THE RELATIONSHIP BETWEEN ELECTRICAL PROPERTIES AND INTERBAND AND INTRABAND TRANSITIONS OF THIN Cr FILMS

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ABSTRACT

Thin chromium films of thickness ranging from (25 to 80 nm) were prepared by thermal evaporation under a vacuum of 10^{-5} Torr. The electrical resistivity was inversely proportional to the thickness of the film. Analysis of the electrical resistivity was treated in the frame of the effective mean free path theory of size effect developed by Tellier et al. Such analysis allows the determination of the mean free path l_o, carrier concentration n_c, relaxation time τ and the Fermi energy F_F. The optical constants (n and K) of chromium thin films were determined in the spectral range of (200 to 25000 nm). The obtained results agree with the optical conductivities predicted theoretically by Moruzzi et al. In addition, the values of n_c, σ, l_o, and τ obtained electrically were found to match with those obtained optically.

INTRODUCTION

The theoretical calculations of the optical constants of metals might be interesting as there is an accumulating amount of experimental data which now become sufficient to allow comparison with theory. Moreover the theory might indicate where further measurements of the optical constants are needed to give more information on the band structure of metals[1]. However, band structure calculations for some of the transition metals have been performed, and in a few recent cases the calculated band structre has been used to obtain theoretical values for the interband optical absorption [2,3].

In the present study, electrical resistivity and optical constants have been obtained. The results were used to estimate the parameter of mean free path, carrier concentration, Fermi energy. Moreover, the contributions of interband as well as the intraband transitions have been extracted from the optical measurements.

2.EXPERIMENTAL TECHNIQUE.

This films of chromium Cr (purity 99.99%) were prepared in vacuum of 10^{-5} Torr by thermal evaporation on glass quartz and potassium bromide substrates held at room temperature, In all cases the cleaned substrates were masked until the source was at its evaporation temperature, and the evaporation rate was made as high as possible. the thicknesses of the films were determined with a multiple-beam interferometer of the Fizeau type. The electrical resistivity (ρ) of the samples was measured by the two-probe technique using a highly sensitive digital multimeter (type DA 8601) and a Univeka (type 141) URAN voltmeter. The transimttance of the prepared samples were measured at normal incidence in the spectral range 200 to 3000 nm using a Carey 2390 spectrophotometer, and in the spectral range 2500 to 25000 nm using Pycunicam 3P3-300IR spectrophotometer.

3 - RESULTS AND DISCUSSION

3.1. : Electrical Resistivity

The electrical resistivity (ρ) of Cr films as a function of thickness (t) is shown in Fig. (1). The data follow the known behaviour, where decreases by increasing t. The Fuchs-Sondheimer expression was used [4] to analyse the thickness dependence of the resistivity taking the surface scattering into account, along with the other kinds of scattering. Structural studies indicate that thin Cr films are polycrystalline[5] ; thus grain-boundary scattering also should be taken into account along with the surface scatting, when discussing the experimental results on film properties. The Mayadas-

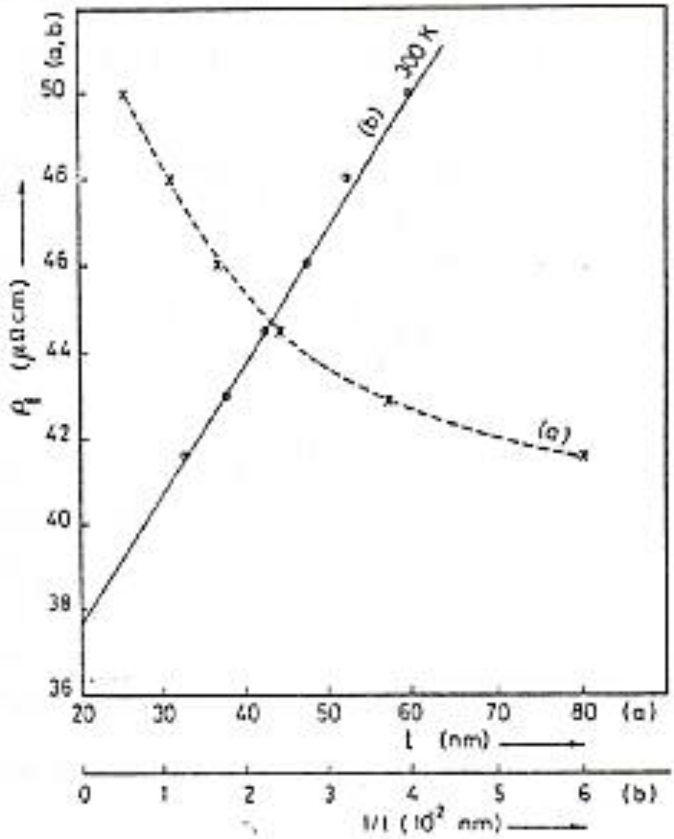

Fig. 1. (a) Relation between the thickness (t) of the Cr films and the corresponding resistivity (ρ).

(b) Relation between the resistivity (ρ) and l/t.

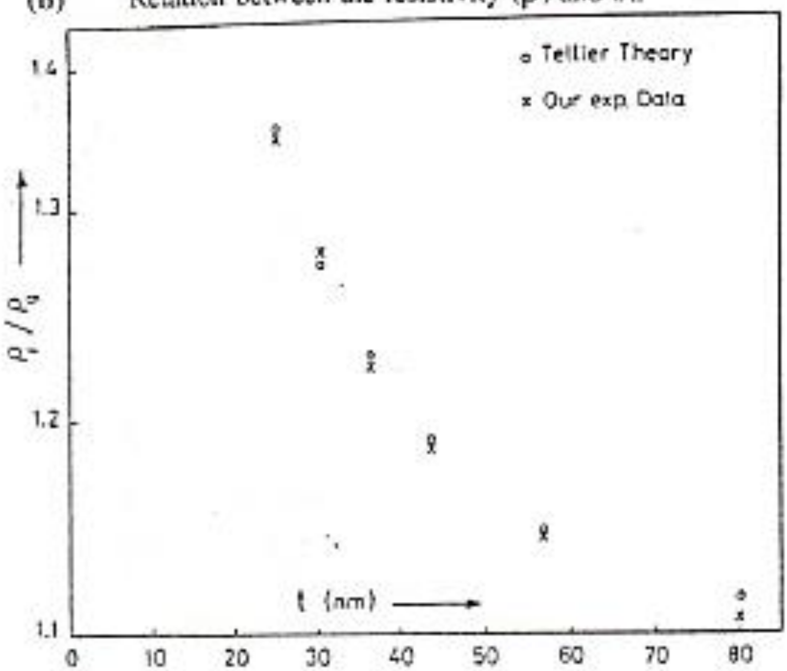

Fig. 2. Relation between the resistivity ratio (ρ_t/ρ_g) and thickness t.

Shatzkes theory takes into account the grain boundary scatting, but its expression [6] is complicated. Mola and Heras[7] , Tellier et al. [8] and Tellier et al.[9] attempted to simplify the analytical expressions of Mayadas-Shatzkes theory. Tellier et al.[9] derived such a simple analytical expression defining an effective mean free path in an infinitely thick film. This model is known as the effective mean free path model. According to which the analytical expression for the film resistivity as a function of thickness is given by :

$$\rho_f = \rho_g \left[1 - \frac{3}{8}\frac{1-\rho}{K_g}\right] \int_1^x \left(\frac{1}{x^3} - \frac{1}{x^5}\right)\left[\frac{1-\exp(-x)}{1-P\exp(-x)}\right] \qquad ...(1)$$

where ρ_g is the resistivity of the infinitely thick film, K_g is the reduced thickness (t/l_g). Under asymptotic conditions, $K_g >> 1$, the above equation can be reduced to the form :

$$\rho_f = \rho_g \left[1 - \frac{3}{8}\frac{L_g}{t}(1-p)\right] \qquad ...(2)$$

It has been shown numerically that the above expression is successfully applicable down to $K_g = 0.1$. It is evident from the above experession that a plot of film resistivity as a function of reciprocal thickness will be linear at a given fixed temperature. Obtained resistivity exhibits linear dependence on 1/t (Fig. 1), which agrees with equation (2). Estimated value of ρ_o was found to be 37.7 μΩcm for Cr films at 300°K. From the slope of the graph, the effective mean free path of carriers, L_o at at 300 °K was found to be 22.5 nm. There values differ from those given by Udachan et al. [10] in $\rho_o = 50$ μΩcm and $L_o = 107$nm with p = 0.3. This difference can be can be accounted for by the fact that Udachan et al. [10] in their study used Fuchs-Sondheimer [4] theory which does not consider the grain-boundary scattering also we can't ignore the difference in purity and condition of preparation, the latter greatly affect of grain boundaries. The dependence of (ρ_f/ρ_g) on (t) obtained was compared with the theoretical values of (ρ_f/ρ_g) according to Tellier theory Fig. 2. Excellent agreement is obviously seen. Mayadas-Shatzkes [6] showed that the presence of the grain boundary scattering reduces the mean free path of carriers, thereby reuslting in an increase in

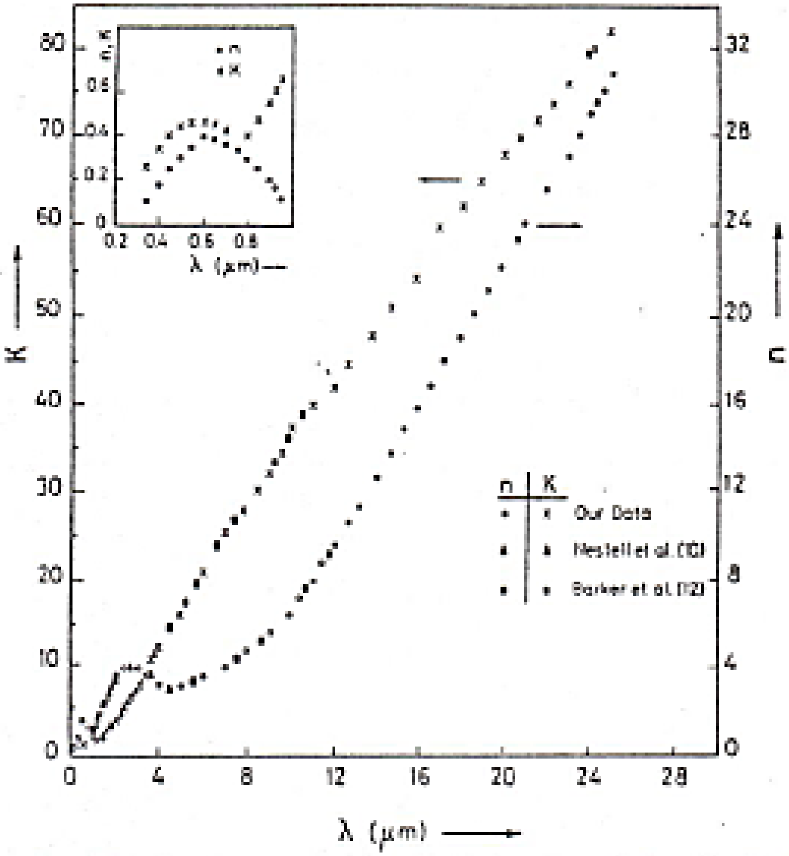

Fig. 3. Relation between n (o) and K (x) against (λ) for thin Cr films.

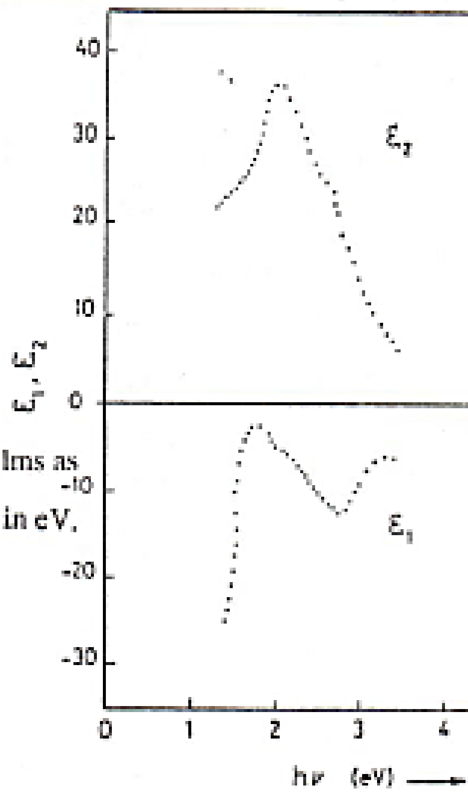

Fig. 4.

Dielectric constants for Cr films as a function of photon energy in eV.

resistivity. They showed that $\rho_g l_g = \rho_o l_o$ where ρ_g and l_g are the resistivity and the mean free path of carriers in the bulk with film microstructure, whereas ρ_o and l_o are for signle crystal. Electron density estimated according to the free electron theory was found to be 1.74×10^{21} cm $^{-3}$ using the equation [6,11] :

$$\frac{1}{\rho_o l_o} = \left(\frac{1}{3\Pi^2}\right)^{1/3} \frac{e^2 n^{2/3}}{\hbar} = \frac{1}{\rho_g l_g} \qquad ..... (3)$$

where $\hbar = h/2\pi$ and h is Planck's constant and e is the electronic charge. The corresponding Fermi energy in such case is found to be 0.246eV using the equation :

$$E_F = \frac{\hbar^2}{2m^*} (3\pi^2 n)^{2/3} \qquad ..... (4)$$

where m* is the effective mass of the electron [$m^* = 9.1 \times 10^{-28}$].

3.2 OPTICAL MEASUREMENTS

The obtained experimental results concerning the transimittance of Cr thin films were used to determine the optical constants (n and K) following the formula given in [12,13]. The refractive index n as well as the absorption index K does not depend on the film thickness. All of the results for films of different thicknesses fall within the range of the estimated error "In K is less than ± 1.5% and n is less than ± 0.4%".

3.2a INTERBAND CONTRIBUTIONS

Optical transitions in a solid can take place by more than one mechanism, the direct transition from the valence band to the conduction on is well known interband transition. However, at low energy I.R. the most probable mechanism in the intraband band, in which the electron transition takes place between two levels in the same band. In general, the electronic transitions in a solid are more directly related to the complex dielectric constant $\bar{\varepsilon} = \varepsilon_1 + i\varepsilon_2$ instead of the complex index of refraction n = n + iK, where $\varepsilon = n^2$ [14]. So that $\varepsilon_1 = n^2 - K^2$ and $\varepsilon_2 = 2nk$. On the other hand, the dielectric constants ε_1 and ε_2 are closely related to the electronic structure of the solids and are more direcly comparable with theory. Obtained dielectric conststants for Cr are presented in Fig. 4, where the curves represent the average of values from four films of thickness 28, 30, 34 and 40 nm respectively. The agreement of the four results is well within the estimated error. It is clear from Fig. 4 that there are two peaks at hv = 2eV and hv = 1.75 eV for ε_1 and ε_2 respecitively. However, for graphical representation of the results in case of the transition metals, the real and imaginary parts of the optical conductivity are more convenient. The optical conductivity (σ_1 and σ_2) are related to the components of the complex dielectric constant by $\sigma_1 = \varepsilon_2\omega/4\pi$ and $\sigma_2 = (1 - \varepsilon_1)\,\omega/4\pi$, where ω ($=2\Pi v$) is the angular frequency of the incident radiation; the real and imaginary parts of the optical conductivity of thin Cr films are shown in Fig. 5. Several attempts have been made previously to determine the free-electron contribution by extending reflection measurements further into the infrared region. Lenham and Treherne[15] made measurements up to 15μm for most of the metals including the chromium. Barker and Ditzenberger [16] used a free-electron gas model to estimate the size of the anomalous skin effect and to calculate the corresponding absorptivity for chromium. Both the theoretical prediction [16] and the experimental evidence [15] indicate that below 2μm all the measurements are presumed to be enetirely within the interband portion of the spectrum.

Accordingly, we can consider that the interband mechanism, is responsible for the interaction in the considered spectral region (hv<4eV). In this interband region of the spectrum, we will compare our results with a recent calculation of optical conductivity. Moruzzi, Williams and Janak[2] have calculated the interband contribution to ε_2 for Cr. These self-consistent effective one-electron calculations, used the approximation treatment of exchange and correlation due to Kohn and Sham,[17], and the "muffin-tin" approximation to both the charge density and the potential. The results are compared with the obtained experimental values of σ_1 in Fig. 6. The agreement is encouraging, the experimental peak near 2 eV can be correlated with the theoretical one, but it is generally broader and lower than the theoretical one. This difference may be partly because the measurements were made at room temperature, and that the temperature dependence can be strong enough to account for

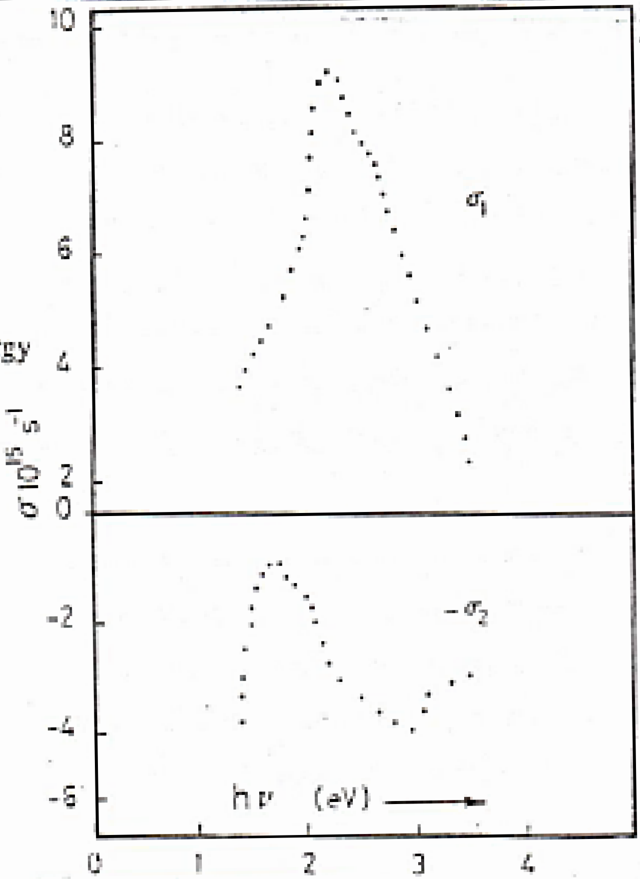

Fig. 5

Optional conductivity in $10^{-15}s^{-1}$ for Cr films as a function of phonon energy in eV.

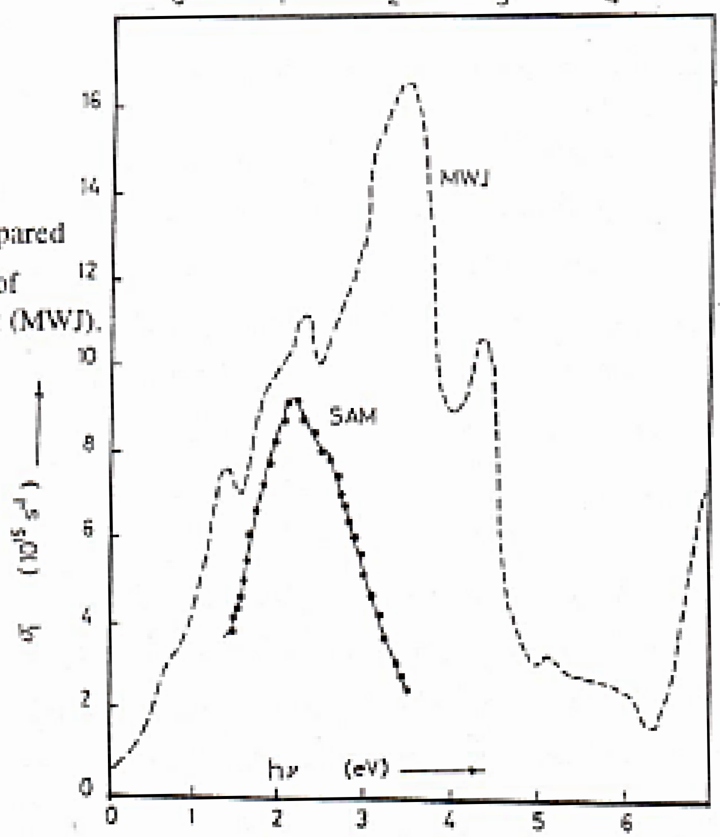

Fig. 6.

Our experimental optical conductivity (SAM) compared with the theoretical curve of Moruzzi, Williams and Janak (MWJ).

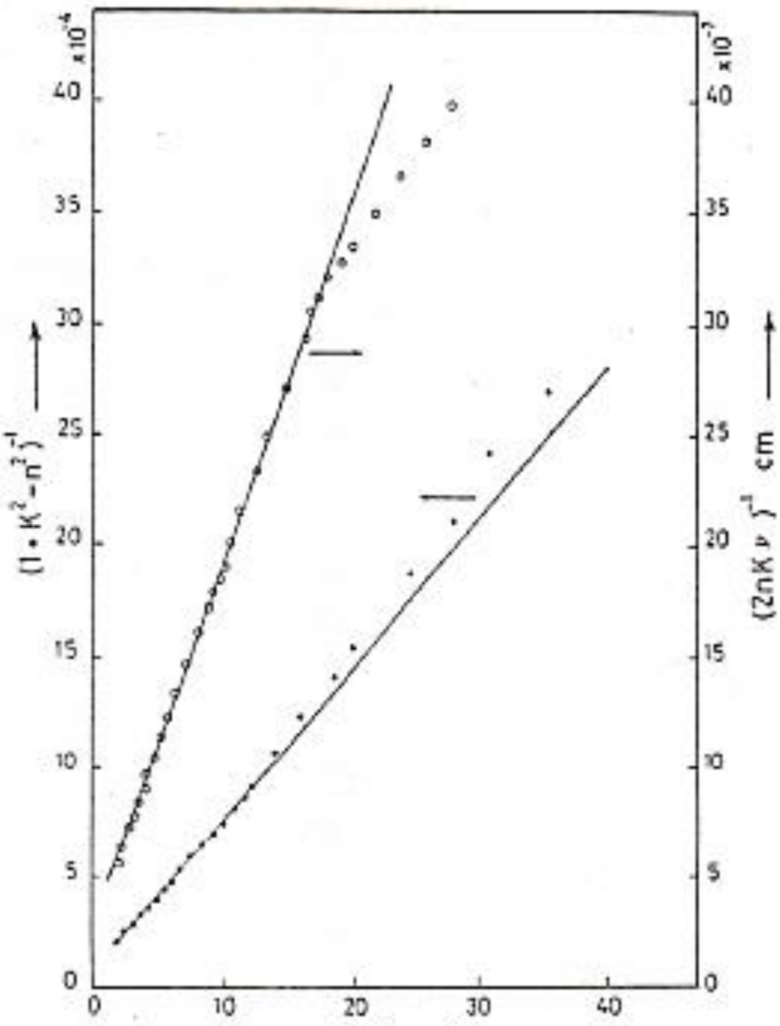

Fig. 7. Relation between $(1+K^2-n^2)$ and $(2nKn)^{-1}$ with v^{-2} for Cr thin films.

the entire discrepancies. The theoretical curves of Cr have a peak just above 2 eV corresponding to 3 $\rightarrow$ 4 transitions. Contributions are coming from transitions over a large volume of k space along the three major directions (Λ, Δ and Σ). The 3 - 4 transitions corresponding to the 2 eV peak probably occur away from a symmetry point along A or near G.

3.2b INTRABAND CONTRIBUTIONS :

Most stuides in which the intraband and interband processes have been separated, have been separated, have used the Drude theory [18]. According to the previous theory, there are two relations relating the optical constants (n and K) to the wavenumber of the incident radiation (v). These two relations are :

$$(1+K^2-n^2)^{-1} = (\bar{v}_o)^{-2} [(\bar{v})^2+(\bar{v})^2+(\bar{v}_R)^2] \quad \text{..............(5)}$$

and

$$(2nK\bar{v})^{-1} = (\bar{v}_o)^{-2} (\bar{v}_R)^{-1}[(\bar{v})^2+(\bar{v}_R)^2] \quad \text{..............(6)}$$

where $\bar{v}_o$ is the plasma frequency and $\bar{v}_R$ is the damping frequency. Thus the values of $\bar{v}_o$ and $\bar{v}_R$ were obtained from Fig. 7 and were found to be 3.82 x 10 cm^{-1} and 413.3 cm^{-1} respectively. In addition to equations (5) and (6),

$$(\bar{v}_o)^2 = n_c e^2 / \Pi m^* c^2 \quad \text{..............(7)}$$

and

$$\bar{v}_R = 1/2\pi c\tau \quad \text{..............(8)}$$

where n_c is the number of conduction electrons per unit volume, e the electron charge, τ the relaxation time, m* the effective mass and c the light velocity in vacuum. Using the values of $\bar{v}_o$ and $\bar{v}_R$ in conjunction with equations (7) and (8) n_c, τ were calculated and are given in Table (1). Taking into account that $m^* = m = 9.1 \times 10^{-24}$ gm. Knowing n_c and τ the static conductivity (σ) in Cr films can be calculated using.

$$\sigma = n_c e^2 \tau / m^* \quad \text{..............(9)}$$

It was found that s = 5.28x1016 (e.s.u.). The electron velocity at the Fermi surface V_F can be determined using the following equation :

$$V_F = (3n_c h^3/8m^{*2}e^2)^{1/3} \qquad \text{.............(10)}$$

V_F was calculated and tabulated in Table (1). It was possible to define l_g the mean free path by using the formula $l_g = \tau V_F$. It was found that l_g = 19.9nm. The small values of l_g indicate that electrons are strongly interacting with each other and with lattice. This is a known effect for transition metals, which are characterized by a narow 3d band. All the calculated parameters using the optical properties are given in Table (1) in comparison with those obtained previously from the electrical measurements.

Table. 1. : Values of n_c, σ, τ, l_g and E_F determined from electrical measurements compared with that obtained from optical measurements.

	n_c cm^{-3}	σ e.s.u.	τ (s)	l_g nm	E_F eV	V_F cm/s
Electrical	1.74×10^{21}	2.4×10^{16}	—	22.5	0.246	---
Optical	1.63×10^{22}	5.28×10^{16}	1.28×10^{-14}	19.9	0.234	1.55×10^{8}

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