeros and two poles and can be written in the form

\[ \varepsilon(k, \omega) = \frac{[\omega^2 - \omega_{ph}^2(k)][\omega^2 - \omega_k^2(k)]}{[\omega^2 - \omega_{ph}^2(k)][\omega^2 - \omega_k^2(k)]} . \]

(30)

Taking $\omega_k^2(k) = \omega_{ph}^2 + \beta_k^2 k^2$, gives the hydrodynamic plasmon form plus one dressed $e$-$h$ mode. From the form of Eq.(29), one gets the plasmon pole approximation for the collective mode and one dressed single-particle mode. Let us put

\[ \omega_{ph}^2(k) = \beta_{ph}^2 k^2 + (k^2/4m^2)k^4 . \]

(31)
...and single particle modes.

Figure 2
Qualitative behavior of RPA.
Bare (squares), collective (dot) and dressed (circles) frequencies.
a) Qualitative full RPA.
b) Hydrodynamic-like approx.
c) Approximation that contains e-h pair excitation.

Figure 3
Qualitative behavior of in the RPA approximation.
(Same nomenclature as in Fig. 2, including the polariton-like mode (dot).

Using all this in the sume rule (26) or (27), it yields

\[ b_L^2 + b_{eh}^2 = b_0^2 + b_1^2 \]  \hspace{1cm} (32)

This renders one condition for the four adjustable parameters. A second condition is obtained by imposing the requirement that the plasmon strength equals the correct value. Two further conditions are obtained by fitting to the RPA dispersion relations for the plasmon mode at low k and for the low-k edge of the e-h pair spectrum /29/.

b. Longitudinal and Transverse Bulk Excitations

For external radiation one needs to consider the full dielectric tensor

\[ \epsilon({\vec{k}},\omega) = \epsilon_L({\vec{k}},\omega) \mathbb{I} + \epsilon_T({\vec{k}},\omega) \mathbb{P} \]  \hspace{1cm} (33)

where \( \epsilon_L \) and \( \epsilon_T \) are the longitudinal and transverse dielectric functions and \( \mathbb{I} \) and \( \mathbb{P} \) are the longitudinal and transverse projectors

\[ [-\epsilon_L - k^2 q^2 \epsilon_T] \cdot \mathbb{E} = (4\pi/\omega) i \mathbb{J}_{ext} \]  \hspace{1cm} (34)

From Maxwell equations, the equation for \( \mathbb{E} \) in Fourier transforms is \( (q^2 \omega/c) \)
For the vector potential one obtains
\[ \hat{A}_{\text{tot}} = \frac{1}{\varepsilon_L} \left( \frac{k^2 - q^2}{k^2 - \varepsilon_L^2 \omega^2} \right) \hat{A}_{\text{ext}}. \]  
(35)
For the longitudinal part we recover the picture of longitudinal screening seen in the preceding sub-section, whereas if one takes the transverse part one obtains a description of transverse screening. The transverse normal modes have the dispersion relation
\[ k^2 - q^2 \varepsilon_T(\vec{k}, \omega) = 0. \]  
(36)
One associates this with the polariton mode. In the quasistatic limit, the polariton becomes the transverse dynamical mode given by the poles of \( \frac{1}{\varepsilon_T} \) /33/ for \( c = \varepsilon \) i.e.,
\[ \frac{1}{\varepsilon_T(\vec{k}, \omega)} = 0. \]  
(37)
Figure 3 shows the qualitative behavior of \( \varepsilon_T(\vec{k}, \omega) \). The picture is qualitatively the same as the previous one, except for the static limit (\( \omega \rightarrow 0 \)). However the physical picture is different. The renormalized frequencies \( \omega_{\nu} \) of the interacting electron gas coupled to the EM field appear now at the intersections of the curve \( \varepsilon = (\varepsilon_0, \omega)^2 \). As in the longitudinal case, for low \( k \) there is a distant mode at high \( \varepsilon \), and a quasi-continuum of modes accumulated in the low-frequency range. The distant mode is the (predominant) collective transverse excitation which results from the coupling to the EM field. This is the polariton or light polariton, which could also be viewed as a photon dressed by its interaction with a medium described by \( \varepsilon(\varepsilon_0, \omega) \). The lower-frequency modes have the nature of dressed transverse single-particle-like excitations. Their frequencies differ very little from the bare excitation frequencies \( \omega_{\nu} \). Indeed, the Coulomb electron-electron interaction is longitudinal and by itself would not produce transverse excitations. However, there is a small indirect interaction via the coupling to a transverse EM field, which produces a shift from \( \omega_{\nu} \) to \( \omega_{\nu} \). This change is very small because \( \omega_{\nu}/k = \varepsilon \ll c \), so that the coupling to the EM field is weak. In the limit \( c \rightarrow \infty \), the transverse eigenmodes are the poles of \( \varepsilon_T \), as is seen in the dispersion relation. The corresponding eigenfrequencies represent formally the quasistatic (\( c \rightarrow \infty \)) limit of the transverse elementary excitations. They are just bare frequencies, as they must, since on switching off the coupling to the EM field the electrons have no transverse interaction and their eigenfrequencies can only be those of the bare models. Incidentally for very high frequencies both \( \varepsilon_L^2 \) and \( \varepsilon_T \) behave as \( i \omega \rho/\mu^2 \) and then the polariton dispersion relation is
\[ \omega^2 = \omega_p^2 + c^2 k^2 \]  
(38)
which is essentially a long-wave approximation. For arbitrary \( k \) the polariton has a more complicated dispersion relation, reflecting the \( k \) dependence of \( \varepsilon_T \), i.e., spatial dispersion effects in the transverse response.

It should be noticed that in Fig. 3 there are \( N+1 \) modes when there are \( N \) in Fig. 2. Thus the lowest approximation needed to obtain a dispersive collective longitudinal mode (plasmon) requires one single-particle bare mode in \( \varepsilon_L \), whereas a strictly local \( \varepsilon_T \) is sufficient to yield the collective transverse dispersive mode (polariton) which results from dressing the photon. In the hydrodynamic model, one has
\[ \varepsilon_T(k, \omega) = 1 - \frac{\omega_d}{\omega} \left( 1 + \frac{\omega_d^3}{\omega - \beta^2 k^2 - (k^2/4a^2) k^2} \right) \]  

(39)

where the idea is to introduce one single-particle mode in \( \varepsilon_T \), consistently with the approximation (28) for \( \varepsilon_T \), and to provide for the act that in the quantum mechanical RPA result the \( k \to 0 \) limit of \( \varepsilon_T \) is \( 1 - \omega_d^2/\omega^2 \). One could therefore use a simpler form, namely

\[ \varepsilon_T = 1 - \frac{\omega_d^3}{\omega - \beta^2 k^2} \]  

(40)

which has classical behavior.

One possible way to work out a phenomenological analysis of the response to EM fields is to formulate everything in terms of the surface impedance. Therefore we try to extend the definition given in I. The surface impedance tensor is defined by

\[ \bar{E}_s(0) = (c/2x)Z \cdot \hat{n} \cdot \bar{H}(0). \]  

(41)

If \( Z \), denoted the surface impedance of the medium contained in the respective domain \( x \), one gets for the P-mode

\[ Z_p = \frac{4}{c} (E_x/H_y)_0 \]  

(42)

and for the S-mode

\[ Z_s = \frac{4}{c} (E_y/H_x)_0 \]  

(43)

As was seen in I, if there is a medium 1 on the side \( x < 0 \) and a medium 2 on the other side, the reflection coefficients in both geometries are given by:

\[ p_p = \frac{Z^<}{Z^>} \hspace{1cm} p_s = \frac{Z^<}{Z^>} \]

(44)

The poles of the reflectivity \( R = |R| \) are the surface modes of the system.

In our case, medium 1 is vacuum and medium 2 is the metal under study. If light is incident at an angle \( \theta \) with the normal to the surface, one has

\[ R_p = \frac{Z^< + \cos \theta l}{Z^< - \cos \theta l} \]  

(45)

Therefore the problem is reduced to evaluate the surface impedance of the medium. One needs to know the surface response functions, which in turn depend on the surface model. The standard EM matching conditions do not suffice in general to solve the problem, and this is expressed sometimes by saying that one needs additional boundary conditions (ABC). A proper description of the surface region will not need any ABC. Some calculations employing this scheme are presented in the following section.
4. ABSORPTION AND REFLECTION

a. Classical Joule Heating and Surface Contribution

It has been shown in I that the dielectric properties of the interface change in a rapid but continuous way across the surface region. Let us calculate now the power absorption of light, which will give also an alternative derivation of the reflectivity $p$ discussed in the previous section and introduced in paper I.

To calculate the absorption of light, one can use an equivalence between the Golden Rule and the Joule heating /47/. The absorption $a(\omega)$ is given by the time averaged power absorption in the metal normalized to the incident flux $I_s = (c/8\pi)A\cos\theta$, where $A$ is the irradiated area and $\theta$ is the angle of incidence from vacuum ($\varepsilon = 1$):

$$ a(\omega) = \frac{1}{2\hbar} \text{Re} \int \mathbf{J} \cdot \mathbf{E}^* - \frac{\omega}{8\pi\hbar} \text{Im} \int \mathbf{J} \cdot \mathbf{D}^* \cdot \mathbf{E} $$

(46)

where Re and Im denote the real and imaginary parts respectively. It is convenient to split $a(\omega)$ into two parts $a = a_1 + a_2$ in the following way:

$$ a_1(\omega) = -\frac{\omega}{8\pi\hbar} \text{Im} \int \mathbf{J} \cdot (\mathbf{D}_+^* \cdot \mathbf{E}_+ + \mathbf{D}_-^* \cdot \mathbf{E}_-^*) $$

(47)

and

$$ a_2(\omega) = -\frac{\omega}{8\pi\hbar} \text{Im} \int \mathbf{J} \cdot \mathbf{D}_-^* (\mathbf{E}_- - \mathbf{E}_-^*) $$

(48)

Here the perpendicular field component $E$ for a sharp interface, with a step in the field at $z = 0$ connected to a singular classical surface charge, has been added and substracted.

It should be noticed that $a_1(\omega)$ is a bulk-like contribution whereas $a_2(\omega)$ is, by construction, a surface property. It depends on the difference between the actual field perpendicular to the surface and the field for a sharp interface so that $a_1 = 0$ for a sharp surface.

Let us first evaluate $a_1(\omega)$. For frequencies lower than the bulk-polariton propagation frequency ($p_e < 0$: cf. Sec.2 in I), the $z$-integral converges automatically. One obtains readily:

$$ a_1(\omega) = -\frac{1}{2\cos\theta} \text{Im} \varepsilon \left[ E^p \right]_{2\text{Im}(p_e)}^{-1} $$

(49)

We have used $\mathbf{D} = \varepsilon \mathbf{E}$ for the slowly varying fields involved. When the bulk polariton is not evanescent any longer, a finite Drude damping will still provide a finite imaginary part for $p$ and Eq.(49) still holds. Im$(p_e)$ is directly related to the skin depth (cf. following sub-section). We assume that $\mathbf{E}$, $\mathbf{P}$ and $\mathbf{D}$ have the same form as in classical theory. The usual matching boundary conditions ($\mathbf{E}$ and $\mathbf{P}$ continuous) give at once:

$$ a_1(\omega) = \cos\theta \left| -p_e \varepsilon_k (1+\kappa/|p_e|^2) \int_{2\text{Im}(p_e)}^{-1} \right| $$

(50)

$\varepsilon_k$ being Im$(\varepsilon)$. A careful examination of the wave vectors involved gives the equivalent, but more transparent, result:

$$ a_1(\omega) = \cos\theta \left| -p_e \text{Re}(1/Z_e^*) \right| $$

(51)
and single particle modes.

As usual, q is the wave number of the incident wave and p is the z-component for the transmitted wave. e = ε(ω) is the classical dielectric function. Z is the surface impedance which is related to the reflectivity through Eq. (5.4) in I, and Z\textsuperscript{cl} is the classical surface impedance p_τ/q_ε.

For a_2(ω) one obtains after integrating by parts:

$$a_2(ω) = \frac{q}{\cosθ} \text{Im} \int dz \frac{d}{dz} D_\parallel (E_A - E_{A\perp}) \text{.} \quad (52)$$

To lowest order in k times the extension of the region where E_\parallel and E_{A\perp} differ D_\parallel is a constant (from ν.D=0) and dE_\perp/dz = 4ρ_{ind} (from ν.e = 4ρ_{ind}) and thus, in this "dipole approximation" (k-1/incident wavelength),

$$a_2(ω) = \frac{q}{\cosθ} |D_\parallel|^2 \text{Im} \int dz \frac{d}{dz} D_{\parallel\perp} / D_\parallel \text{.} \quad (53)$$

Now 4ρ_{ind} = E_{\parallel}(\infty) - E_{\parallel}(\infty) = D_\parallel(1/e - 1)). One can thus eliminate D_\parallel from Eq. (53), giving us the final result:

$$a_2(ω) = \cosθ |1 - p|^2 \text{Im} \frac{q \sinθ}{\sinθ} - \frac{ε}{ε_0} |Z|^2 \quad (54)$$

where the sin^2θ factor comes from |D_\parallel|^2 = sin^2θ|D_\parallel| and d_\parallel has been defined in I.

One can also express the power absorption a(ω) directly in terms of the surface impedance Z since

$$a(ω) = 1 - |p|^2 = \cosθ |1 - p|^2 \text{Re} \frac{1}{Z^{-1}} \text{.} \quad (55)$$

Comparing this formula with Eqs. (51) and (54), one finds a surface contribution to Z which is proportional to d_\perp(ω):

$$Z = Z^{cl} + \frac{1}{\sinθ} \frac{ε}{ε_0} d_\perp(ω) \text{.} \quad (56)$$

The classical surface impedance Z\textsuperscript{cl} = p_τ/q_ε is the result for a sharp interface, where by definition d_\perp(ω) vanishes. The factor sin^2θ is essential and shows how important the perpendicular field is to get a surface contribution.

It has been shown that the optical properties of the vacuum-metal interface are completely determined by the formulas for p_τ and Z and that the surface properties are determined by the function d_\perp(ω). The arguments are elementary: one calculates the Joule heating using the asymptotic form of the fields outside and inside the medium. No ABC's have been used. The results follow from the continuity of the tangential components of the electric and magnetic fields. The result for the surface impedance is exact to first order in the ratio between the width of the surface region to the wavelength of the incident radiation.

In the following these optical properties are investigated in a rather different way by using the results from Sec. 5 of I and the response functions discussed in Sec. 3b.

b. Anomalous Bulk Absorption

The bulk optical absorption due to e-h pairs excitations using the SCIB model is described in this subsection.
As it was discussed in Sec.2, the pure surface effect arises in the strong variations of the fields within the very surface region of the solid. The intraband volume term, on the other hand is usually neglected. Its contribution to absorption originates from the excitation of e-h pairs in the interior of the metal. An illustrative derivation of this contribution in FE metals perturbed by photons of small wave number is presented here. It is an extension of a previous report /49/. Excitation by low energy electrons have been considered in recent works /34,36/.

For a plane EM wave incident at an angle \( \theta \) with the surface normal, the surface impedance \( Z \) in the SCIB model is given by Eq.(5.23) in I. In principle one can take for the calculations the bulk dielectric functions of Lindhard /46/. Adding and subtracting to Eq.(5.23) the quantity \( \sin^2 \theta / \epsilon(\omega) \), where \( \epsilon(\omega) \) is a local dielectric function, the Drude dielectric constant for this case, \( Z \) can be divided into its transverse (\( Z_T \)) and longitudinal (\( Z_L \)) parts, namely:

\[
Z_T = i q \int dp \left[ \frac{\sin^2 \theta}{\epsilon_{\parallel}(q,\omega)} - \frac{p^2}{q^2 - q^2 \epsilon_T(q,\omega)} \right] / q^4 
\]

(\( q^2 = p^2 + k^2 \)) and

\[
Z_L = i q \int dp \left[ \frac{1}{\epsilon_L(q,\omega)} - \frac{1}{\epsilon(\omega)} \right] 
\]

It should be noticed that the \( q = 0 \) pole is cancelled for both \( Z_T \) and \( Z_L \) due to the properties of the Lindhard dielectric functions \( \epsilon_T \) and \( \epsilon_L \) when \( q^2 = 0 \) (cf. Sec.5 in I and Sec.2 in this paper).

The longitudinal contribution is seen to be zero for a transverse EM field (s-polarized light). On the contrary, it becomes eventually the main contribution as the parallel component of momentum of the incident field increases. For the simple illustration of interest here, only normal incidence is considered.

In terms of the surface impedance, the absorption is given by

\[
\alpha(\omega) = \frac{4 \cos \theta \text{Re}(Z)}{|\cos \theta + Z|^2} 
\]

According to Eqs.(57) and (59), the basic ingredient for the optical absorption is the real part of the surface impedance which under the assumption of normal incidence takes the form:

\[
\text{Re}(Z_T) = \frac{\text{Im} \epsilon_T(q,\omega)}{1 - q^2 \epsilon_T^2(q,\omega)} \left( q^2 - q^2 \text{Re} \epsilon_T^2 \right) + |q^2 \text{Im} \epsilon_T|^2 
\]

It is convenient at this point to establish a parallelism between the loss function given by \( \text{Im}(-1/\epsilon_L) \) in Eq.(5.37) in I and the actual contribution coming from \( \text{Im} \epsilon_T \) in Eq.(60). It must be pointed out that we are not looking here for the surface plasmon polaron given by the pole of Eq.(59). This will be considered in the following section, where it will be seen essentially as a classical (local) contribution. We are interested indeed in low frequency excitations, well below \( \omega_p \).
To further facilitate a division into various optical regimes, let us proceed to make some approximations based on the very small value of q compared to q's characterizing e-h pair excitations. For that purpose one can replace \( \text{Re}(\varepsilon) \) by \( \varepsilon(\omega) \) given by the Drude dielectric function in the limit \( \omega' \to \infty \). It is convenient at this point to use the length scale parameters introduced in section 3 in I. It will be seen that the relationship between these lengths characterize very well the different regions discussed in that section.

### b.1. Classical Absorption

It is easily seen that \( \varepsilon_0 \) defined in Eq. (3.2) in I is the inverse of the imaginary part of the wave number associated with the penetrating EM field in the limit \( \omega' \to \omega_p \) for normal incidence. For that purpose one replaces \( \varepsilon \) by a local dielectric function, e.g., the Drude form, and performs the \( \rho \) integration. One obtains readily the very-well known result for the classical surface impedance, namely:

\[
Z^c = p_t / q_e 
\]

where \( p_t \) is the z-component of the wavenumber of the penetrating field:

\[
p_t = \sqrt{\varepsilon_0} n \omega / \omega' \tag{62}\n\]

Explicit use of the Drude dielectric function shows clearly that \( \varepsilon_0 \omega \to \omega_p \) by Eq. (3.2) in I is nothing but \( (\text{Im}(p_t))^{-1} \) in the limit \( \omega' < \omega_p \) and \( \theta = 0 \). Let us rewrite the local dielectric function in terms of the length parameters \( \ell \) and \( \lambda_e \) defined by Eqs. (3.1) and (3.3) in I:

\[
\varepsilon(\omega) = 1 - \left( \omega_p^2 / \omega^2 \right) \frac{s}{s+i} \tag{63}\n\]

where

\[
s = \ell / \lambda_e = \omega / \tau \n\]

Using the definition of the electronic length, Eq. (3.3), in (62) yields for the real part of the surface impedance the following result:

\[
\text{Re}(Z^c) = \left( 1/2 \right) \left[ \left( 2s \right)^{1/2} \right] \left( \frac{s}{s+i} \right) \left( \frac{2s}{s+i} \right)^{-1/2} \tag{64}\n\]

Before going further, it is instructive to consider different limits of the classical skin effect. With \( \lambda < \ell \), at very low frequencies, one is in the classical skin region (CSR) characterized by \( \omega \tau < 1 \). One can write for this region

\[
\text{Re}(Z^c) = \left( 1/2 \right) \left( \frac{2s}{s+i} \right)^{1/2} \tag{65}\n\]

In this region the electrons experience many collisions during each oscillation of the field, and the screening mechanism is quite efficient. At rather high frequencies, \( \omega > \omega_p \), the damping is negligible to zeroth order \( (\ell - 0) \). The electrons respond as being free without affecting the field, and collisions are of no significance: this is the transmission region. A more interesting situation arises at the intermediate region called the relaxation region (RR), where absorption takes place due to bulk phonon excitation, scattering against impurities, etc. This region is accessible from our simple scheme above. One gets from Eq. (64):
Although a unique relaxation time cannot be assigned to the electrons, collisions are of great importance in the RR (cf. Fig. 2 in 1).

Scaled to \( \tau = \tau_0 / \xi_e \) (= \((V_F/c)(\omega_p/\omega)\)), Eq. (64) takes the form:

\[
\text{Re}(Z^{\text{cl}}) = \left(1/2\omega_p \tau\right)^{-1/2} \left(\frac{1}{1+\left(\omega/c^2\right)^2}\right)^{1/2}
\]

where \( \alpha = (1/\omega_p \tau)(c/V_F) \). The factor \( \omega_p \tau \), usually between 10 and 1000, contains the properties of the material. It depends on the density parameter, sample purity and preparation, temperature, etc.

Obviously the discussion above does not contain any information about the e-h pair mechanism. This can only be achieved with a non-local expression for the dielectric function including spatial dispersion.

b.2. Electron-hole Pair Excitations.

An estimate of the e-h pair contribution is obtained from Eq. (40):

\[
\text{Im}(\varepsilon) = \frac{\omega_p^2}{2\omega B} \left[ \left( \omega_B - \omega_i \right) + \delta \omega / \varepsilon \right]
\]

This response function has been found to represent most of the physics contained in the full Lindhard expression. It is seen to include e-h pairs (\( \varepsilon \tau = \omega_p \)) for small \( \omega \) and \( \omega_i \). Within this region, \( \text{Im}(\varepsilon) \) in the denominator of Eq. (60) can be neglected.

Its pole form

\[
\text{Re}(Z^{\text{eh}}) = \left(1/\omega_p \tau\right) \beta \left(\frac{\omega_p}{c}\right)^2 \left[1 + \left(\frac{\beta r}{\xi_e}\right)^2\right]^{-1/2}
\]

It is instructive to consider the two limits of this expression:

\[
\text{Re}(Z^{\text{eh}}) = \begin{cases} 
\beta \left(\frac{\omega_p}{c}\right)^2, & \beta r \ll 1 \\
\beta \left(\frac{\omega_p}{c}\right)^{-3}, & \beta r \gg 1
\end{cases}
\]

It must be kept in mind that these limits are always within low-frequency ranges.

The important result from this subsection is the ability to access the AA region using our simple approach. A cooled sample can fulfill the requirement \( \beta \gg \xi_e \).

Thus the field amplitude goes to zero in less than one mean free path. At very low frequencies, \( \omega_p \gg \omega_i \) and the difference in phase between the field and the induced current is small. This is called the anomalous skin effect (ASE). The absorption decreases by lowering the frequency. Increasing the frequency, one reaches an ultimate complex situation where the field has vanished and oscillated many times within each mean free path (\( \beta \gg \xi_e \gg \xi_e \)), denoted the extreme anomalous skin effect (EASE). Now the absorption decreases by increasing the frequency. Evidently, the
bulk e-h pair absorption mechanism dominates the picture in the anomalous region where $\epsilon \gg \epsilon_0$, and a dielectric response including e-h pairs is of crucial importance.

Figures 4 and 5 compare the classical absorption (Eq. (67)) and the e-h pair excitations (Eq. (70)) described within the simple approach followed above. It is instructive to present the results in terms of $r = \lambda_e / \lambda = 1 / \omega_c$. Figure 4 was obtained with $\lambda_e / \lambda \ll 1$, where a classical description is expected to give reliable results. Figure 5 shows the results when e-h pairs play an important role ($\lambda_e / \lambda > 1$). The different regions of the skin effect are explicitly indicated in both figures. The relationship between the quantum effect (e-h pairs) and the classical contribution can be written as

![Figure 4](image)

**Figure 4.**
Schematic picture of the electron-hole pair contribution to absorption for normal incidence and the classical one when the mean free path is small compared to the penetration length $\lambda_e$. CSE stands for the classical skin region and RR indicates the relaxation region. Re(Z) is measured in units of $A^2/\omega$.

![Figure 5](image)

**Figure 5.**
Same as in Figure 4 for $\lambda_e / \lambda \gg 1$, ASE stands for the anomalous skin region and EASE is the extreme ASE. The change in scale should be noticed.

![Figure 6](image)

**Figure 6.**
Results from a numerical calculation using the full Lindhard dielectric function (solid lines) and our approximate approach given by Eq. (78). The results have been normalized to the maximum value obtained using the full dielectric function. The scale has been chosen as 3.77 to get the maxima at the same place.
where \( f(1/\omega^2) \) goes to zero both at high and extremely low frequencies. The most interesting feature in this relationship is the simple \( k_0 \) dependence (neglecting frequency variations). This is gratifying since \( k_0 \) characterizes \( Z_e^i \) and \( k_0 \) is associated with \( Z_e^h \). It becomes obvious from this simple analysis that when \( k_0 \) is an order of magnitude smaller than \( Z_e^h \) the e-h pair mechanism is the dominant one, as it is meant by a smaller \( Z_e^i \) associated penetration depth \( l_0 \). At very low frequencies and at high energies, our results should not be taken too literally.

Figure 6 establishes a comparison between the results obtained by using our simple scheme and a numerical calculation performed using the Lindhard transverse dielectric function /46/. It is encouraging from these results that the main features of the absorption coefficient within a region of tens to hundreds of meV can be described with the simple approach proposed here.

5. SURFACE EM WAVES AND SURFACE PLASMONS

As is very well known, an ideal (local) metallic surface is perfectly reflecting for \( \omega \equiv \omega_c / \cos \theta \), where \( \theta \) is the incident angle of the EM wave. This follows at once from the classical Fresnel formulas. It is also perfectly reflecting for frequencies below \( \omega_c \) since \( \omega < 0 \), and no EM fields can penetrate the metal due to the screening effect at the surface. For a smooth surface there is indeed a penetration depth and it extends up to several \( A \), as self-consistent calculations reveal. At \( \omega = \omega_c \) there is a longitudinal excitation available. Non-local effects introduce a dispersion relation which in the hydrodynamical approach is (cf. Sec.3)

\[
\omega^2 = \omega_p^2 + (3/5)q^2 \omega_F^2
\]

(73)

The plasma mode is limited to p-polarized light. A thin metallic film and a metal superlattice amplifies the effect /31,32/. This will be discussed in paper IV of this series.

Analogous to the dispersion relation (73) for the longitudinal mode, one can derive a dispersion equation for transverse waves. This is the already discussed polariton mode (see I). In a local non-absorbing medium it is

\[
\omega^2 = \omega_p^2 + \epsilon q^2
\]

(74)

In this section surface effects and surface modes are examined. We start with the surface-plasmon polariton. Figure 7 illustrates this mode between two semi-infinite isotropic media. The shape of the surface waves are of the type

\[
a_j e^{i(k_j \cdot \mathbf{r} - \omega t)} e^{-k_z z}, \quad z > 0.
\]

(75)

Here \( \beta_j \) and \( \beta_z \) are determined by Maxwell equations. The result is

\[
\beta_j^2 = (k_j^2 - \epsilon_j q^2)
\]

(76)

Matching boundary conditions give, after some algebra, the (classical) dispersion relation
A non-radiative surface mode propagating along an interface without decaying in the propagation direction, although decaying in a direction perpendicular to the propagation, is commonly referred to as a Fano mode. This is the solution for \( \varepsilon_2 < 0 \), which can occur for, e.g., a metal in part or all of the optical frequency range. The other solution with \( \varepsilon_2 > 0 \) (the Brewster radiative mode) is actually represented by the same mathematical equation. This mode is only bound to the surface if \( \varepsilon_2(\omega) \) has a small imaginary part /50/.

\[
\begin{align*}
  k_z^2 &= \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} > 0
\end{align*}
\]

Figure 7. A surface wave (LH mode) propagating at the boundary between a semi-infinite dielectric (or vacuum) and an electron plasma.

Figure 8. Dispersion of surface modes (solid lines) and bulk polariton (dashed line) for a non-dispersive free-electron metal. The asymptotic behavior is also indicated.

If the metal dielectric function is taken as given by Drude, i.e., \( \varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2} \), the explicit form for the dispersion equation has solutions /51/

\[
\omega^2(k_x) = \frac{\omega_p^2}{2} + \frac{\omega_p^2}{2} \left( \frac{\epsilon_2}{\epsilon_1} \right) \left( \frac{(ck_x)^2}{\epsilon_1} \right)^{1/4}
\]

A small \( k_x \) expansion gives

\[
\omega^2(k_x) = \begin{cases} (ck_x)^2 / \epsilon_1 & \text{(Fano mode)} \\ \omega_p^2 + (ck_x)^2 / \epsilon_1 & \text{(Brewster mode)} \end{cases}
\]

for the Fano and Brewster mode respectively. The general behavior is represented in Fig.8. The Fano mode is the only one of interest to the present discussion. It will be assumed that medium 1 is vacuum (\( \varepsilon_1 = 1 \)).
Within a classical analysis, there is still another way to look upon the surface \( \varepsilon \) mode. The dispersion relation can be obtained from the pole of the reflection coefficient. Having dealt with a generalization of the classical Fresnel expression in \( I \), it is worthwhile to try to extract more information from its non-local equivalent (Eq. (5.40) in \( I \)). Starting from this formula and using Eq. (56) for the surface impedance, one can write

\[
P_p = \frac{ep - p_t - ik_s(e-1)d}{ep + p_t + ik_s(e-1)d} \quad (79)
\]

The pole of the reflection coefficient gives the dispersion \( \omega = \omega(k) \) of the surface mode. Neglecting the surface contribution, this pole can be found from \( \cos \phi + Z = 0 \). One recovers Eq. (77) in a general case. For a vacuum - metal interface, this can be rewritten in the more familiar form

\[
(k_s/q)^2 = \varepsilon / (\varepsilon + 1)
\]

which gives the dispersion relation for the surface polariton. The corresponding relation for bulk polaritons at very low \( k \) is (cf. Sec. 5 in \( I \)):

\[
(k/q)^2 = \varepsilon \quad (81)
\]

For the surface EM mode, one has that the wave length is long and of the order \( k \); \( \omega / c \). For such long wavelengths, one has no effects of spatial dispersion and one can take the classical dielectric function \( \varepsilon(\omega) = 1 - \omega_p^2 / \omega^2 \) in (80). Solving this relationship for \( \omega(k) \) with \( \varepsilon = 1 \) and discarding the other solution which is now unphysical, one retrieves the dispersion relation for surface polaritons obtained above in Eq. (78):

\[
\omega^2(k_s) = \frac{1}{2p} + (ck_s)^2 - \frac{1}{2} \omega_p^2 + 4(ck_s)^2 \quad (82)
\]

An inspection of Fig. 8 shows that the Fano mode of the surface polariton starts at \( \omega = 0 \) for \( k = 0 \). It follows closely the photon line but flattens out and tends to a constant value for \( k > \omega / c \). As a result, one cannot excite a surface EM wave with an external light since energy and momentum cannot be conserved simultaneously at a planar surface. For a rough surface, an external wave can couple to a surface EM wave.

Going the other way around, the bulk polariton (cf. Sec. 5 in \( I \)) starts out as a plasmon and at high \( k \) it approaches asymptotically the photon line \( \omega = ck \). (Note that this occurs in the limit \( \omega > \omega_p \), where \( \varepsilon(\omega) = 1 \).)

Several techniques are available in the experimental study of surface plasmon polaritons. The choice of one or another technique is ultimately determined by the frequency and wavenumber range in which the excitation is examined. One method to study surface polaritons is to place above the surface a diffraction grating that provides the momentum. At short wavelengths, the surface polariton is most plasmon-like, the most suitable probe of the dispersion equation is an experiment involving electrons (EEL). This is due to the fact that, for a given \( \omega \), a moving electron always provides a \( k \) in correspondence with the surface mode. Therefore this can be excited while the electron is reflected losing energy \( \hbar \omega \) and momentum \( \hbar k / \ell \). Conversely, at very small wave number values, where the surface polariton is most photon-like, a useful probe is an experiment involving light of the appropriate frequency. In this case it is very suitable to make use of a technique known as attenuated total reflection (ATR) /11/.

ATR involves the use of the evanescent wave that is set up at a medium-air interface when light in a high refractive index medium, such as glass, suffers...
total internal reflection. It is the reduction of the reflected wave due to absorption that is called ATR. If the weakening occurs by some other means it is usually called frustrated total internal reflection (FTIR). In the former case a prism is placed above the crystal surface and a light wave is shone in under the condition of total reflection in the face parallel to the crystal surface. There will be an evanescent wave in the air gap between the prism and the surface. This wave provides the momentum required to excite a surface polariton. This method is illustrated schematically in Fig. 9.

Going back to Eq. (79) and considering the non-retarded limit, one obtains the condition for the surface mode in the electrostatic limit by letting $c = \infty$ in that equation:

$$\varepsilon(\omega) + 1 = 0.$$  \hfill (83)

It is equivalent to evaluate the pole in (79) to zeroth order in $k_d$. Combining it with the classical dielectric function $\varepsilon = 1 - \omega_p^2 / \omega^2$, the well-known result for the surface plasmon is obtained:

$$\omega_s = \omega_p / \sqrt{2}.$$  \hfill (84)

In a more general case the non-dispersive surface plasmon frequency is given by

$$\varepsilon_s(\omega) + \varepsilon_s'(\omega) = 0.$$  \hfill (85)

Let us consider next the case often referred to as "large" $k$. By this it is meant that $k_s$ is much larger than that of the light wave of frequency $\omega$ so that retardation effects can be neglected, but also that the wavelength is long compared with the lattice spacing or the Fermi wavelength. Therefore wave numbers in the interval

$$\omega/c \ll k_s \ll k_F$$

are considered here. To first order in $k$ one obtains

$$\omega^2(k_s) = \omega_s^2(1 + k_s d(\omega))$$  \hfill (86)

This result was first obtained by Harris and Griffin /52/ using RPA and soon thereafter derived in a model independent way using EM theory by Flores and Garcia-Wolinear /53/. A calculation of $\omega(k)$ by Inglesfield and Winkborg using the QIB model /54/ gives the dispersion relation over a wide range of momenta up to the plasmon cut-off momentum $\omega_c$.

It is quite interesting that one is able to derive the dispersion relation for the mode which exists at a vacuum-metal interface without in any way specifying the underlying model for the dielectric response. Moreover, Eq. (86) shows that the long wavelength limit is a purely classical result. Quantum mechanics enters at the first correction in the form of the center of gravity of the induced charge.

If the induced charge at $\omega_s$ is relaxed outside of the jellium edge or in the other direction, it will appear in an experiment as a shift from the classical value for this collective surface mode. Since $d$ contains an absorptive part, Eq. (80) also tells us about the damping of the mode in question, through its decay into e-h pair excitations.
Figure 9. Attenuated total reflection (ATR) configuration to excite surface plasmon polaritons using a hemicylindrical prism and an air gap.

Figure 10. Illustration of a more realistic (non-local) behavior of the surface mode. For k < q the pressure in the electron gas tries to counteract the electron density variation.

For higher k, it is seen in Fig. 10 that the dispersion relation bends upward. This has to do with the fact that when one tries to build up an electron density variation for the surface plasmon with wave numbers k < k_F, the pressure in the electron gas will counteract this and the mode will be stiffen, thereby increasing its frequency. In between, the small k transverse plasmon and the high longitudinal plasmon, one has a regime where the dispersion relation contains important information about the surface charge spatial distribution. The calculation of d_\perp within RPA seems to agree rather well with experimental findings, whereas calculations not based on a self-consistent density profile disagree strongly, even giving the wrong sign for d_\perp (cf. Table II). This discrepancy occurs because the electron density profile in most models is not enough relaxed, forcing the induced density to be too far into the metal. The d_\perp in Table I and Eq. (79) refers to the jellium edge. In a more general theory d_\perp has to be replaced by d_\perp = d_\perp, which is invariant under translations /3/.

Some comments are at hand when one takes a closer look at Table I. The first three models have an infinite barrier at the surface and therefore, by construction, one can never get a negative value respect to the positive background and gives therefore a smaller value. The hydrodynamic model only contains plasmons and no e-h pairs, therefore it does not give any damping for the model. A reduction in the barrier, as in the infinite step model, increases the damping considerably but the electron density is still not relaxed enough: the induced density sits inside the positive background. It is not until one goes to the self-consistent scheme based on the density functional scheme that the induced density is outside of the positive background, since in a density functional both, the induced density and the electron potential, are allowed to change in response to each other. In this case, the damping is also considerably larger, being almost an order of magnitude larger than in the other models. This will have some important consequences for the damping of excited molecules located above a metal surface. Finally it should be pointed out that the experiments are very hard to perform, as it is indicated by the spread in the experimental data.
...and single particle modes.

The surface plasmons correspond to an electron density oscillation in the surface. The free oscillations have the same characteristics as the induced oscillations discussed in I. It has a pronounced maximum not far from the distance $d$ from the jellium edge and shows an oscillatory decay into the bulk. One can notice that even for small $k^*$ ($k^* < k_F$) there is a finite damping of the surface plasmon due to the strong coupling to real excitations of e-h pairs.

**TABLE I**

<table>
<thead>
<tr>
<th>MODEL</th>
<th>REF.</th>
<th>$Re\left(\omega_0\right)$ (A)</th>
<th>$Im\left(\omega_0\right)$ (A)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrodynamic</td>
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<td>0.90</td>
<td>0.0</td>
</tr>
<tr>
<td>Finite step</td>
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<td>0.32</td>
<td>-0.18</td>
</tr>
<tr>
<td>Q I B</td>
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<td>0.26</td>
<td>-0.05</td>
</tr>
<tr>
<td>S C I B</td>
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<td>0.98</td>
<td>-0.06</td>
</tr>
<tr>
<td>Long-Kohn</td>
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<td>-0.42</td>
<td>-1.14</td>
</tr>
<tr>
<td>Experimental</td>
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<td>-0.14</td>
<td>-0.14</td>
</tr>
<tr>
<td>Experimental</td>
<td>59</td>
<td>-0.36</td>
<td>-0.60</td>
</tr>
</tbody>
</table>

6. CONCLUSIONS

Electronic excitations and spectroscopic techniques in condensed matter and surface studies are different points of view to refer to the same subject. In both cases one is essentially looking for the response of the electron plasma. To understand the role of the normal-mode spectrum is of paramount importance in many studies of solids and surfaces. One of the most extended techniques to analyse interface processes is the measurement of quantities arising from collective excitations. These are usually analyzed employing classical schemes. This is often not enough in surface analysis as recent experiments have shown. Also the standard optical experiment contains non-local ingredients that cannot render fine information about surfaces unless a proper description of the physics involved is at hand. An adequate treatment of non-local (quantum) aspects would involve a self-consistent treatment of the dynamical surface screening. Though recent advances permit carrying out these studies, there are some disadvantages when proceeding with brute force calculations. In particular, the physics involved is usually hidden in heavy computations. Additional limitations of a self-consistent approach using a jellium model are found on trying to apply it to more realistic systems. Therefore simple models are of worth whenever they can provide an understanding of the main features of the processes taking place and as much as they can give a qualitative insight of the physics involved.

An overview of the role played by the different types of elementary excitations in surface electrodynamics has been the main subject of the paper. The response to an external longitudinal stimulus (particles) and to an external transverse stimulus (radiation) have been taking into account. The emphasis has been on the basic physics rather than on advanced calculations that may be needed for accurate results. The SCIB model does provide an adequate framework to these purposes. This model, that at least can give a qualitative description of the bulk contribution due to the surface presence, and bulk dielectric functions that contain the main aspects of non-locality for plasma resonances and e-h excitations at low frequencies, have been employed. In this way a discussion of reasonable approximations have been done and illustrated. It turns out that in some cases one can even obtain surprisingly good accuracy. Non-specular surface scattering,
where one expects the SCIB model to give accurate results, has not been worked out. It is of interest to extend the above analysis to this and similar situations, and this is the goal of a forthcoming paper. We have also restricted to semi-infinite metals, leaving the discussion of other geometries to papers III and IV of this series. Some other aspects related to the adsorbate-substrate complex, a broad topic of major interest, will be discussed in paper V.

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...and single particle modes.


15. See, e.g., E. Merzbacher, "Quantum Mechanics" (Wiley, New York, 1970), Ch. 18.


