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Optical and Electrical Properties of Au and Ag in Relation to Free Electron Theory

The results of new measurements of the refractive index n and the absorption coefficient k of Au and Ag are discussed with respect to free electron theory. This permits for the calculation of the concentration N of free electrons and D.C. conductivity σ_0, which are compared with values obtained from the electrical conductivity and Hall effect measurements taken for the same films.

1. Introduction

Outside the range of interband transition due to bound electrons, the optical constants n and k at wave length λ (frequency ω) are related to the electronic parameters of the metal in the following equations, based on the free electron theory of metals [1, 2, 3].

$$-\varepsilon = k^2 - n^2 = -1 + \frac{4\pi Ne^2/m^*}{\omega^2}$$

$$= -1 + \frac{4\pi\sigma_0\omega'}{\omega^2} = -1 + \frac{Ne^2/m^*}{\pi c^2}\lambda^2, \qquad (1)$$

$$\sigma = \frac{nk\omega}{2\pi} = \frac{Ne^2/m^*}{\omega^2}\omega'$$

$$= \frac{\sigma_0\omega'^2}{\omega^2} = \frac{\sigma_0\omega'^2}{4\pi^2c^2}\lambda^2, \qquad (2)$$

where, ε and σ are the frequency-dependent dielectric constants and electrical conductivity of the metal, respectively, N is the number of free electrons par unit volume, m^* is the effective mass of the electron, ω' is the self frequency of the electron defined as the reciprocal of the relaxation time τ, σ_0 is the D.C. conductivity, and c is the velocity of light in vacuum.

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2. Results and discussion

The results of the optical constants for Au and Ag have been reported in the previous paper. According to equation (1), the relation between $(k^2 - n^2)$ and λ^2 is linear. This is verified in case of Au and Ag, as shown in Fig. 1 and Fig. 2,

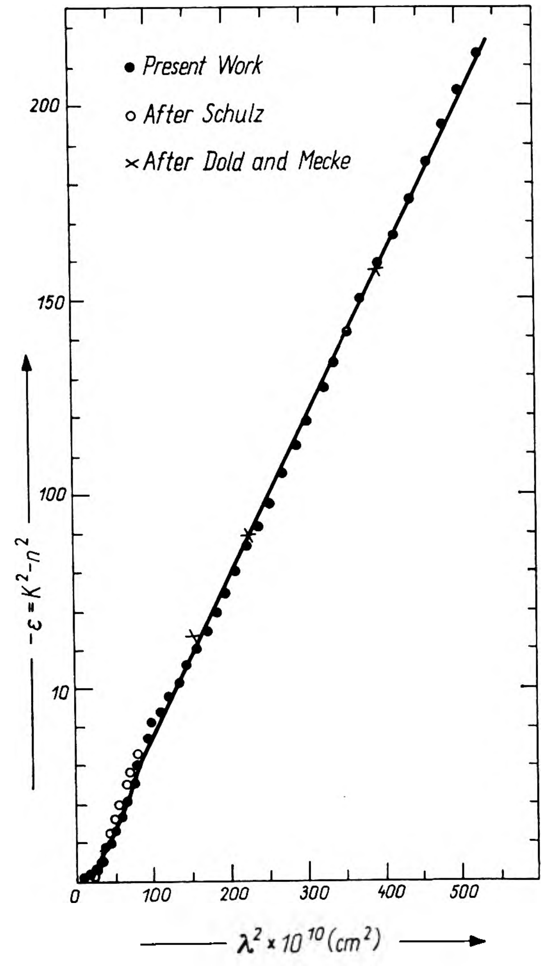

Fig. 1. Relation between $(k^2 - n^2)$ and λ^2 for Au

respectively, with data of previous authors for comparison [1, 2, 4]. N (optical) has been calculated from the slope of the straight line, considering $m^* = m = 9.1\times 10^{-28}$ gm. N_a (theoretical) has been also calculated ($N_a = dL/A$, where d is the density of the metal, A its atomic weight and L Avogadro's number) considering one free electron per atom. N (electrical) was deduced from the present Hall effect measure-

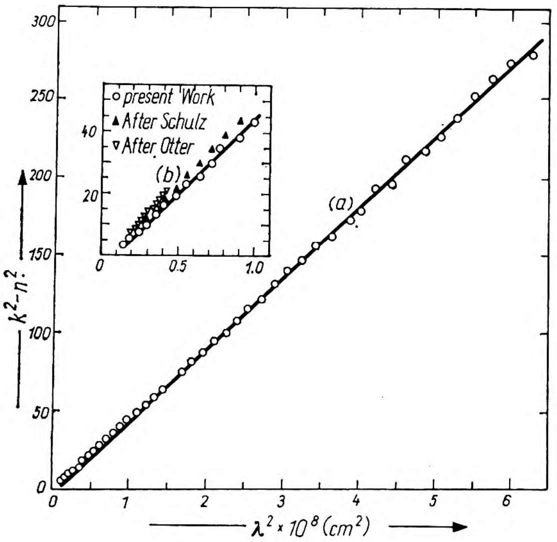

Fig. 2. Relation between (k^2-n^2) and λ^2 for Ag

ments for both Au and Ag films, which were used before in the optical measurements. The data thus obtained are listed in Table 1. As it is visible N (optical) is in fair agreement with N (electrical). The effective number of free electrons per atom N/N_a (optical) being also given. Hence, the optical effective mass m^*/m (opt.) may be calculated. The resulting values are comparable with that obtained by Čorak [5], Beaglehole [6] for Au (1.16 ± 0.08) and that reported by Givens [3] for Ag (0.98), respectively.

Table 1

	Au	Ag
N (optical	4.79×10^{22} elec./c.c.	5.2×10^{22} elec./c.c.
N_a (theoretical)	5.89×10^{22} elec./c.c.	5.9×10^{22} elec./c.c.
N (electrical)	4.96×10^{22} elec./c.c.	5.2×10^{22} elec./c.c.
N/N_a (opt.)	0.81	0.88
m^*/m $= N_a/N$ (opt.)	1.23	1.13

Fig. 3 represents the dependence of the conductivity $\sigma = nk\omega/2\pi$ on the wavelength λ for Au, giving a threshold of interband transition at 0.6 μm, corresponding to an energy $E = 2.07$ eV, due to the excitation of d electrons

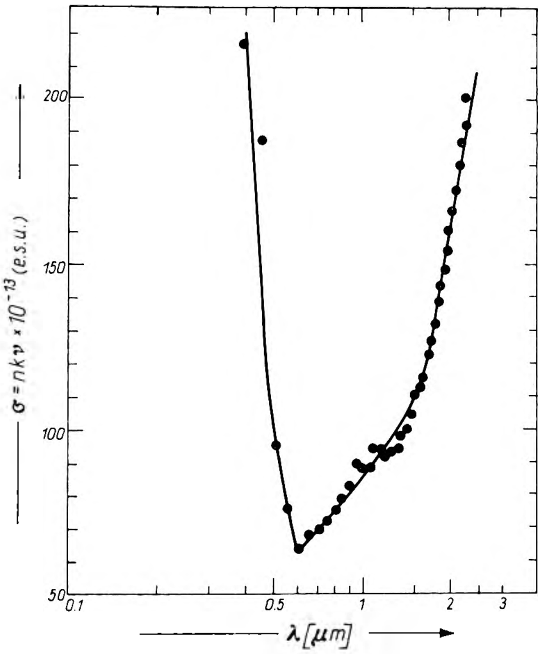

Fig. 3. The dependence of σ on λ for Au

to the conduction band [7, 8]. At wavelength region longer than the absorption edge, the condutivity σ increases with increasing λ, as expected from eq. (2).

Fig. 4 represents the dependence of $2nk/\lambda = 2\sigma/c$ on λ^2 for Ag, showing similar behaviour

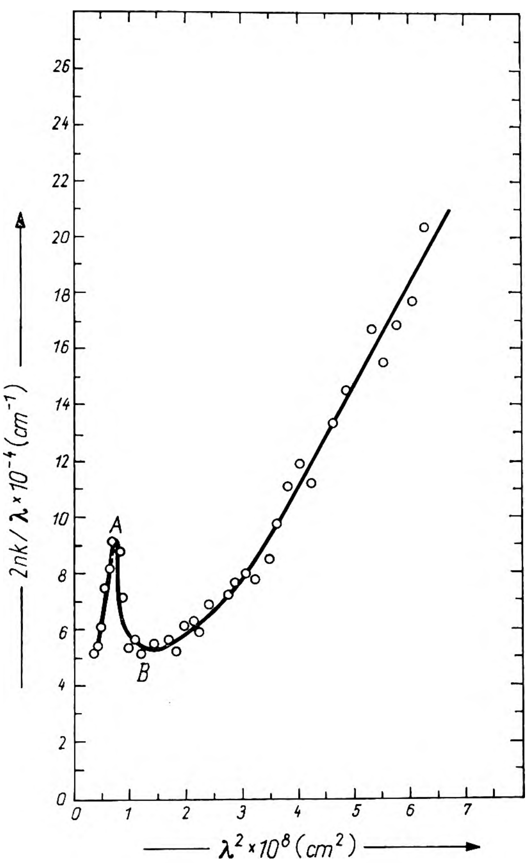

Fig. 4. Variation of $2\,nk/\lambda$ with λ^2 for Ag

as Au, and indicating a peak at $\lambda = 0.95$ μm, which is possibly associated with interband transition of electrons either from the Fermi-surface to the next higher empty band or from a lower lying filled band to the Fermi-surface [1, 9, 10, 11]. Beyond 1.5 μm, the curve shows a continuous increase of σ with increasing λ as the theory (eq. (2)) expects.

According to eqs. (1) and (2), the Argand diagram for Au in Fig. 5 represents (k^2-n^2+1) against $\sigma = nk\omega/2\pi$ showing two straight lines

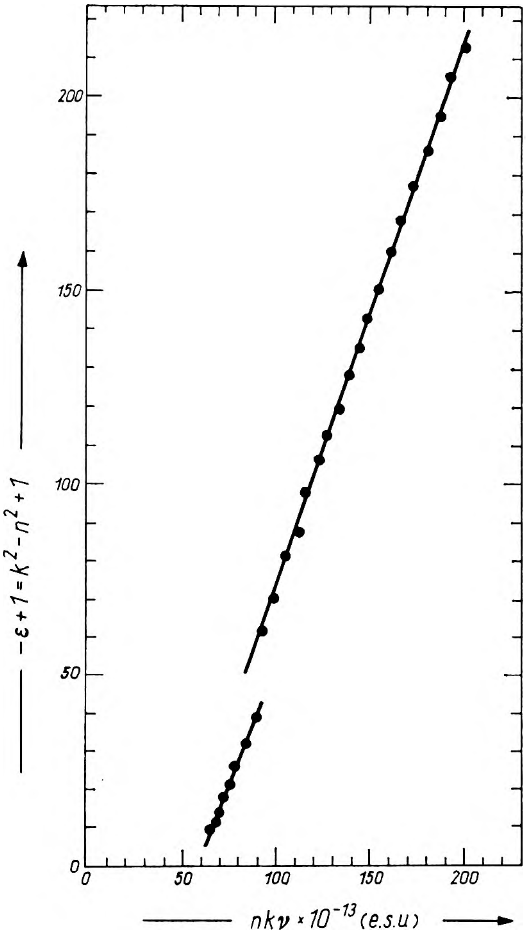

Fig. 5. The Argand diagram for Au

of different slopes (slope $= 4\pi/\omega' = 4\pi\tau$) corresponding to two values of relaxation time $\tau = 0.99 \times 10^{-14}$ and 1.1×10^{-14} s. This may be due to the fact the Fermi surface in Au is non-spherical [4, 12, 13, 14].

The relaxation time τ of the free electron in Ag, calculated from the slope of Fig. 2 and the slope of the linear part of Fig. 4, gives $\tau = 0.68 \times 10^{-14}$ s.

Using the values of N (opt) and τ, the D.C. conductivity σ_0 is calculated ($\sigma_0 = Ne^2\tau/m$). Values thus obtained are listed in Table 2 with the values of σ_0 deduced from the present electrical measurements on the same films used before in the optical measurements.

Table 2

	Au	Ag
τ (opt.)	1×10^{-14} s	0.68×10^{-14} s
σ_0 (opt.)	1.3×10^{17} e.s.u.	0.9×10^{17} e.s.u.
σ_0 (elect.)	4.2×10^{17} e.s.u.	1.92×10^{17} e.s.u.

It is clear that σ_0 (opt.) $<$ σ_0 (elect.). This is attributed to the fact that the electrons near the surface have frequent collisions; therefore they have a shorter mean free path and a smaller relaxation time τ than the electrons located deeper in the metal, which determine σ_0 (elect.). Since the light waves penetrate to a very short distance into the metal they interact only with the electrons near the surface, therefore σ_0 (opt.) is reduced [15, 16].

Les propriétés optiques et électriques de Au et de Ag rapportées à la théorie des electronst libres

On a examiné, par rapport à la théorie des électrons libres, les résultats des mesures effectuées pour Au et Ag et concernant l'indice de réfraction de la lumière n et le coefficient d'absorption k. Ceci permet de calculer la concentration des électrons libres N ainsi que la conductivité spécifique en courant continu σ_0. Les résultats ont été comparés avec les valeurs de N et de σ_0 obtenues des mesures de la conductibilité électrique et de l'effect Hall qu'on avait effectuées sur les mêmes couches.

Оптические и электрические свойства Au и Ag с точки зрения теории свободных электронов

Результаты измерений коэффициента преломления света и коэффициента поглощения k, произведенных для Au и Ag, обсуждены с точки зрения теории свободных электронов. Благодаря этому становится возможным расчет концентрации свободных электронов N и удельной проводимости для постоянного тока σ_0. Результаты сопоставлены со значениями N и σ_0, полученными путем измерений электропроводности и эффекта Холла, проведенных на тех же пленках.

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Received, March 24, 1974

Received in the revised form, July 20, 1974