

CORRELATION OF THE OPTICAL PROPERTIES AND INTERBAND TRANSITION OF VANADIUM THIN FILMS

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The possibilities of using the optical constants n and k of vanadium thin films, to calculate the free-electron, optical effective masses and relaxation time which were derived from the results, are discussed from the theoretical and experimental point of view. Throughout the optical constants, the optical conductivity $\sigma = \sigma_1 + i\sigma_2$ as well as the volume and surface energy loss function are derived. The interband contribution to the imaginary part of the dielectric constant is also obtained by subtracting the free electron contribution.

Key words: Optical constants (n & k), Vanadium thin films, Transition.

Introduction

The optical constants for a limited number of transition metals have been accurately determined throughout a continuous spectral range from the near infrared to the near ultraviolet. However, band structure calculations for some of transition metals are now being performed, and in a few recent cases, the calculated band structure have been used to obtain theoretical values for the interband optical absorption [1,2]. The determination of the optical constants n and k of vanadium in thin film form of thickness varied from 50 to 100 nm, in the spectral range of 2.5-8.5 μm was previously discussed [3]. The previously obtained results revealed a relatively small discrepancy in the calculated values of n and k , in comparison with the mean value in the whole range of wavelength. This leads us to the conclusion that both parameters (n , k) are independent on the film thickness.

The purpose of this study is to use the obtained optical constants of thin vanadium films in the spectral range of 2.5 - 8.5 μm in the determination of some recent theoretical calculations of the interband optical conductivity as well as the lattice dielectric constant.

Results and Discussion

As mentioned before, the optical constants (n & k) of V films of different thicknesses in the thickness range between 50 and 100 nm do not depend on the film thickness, thus the absorption A can be calculated using the following formula:

$$A = \frac{4n}{(n+1)^2 + k^2} \quad \dots\dots\dots(1)$$

Figure 1 illustrates A against $h\nu$. This relation has a peak at $h\nu = 0.415$ eV corresponding to wavelength (λ) = 2.99 μm .

The electronic transition in a solid is more directly related to the complex dielectric constant $\epsilon = \epsilon_1 + i\epsilon_2$, instead of the

complex index of refraction $n = \pi + ik$. These are connected by $\epsilon = \bar{n}^2$, so that $\epsilon_1 = n^2 - k^2$ and $\epsilon_2 = 2nk$. The dielectric constant is determined by the relaxation time and the optical mass of the electrons according to the Drude free-electron theory:

$$\tilde{\epsilon}^f(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i/\tau)} \quad \dots\dots\dots(2)$$

where $\omega_p^2 = \frac{4\pi N e^2}{m_0}$. Here N is the density of the

conduction electrons, m_0 is their effective optical mass and τ is the relaxation time. Separating $\tilde{\epsilon}^f$ into its real and imaginary parts, we get:

$$\epsilon_1^f = \frac{1 - \omega_p^2 \tau^2}{1 + \omega^2 \tau^2} \quad \dots\dots\dots(3)$$

and

$$\epsilon_2^f = \frac{1 - \omega_p^2 \tau}{\omega(1 + \omega^2 \tau^2)} \quad \dots\dots\dots(4)$$

For metals at near-infrared frequencies one finds that $\omega \gg 1/\tau$. Therefore:

$$\epsilon_1^f \cong 1 - \frac{\omega_p^2}{\omega^2} = 1 - \frac{\lambda^2}{\lambda_p^2} \quad \dots\dots\dots(5)$$

and

$$\epsilon_2^f \cong \frac{\omega_p^2}{\omega^3 \tau} = \frac{\lambda^3}{\lambda_p^2} \tau' \quad \dots\dots\dots(6)$$

where $\lambda_p^2 = Ne^2 / \pi m_0 c^2$ and $\tau' = 2\pi c \tau$. We can notice that the optical mass can be determined using the experimental results for ϵ_1^f , while τ can be then found from the results for ϵ_2^f . Therefore, a plot representing ϵ_1 against λ^2 will yield a

straight line, the point of intercept on the y-axis yields the required value of ϵ_L , while the optical mass is determined from the slope [4]. Figure 2 illustrates $\epsilon_1 = f(\lambda^2)$. It is seen that the lattice dielectric constant ϵ_L and the optical mass are found to be 17 and 1.3 m respectively.

On the other hand, Fig. 3 shows the relation between ϵ_2/λ and λ^2 , from which τ is found to be 10×10^{-15} S. These results are in good agreement with those reported by other workers [3-5].

The volume and surface energy-loss functions $-\text{Im}(1-\epsilon)$ and $-\text{Im}(1/\epsilon + 1)$ describe the probability that fast electrons will lose energy when traversing the bulk and surface of the material respectively. They are related to the real and the imaginary parts of the complex dielectric constant (ϵ) by the

following relation [6]:

$$-\text{Im}\left(\frac{1}{\epsilon}\right) = \frac{\epsilon_2}{\epsilon_1^2 + \epsilon_2^2} \dots\dots\dots (7)$$

$$-\text{Im}\left(\frac{1}{\epsilon + 1}\right) = \frac{\epsilon_2}{[(\epsilon_1 + 1)^2 + \epsilon_2^2]} \dots\dots\dots (8)$$

Figure 4 illustrates the spectral behaviour of both volume and surface energy loss functions against the photon energy of the incident radiation ($h\nu$). It is clear that, the photon energy lost on traversing the bulk is nearly equal to that lost on traversing the surface. Both functions show a peak at 0.415 eV. However in the case of transition metals, the optical conductivity $\sigma_1 + i\sigma_2$ is more convenient. The optical conductivity (σ_1 & σ_2) are related to the components of the complex dielectric constant by $\sigma_1 = \epsilon_2 \omega/4\pi$ and $\sigma_2 = (1 - \epsilon_1)\omega/4\pi$ where $\omega (=2\pi\nu)$ the

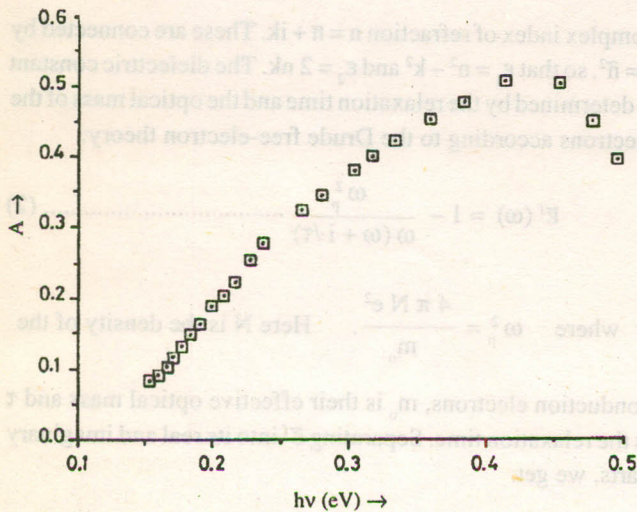


Fig. 1. Variation of the absorption (A) with the photon energy (hv).

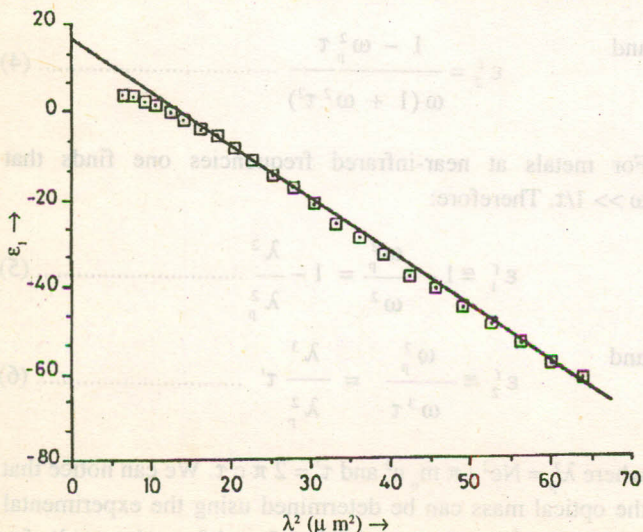


Fig. 2. Plot of real part of the dielectric constant with λ .

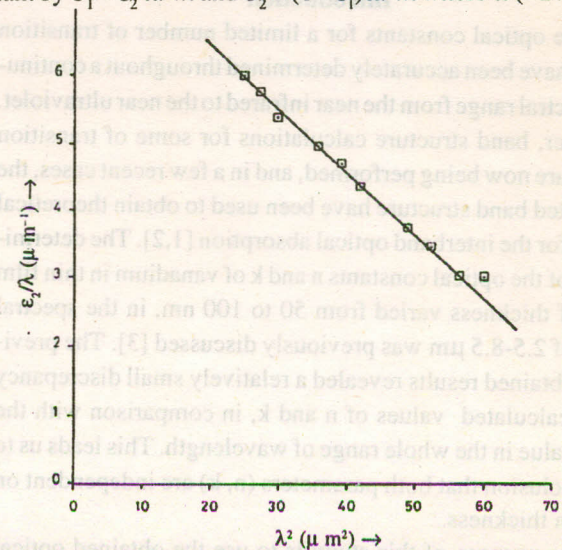


Fig. 3. Plot of ϵ_2/λ vs. λ^2 .

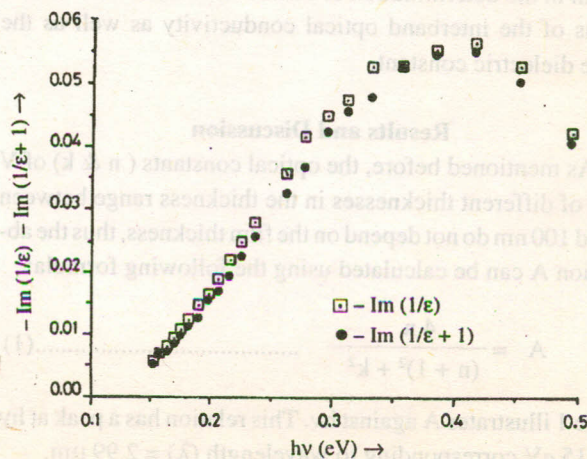


Fig. 4. The spectral behaviour of the volume energy loss function $-\text{Im}(1/\epsilon)$ and the surface energy loss function $-\text{Im}(1/\epsilon + 1)$ vs. ($h\nu$).

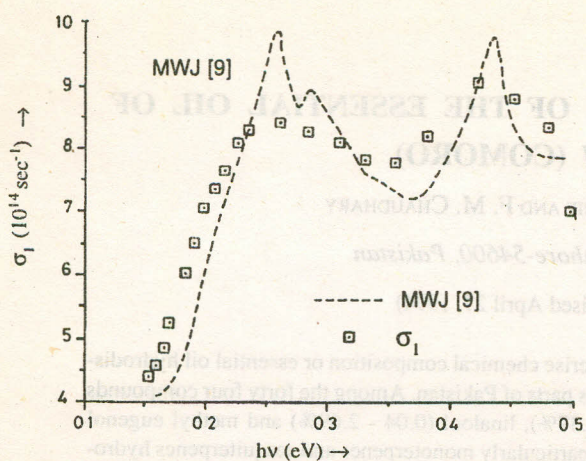


Fig. 5. The real part of the optical conductivity (σ_1 as a function of $h\nu$), compared with the theoretical curve of [9].

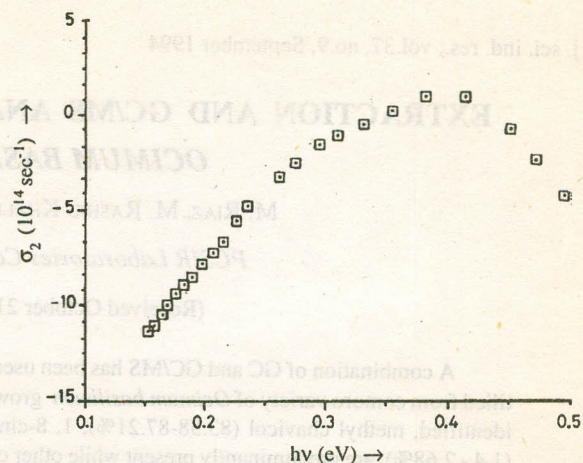


Fig. 6. The plot of $(-\sigma_2)$ of the optical conductivity vs. $(h\nu)$.

angular frequency of the incident radiation σ_1 and σ_2 calculated from the optical constants are illustrated as a function of $h\nu$ (eV) (Figs. 5 and 6). It is seen that $\sigma_1 = f(h\nu)$ shows two distinct maxima, the sharp one at 0.416 eV and the broad one at 0.25 eV. The peak existing at 0.416 eV is in harmony with those peaks observed throughout the $A=f(h\nu)$ represented at 0.415 eV, and that given through the volume and surface energy loss function represents at 0.415 eV. This peak may be attributed to spin-orbit splitting, which is in good agreement with Hedin and Kohn [1 and 7]. Several attempts have been made previously to determine the free-electron contribution by extending reflection measurements further into the infrared region. Lenham and Treherne [8] made measurements up to 15 μm for most of the metals including the vanadium. At these long wavelengths, they determined values for the optical mass and the relaxation time, but even their plots clearly showed contributions of interband transitions. In this interband region of the spectrum, we will compare the obtained results with two recent calculations of optical conductivity. Moruzzi *et al.* [9] have calculated the interband contribution to ϵ_2 for vanadium, chromium and nickel. These self-consistent effective one-electron calculations use the approximate treatment of exchange and correlation due to Kohn and Sham [7] and the 'muffin-tin' approximation to both the charge density and the potential. These approximation permitted the evaluation of the excitation energies and probabilities throughout the Brillouin

zone. The results are compared with the obtained experimental values of σ_1 in Fig. 5. The agreement is encouraging, all the experimental peaks can be correlated with the theoretical ones, but they are generally broader and lower than the theoretical one. The theoretical curve for V have high-energy peak (near 0.25 eV), this peak appears to be due to transitions between band 1 \rightarrow 6 (where the bands are numbered in order of increasing energy at each k), in the case of 1 \rightarrow 6 transition, both bands appear to be more free electron-like.

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