

# Theoretical views of Linear and nonlinear response function in metal optics

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**Abstract:** As we know the light interacting with metal surface or interfaces can excite longitudinal plasma waves inside the metal. This phenomenon becomes more important at frequencies of the order of metal plasma frequencies ( $\hbar\omega=10\text{eV}$ ). This is the region in which the advent of synchrotron radiation has opened up a rapidly growing field of optical experiments. One can demonstrate that reflection from and transmission through metal layers, electro reflection and ellipsometry from metal surfaces, the surface Plasmon and Eigen modes in this layer and spheres and also the field near the surface as encountered by the photoemission. These things cannot be understood been qualitatively without taking plasma waves into account.

**Keywords:** Plasmon frequency, Hydrodynamic approximation, non local effect, Drude-type theory

**Introduction:** In the study of present thesis, we have shown that it is not necessary to include plasma waves in the calculation of the optical response of a variety of systems containing metal surfaces and layers, but it is also possible to do so within a rather simple straight forward extension of classical Fresnel optics, then so called Hydrodynamic approximation (HD). This HD has a clear physical concept and is easy to handle mathematically. In addition to the familiar transverse electromagnetic waves, it includes longitudinal waves which are also solution of Maxwell's equation if the wave number dependence of the dielectric function i.e., spatial dispersion is taken into account in Drude-type theory. From a theoretical point of view, the HD is the simplest approximate version of "non-local optics". To discuss its relation to other phenomenological as well as the more general microscopic approaches is the second major aim of this thesis. It turns out that for many purposes the HD provides a very useful interpretation scheme. Which allows a qualitative understanding of the underlying physics with one requiring a complicated theoretical and numerical machinery.

One therefore, wants to encourage experiments to use HD for the interpretation of experiments and to start with the discussion of slight extension of standard optics. One presents the method and demonstrates its successful applications in many circumstances. This scheme of calculating the optical response of systems with conduction electrons is conceptually no more difficult than calculation of reflectivity of glass plate. One can also study the several more sophisticated treatments of the response of single metal surface to electromagnetic radiation. The relation between general forms of the response functions and additional boundary conditions are discussed for the variety of phenomenological models, the HD, the specular reflection model, the dielectric approximation the semi-classical infinite barrier model and several others. Also microscopic surface response calculations based on a quantum mechanical model containing a surface potential for the electrons are presented and their relationship to the HD and the SCIB model are illustrated. One also deals with the two surface response functions  $d_{\perp}(\omega)$  and  $d_{\parallel}(\omega)$  which contain the integrated effects of spatial dispersion and surface properties on measurable quantities such as reflection coefficient or surface Plasmon dispersion. Their relation to measurements as well as to microscopic or phenomenological response calculation is discussed.

The optical surface effects occur in principle for all frequencies of the incident light and are closely related to the perturbation of the surface charge distribution. This charge distribution is always induced by polarized light (i.e. light with a finite normal component of the electric field vector) at the interface between media of different bulk dielectric constants. At a metal surface, these induced screening charges are confined to a narrow but finite surface region, typically a few Angstroms thick or less in principle, a modification of the charge distribution at the surface, for instance by and atoms or an applied static electric field, affects the distribution of optically induced charges and thus the surface electromagnetic field and the response properties. The optical techniques mentioned in the beginning and so sensitive, that already minute modification of the surface constitution, for instance by means less than a monolayer of ad-atoms, can be clearly detected. In many cases, for instance of metal electrodes in electrolytic cell, these optical techniques provide the most detailed information available. Hence one is not considering the inelastic light scatterings which occur at certain frequencies owing to transition between particular collection modes or single electron states of the metal or an adsorbate. The aim of optical instruments is to obtain information about the constitution of the surface e.g. the electronic charge distribution. Clearly, a reliable theoretical basis for the interpretation of such optical surface data is highly desirable. Unfortunately, classical Fresnel optics, which has been applied successfully to many problems of metal optics over many decades, is not sufficient for an adequate understanding of many experimental results on a variety of metal surfaces. The reason for the failure of classical optics in this context is easy to understand. The standard procedure of classical Fresnel optics<sup>1</sup> is based on the assumption that a boundary between media can be approximated by a plane between two

homogeneous regions of different dielectric properties and that the general solution of Maxwell's equations on both side of interface on transverse divergence free electromagnetic waves, which are matched by the standard boundary conditions. For p-polarization,  $E_{\parallel}$  and  $D_{\perp}$ , the tangential component of the electric field and the normal component of the displacement field must be continuous. Therefore, the discontinuity of  $E_{\perp}$  and thus the induced surface charge is proportional to the discontinuity of the inverse dielectric constant  $1/\epsilon(\omega)$  across the interface. In order to simulate within this approach the effect of slightly modified surface, for instance by ad-atoms or an applied electric field one has to assume a thin surface layer (thickness of the order  $1\text{Å}^0$ ) with a modified dielectric constant  $\epsilon_s(\omega)$ . Near the plasma frequency of this surface layer,  $1/\epsilon_p(\omega)$  becomes layer and the classical Fresnel theory predict a layer induced change and a layer optical response which has no correspondence in the experimental findings. In real systems there is no perfect screening and no singular surface charge density. The induced charges and spread over a distance of screening length, typically of the order  $1\text{Å}^0$  in metals, and induced charges in a thin surface layer are coupled those in the substrate. As a consequence, at the plasma frequency of the layer material no resonant plasma oscillations are found in a very thin ad layer.

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