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MEASUREMENTS OF SOME PHYSICAL PROPERTIES
OF SOLIDS

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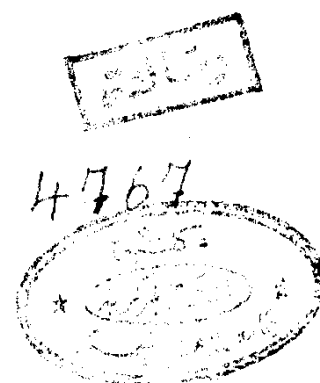
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ABSTRACT

In the present work , the electron theory of solids is studied experimentally ; therefore some physical properties of Gold are measured .

The optical constants - described by the refractive index n and the extinction coefficient k - for spectroscopically pure gold have been determined. Hence , the concentration of conduction electrons N , the D.C. conductivity σ_0 , the relaxation time τ , the plasma or critical wavelength λ_p , the energy corresponding to the interband transitions , the Fermi energy E_F and the Fermi velocity V_F were calculated.

Measurements were made at room temperature using three uniform thick films in the spectral range from 0.4μ to 2.3μ . The films were prepared by evaporating gold on a glass microscope slides under vacuum of about 10^{-5} mm Hg.

Beattie's method , which depends on the analysis of elliptically polarized radiation reflected from the metallic surface , has been used for the present measurements. The incident radiation was plane polarized at 45° to the plane of incident light. This was reflected by a mettalic film , at an angle of incidence 80° , into an analyzer. As the

analyzer was rotated , the reflected intensity varied sinusoidally. The phase and the amplitude of this variation gave the elements of the ellipse vibration. The results obtained from the ellipse vibration together with Crosby tables were used to calculate the optical constants.

The calculated values of the microscopic parameters of gold were found to be as follows :

- (i) $N = 4.7922 \times 10^{22}$ electron/cm³
- (ii) $\sigma_0 = 12.0493 \times 10^{16}$ e.s.u. for $\tau = 0.994 \times 10^{-14}$ sec.
- (iii) $\sigma_0 = 13.3464 \times 10^{16}$ e.s.u. for $\tau = 1.101 \times 10^{-14}$ sec.
- (iv) $E_F = 4.803$ ev.
- (v) $V_F = 1.304 \times 10^8$ cm/sec.

These values , calculated by using the optical constants measured in the present work , show agreement with those obtained by previous authors.

The threshold frequency of interband transitions for gold was found to lie in the visible range of spectrum at an energy $E_g = 2.36$ ev and this agrees well with the value obtained theoretically.

The critical wavelength was found to be $\lambda_F = 1527.4 \text{ \AA}$ corresponding to an energy $h\nu_p = 8.13$ ev .

Some of the electrical properties were also carried out ; using the same evaporated gold films. The concentration of free electrons N was determined from Hall effect measurements and the electrical D.C. conductivity σ_0 was also measured. The physical constants measured by the electrical and optical methods were compared.

INTRODUCTION

The study of the optical properties of absorbing materials specially those which are highly absorbing for electromagnetic radiation , like metals , is of great significance. Usually the optical properties of materials are described by the characteristic constants : the refractive index n and the extinction coefficient k .

These constants , which can be measured optically , give important information about the physical properties of the material qualitatively and quantitatively such as :

- (1) The concentration of conduction electrons in the solid.
- (2) The Fermi energy .
- (3) The Fermi velocity corresponding to the maximum filled energy level .
- (4) The energy corresponding to interband transitions .
- (5) The relaxation time .
- (6) The plasma frequency.

Measurements of the optical constants can be done by various methods. The most suitable method involves by measuring the properties of light transmitted or reflected from the surface of the metal under consideration.

Theoretical investigations of the optical properties of metals were been carried out^(1,2,3,4,5).

The oldest classical treatment which is now referred to as Drude's free electron theory,⁽¹⁾ was based on the assumption that a metal contains free electrons which experience viscous damping. This situation is described by the equation of motion of charge carrier in the field of incident radiation. In 1900 Drude⁽²⁾ proposed a formula for the optical properties of metals based on the postulated existence of two kinds of free charge carriers. In 1904 Drude⁽³⁾ abandoned this formulation since it seemed inconsistent with the electron theory which was being developed at that time. He then restricted the charge carriers to one kind. Since that time it has become evident that Drude's restricted formula may be used with only limited success and does not bring optical data into harmony with data for D.C. conductivity unless the optical properties of the interior are assumed to be different from those derived from reflection experiments on the surface.

It will be shown that Drude's more general formula does apply without limit to the free electron contribution to optical properties in all metals which have been studied, and that there need be no essential difference between the optical properties of the surface and those

of the interior to be consistent with the D.C. conductivity since quantum mechanics and the exclusion principle were then unknown, Drude could not have guessed that electrons in different Brillouin zones can behave differently. He have suspected the existence and important function of "holes". Nevertheless, the properties which he ascribed in his earlier paper to positive and negative "ions" show a remarkable resemblance to modern ideas about holes and electrons.

On the basis of the classical theory of free electrons of a metal Drude⁽³⁾ derived expressions for the dependence of the real and the imaginary parts of the dielectric constant on frequency, concentration free electrons and their relaxation time.

The advent of the Pauli exclusion principle, Fermi-Dirac statistics, the band theory of solids^(4a) and the concept of "effective mass" has modified the picture in some respect, but has not altered the predicted relations between the optical constants and the D.C. conductivity. In 1931 Krönig⁽⁵⁾ had shown that the Drude's theory is valid for free electron model obeying ^{quantum} mechanics with a re-interpretation of the parameters. The free electron formulae for the refractive index and the extinction coefficient should be valid for frequencies below the threshold

for transition between electronic bands. This threshold frequency occurs for most metals in the visible spectrum. These formulae have been modified by Dingle in 1955⁽⁶⁾ to allow for the anomalous skin effect and obtained a dispersion formula for the optical constants of metals in the infrared region of the spectrum. In 1948 theory of the anomalous skin effect was originally given by Reuter & Sondheimer⁽⁷⁾. The anomalous skin effect, for which the classical treatment is inadequate, exists whenever the mean free path of the electrons is not small in comparison with the wavelength and penetration distance. In this case, the current density at any point is determined not only by the electric field there, but also by the motion of electrons which arise there from other places at a distance less than or comparable with the mean free path.

The optical constants of metals depend greatly upon the exact physical state of the metal under consideration. Thus evaporated films do not in general have the same optical characteristics as polished metal specimens. Heavens⁽⁸⁾ has reviewed the work on the dependence of the measured optical constants of evaporated films upon the film thickness. The principle effect is due to the state of aggregation of the film. For thick films the optical constants generally approach those of bulk material.

In recent years numerous results of the experiments on the optical properties of noble metals have been published (9,10,11,12,13,14,15).

Dold & Mecke⁽⁹⁾ measured the optical constants of the metals ;Au,Ag & Cu, in the spectral range from 1.25μ to 14μ . The evaluation of the form of dispersion formula particularly in respect of the anomalous skin effect in the infrared region of the spectrum and the evaluation of the relaxation time dependence on frequency enables to determine several metal parameters ; such as the effective electron density , Fermi energy , Fermi velocity and the mean free path of the conduction electrons.

Roberts⁽¹⁰⁾ measured the optical properties of solid copper in a wavelength range from 0.365 to 2.5μ and at temperatures 90° , 300° , and 500°K . At longer wavelengths the optical properties are determined almost entirely by free electrons. Deviations from simple theory are partly explained by the anomalous skin effect and he concluded that the electronic collisions at the metal surface are diffuse. However , he showed that the anomalous skin effect is not sufficient to explain the observed deviations and that a more complete interpretation ought to consider the non-spherical nature of the Fermi surface and variation of the relaxation time over this surface.

Drude⁽¹¹⁾ showed that the surface optical properties of noble metals in the infrared region can be explained by a simple generalized form of the Drude's formula for the free electron theory, but involves two types of free electrons. He concluded that the optical constants in the interior are the same as those measured on the surface.

Schulz⁽¹²⁾ measured the optical constants of liquid metals and noble metals. Comparison was made with the Drude's theory, including the extension to deal with anomalous skin effect.

Hodgson⁽¹³⁾ measured the values of the refractive index and the extinction coefficient for evaporated films of Au, Ag & Cu at wavelengths ranging between 1.0μ and 15μ by a reflection method. Theoretical formulae for the infrared properties of metals were discussed and he showed that the experimental results were not sufficiently accurate to distinguish between the classical free electron theory and later modification proposed by Dingle⁽⁶⁾. His experimental results have been compared with the theory by a graphical method. Some inference about electronic band structures were made from comparison of experimental points with the theoretical curves.

Beattie⁽¹⁴⁾ measured the optical constants of silver in the infrared region and concluded that the theory of the anomalous skin effect proposed by Dingle agrees with experiments in the case of silver. There is one free electron per silver atom, and a fraction 0.77 of the conduction electrons were diffusely reflected at the surface of the metal. Since the mean free path of electron is not small compared to the penetration depth of radiation; the D.C. conductivity of the silver film was found to be 58% of the value for bulk silver.

These data justify a further analysis of the theory of the optical properties of noble metals. In the region between wavelengths 1.0μ to 10.0μ , where free electron absorption predominates, the Drude's theory predicts the optical constants with some success. But in the region of wavelength between 0.1μ to 1.0μ pronounced deviations from the Drude's theory occurs due to the effect of interband electronic transitions⁽¹⁶⁾, so the optical constants may be expected to depend most strongly on the band structure of the metal.

The theory of photoelectric interband absorption, as related to the band structure of a solid, was first put forward by Krönig⁽⁵⁾, and later by Wilson⁽¹⁷⁾ and others.

The theory has been developed further by Sergeiev & Tchernikovsky⁽¹⁸⁾, by Fan⁽¹⁹⁾, and by Butcher⁽²⁰⁾. The necessary assumptions about the band structures were made on the basis of the nearly free electron approximation.

Suffczynski⁽²¹⁾ has proposed a model which allows the calculation of the contribution of interband transitions to the optical constants of metals, taking into account the details which are important near the zone boundary. These details are ; the energy gap, the bending of the energy bands and the Fermi surface.

The principle aim of the present work is to make a study of the optical properties for opaque gold films in the visible and near infrared ranges of spectrum at room temperature. The measurements were carried on opaque evaporated films of spectroscopically pure gold specimens where the properties may be expected to approach those of bulk metal. Gold was chosen because it is not easily oxidised and can be considered stable against oxide film formation in the atmosphere.

The optical constants, the concentration of conduction electrons, D.C. conductivity, the relaxation time, the Fermi energy and the Fermi velocity were compared with those of the previous authors. The D.C. conductivity and the effective number of free electrons determined optically were also compared with those measured electrically in the present work on the same films.