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MEASUREMENTS OF THE OPTICAL CONSTANTS OF NICKEL IN THE INFRARED REGION

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The optical properties were studied and the values of n and k for nickel films deposited on different substrates under different environmental conditions were determined. The variation of the real and imaginary parts of the dielectric constants and optical conductivity was studied. The structures were explained in terms of interband transition at higher energies. At energies lower than 0.18 eV the relaxation mechanism and the variation of ε_1 with λ^2 indicated that the interband acceleration of the conduction electrons dominated over the interband transition mechanism.

1. Introduction

In thin metal films, the optical constants are different from those of the bulk materials. The variation is due to the difference in the microstructure between the film and the bulk material. Usually thin metal films possess granular structure. Inside the grains the optical constants approach those of the bulk material. Following the optical constants of thin films of material, one may be able to get the corresponding values for a film of infinite thickness of the same material which is supposed to approach the bulky sample. In case of thin films, one may be able to employ the transmission technique to get n and k with better values representing the bulk sample and to some extent independent of the surface of the sample.

Kirillova[1] studied the electron interband transition in nickel single crystal. Some features of the interband transitions were studied. Her results reveals some weak bursts at 0.1, 0.14 and 0.17 eV on the optical conductivity-photon energy curve. These features allowed her to conclude that the absorption edge in nickel is apparently located at energies much lower than the value of 0.25 — 0.3 eV suggested previously[2,3]. Moreover she was able to fit her data to an energy band diagram proposed by Krinchik et al.[4] and Hanus et al.[5]. As well she concluded that her data favours the view of interband transition which begins to contribute significantly to the optical conductivity at energies starting just above 0.25 eV. In spite of that her data in the low energy region was assigned to some electron transition between ($s - p$) like $L_2{\downarrow}$ and d-like $L_3{\downarrow}$ bands at the point L in the Brillouin zone.

In the present work, the optical transmission technique was employed in conjunction with thin semi-transparent nickel films. Measurements were made in the spectral region from 2.5 μm up to 40 μm.

2. *Experimental techniques*

For studying the optical properties of thin nickel films in the infrared region, series of samples were deposited on freshly cleaved surface of mica and potassium bromide discs which were held at 160 °C during deposition. The annealed samples were heated in situ just after their preparation. The nickel samples were obtained by vacuum evaporation of nickel wire of 99.999% purity in a vacuum of about 10^{-4} Pa. The filaments used for the evaporation were of tungsten and the deposition rate was about 1.5 nm/s. The thickness of the samples was measured by multiple-beam interferometry[6]. Optical transmision measurements over the range 2.5 μm to 40 μm using double beam recording spectrophotometer (Beckman 4220) giving transmission values accurate to 1%.

3. *Results and discussion*

Fig. 1 shows the variations of k with photon energy for nickel films deposited on KBr and mica substrates. Some structures in the n and k versus energy curves are observed at 0.14 and 0.19 eV on both substrates. The effect of the substrates disappears at energies above 0.3 eV. The effect of thickness on the values of k is clear. Fig. 2 shows the variation of n with photon energy. For thicker films there is no effect for the substrate after the energy exceeds 0.2 eV. As the thickness increases the values of n and k increase. On annealing the thin film at 420 °C for three hours in situ, the values of n and k are raised as shown in Fig. 3 for a film at 40 nm thick deposited on mica at 160 °C. The results are compared with the data of Kirillova[1] for electrolytically polished nickel, and the data of Johnson and Christy[7] for an evaporated nickel layer. The results are in good agreement, however, the values of n are closer to those for electrically polished nickel. This close agreement may be attributed to the good quality of the surface of the samples.

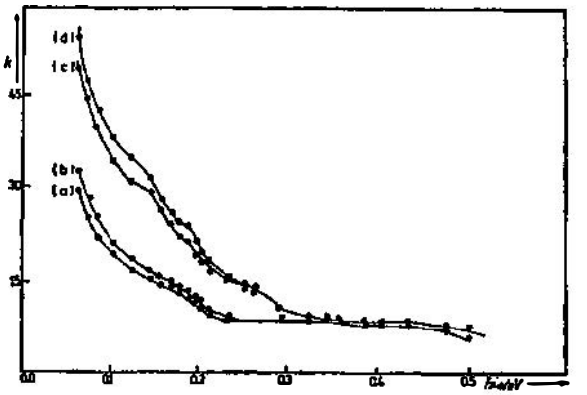

Fig. 1. The variation of k with $\hbar\omega$ in the I. R. region.
(a) $d = 40$ nm on mica, (b) $d = 40$ nm on KBr.
(c) $d = 45$ nm on mica, (d) $d = 45$ nm on KBr.

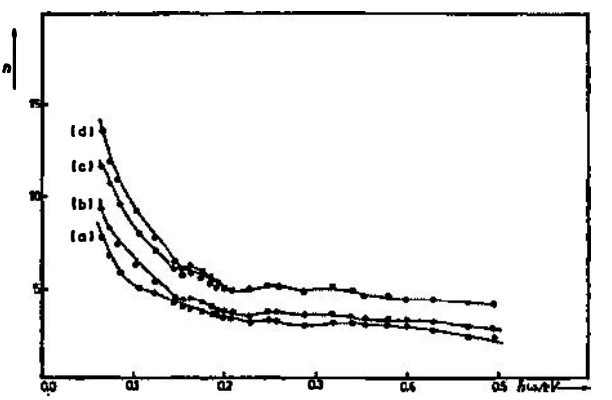

Fig. 2. The variation of n with $\hbar\omega$ in the I. R. region.
(a) $d = 40$ nm on mica, (b) $d = 40$ nm on KBr.
(c) $d = 45$ nm on mica, (d) $d = 45$ nm on KBr.

Fig. 4 shows the dependence of the real and imaginary parts of the dielectric constant on the photon energy. It is clear from Fig. 4 that the dielectric constant at low energy goes to large values and nothing could be got out of the data. On the other hand, the real part of the conductivity $\sigma_1(\omega)$ is finite and exhibits very weak structures at energies below $\hbar\omega = 0.2$ eV. Roberts[8] interpreted his low energy data dealing with the 0.3 eV structure using a formula due to Drude which contains both free carriers and oscillator-like terms. He explained the 0.3 eV structure by invoking several types of free carriers, one high mobility carrier which was assigned to account for the magnitude of the conductivity, and two low mobility carriers characterized by a relaxation time of the order of 10^{-16} s. The low mobility carriers were parametrized to dominate above 0.3 eV and produce the observed weak structure.

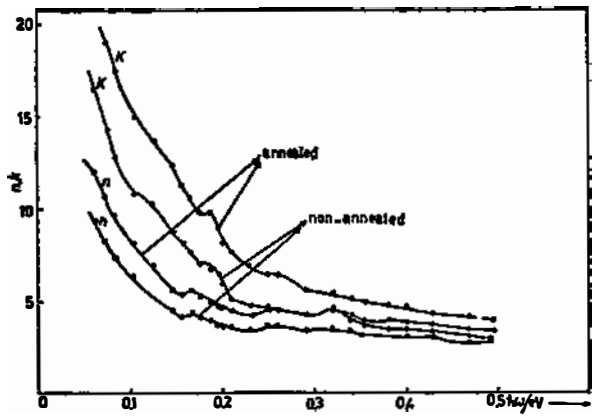

Fig. 3. Variation of n, k for the annealed and non-annealed nickle film 40 nm thick on mica.

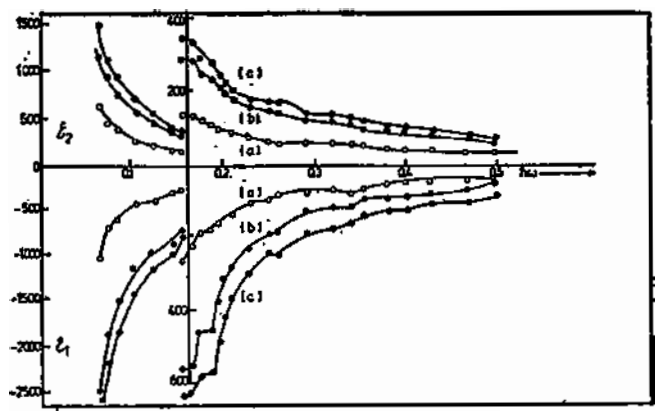

Fig. 4. Variation of ε_1 and ε_2 with $\hbar\omega$ in the I. R. region.
(a) $d = 40$ nm on KBr, (b) $d = 45$ nm on mica,
(c) $d = 45$ nm on KBr.

Fig. 5 shows the variation of the optical conductivity $\sigma_1 = \varepsilon_2 \omega/4\pi$ with the photon energy. The structure $\sigma_1(\omega)$ are listed in Table 1.

TABLE 1

$\hbar\omega$ (eV	Type of structure
0.1 — 0.11	a burst on KBr substrate
0.18	a shoulder
0.25	well-defined weak peak
0.33	well-defined weak peak
0.40	a weak peak with a wide plateau

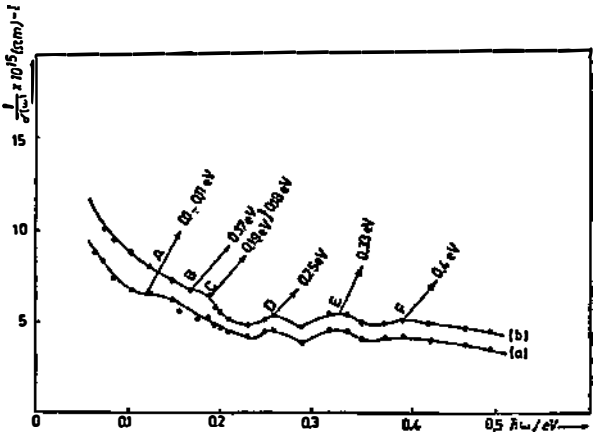

Fig. 5. Variation of $\sigma_1(\omega)$ with $\hbar\omega$ in the I. R. region.
(a) on KBr, (b) on mica.

The structures appeared on the variation of the optical conductivity with photon energy.

The existence of structures at 0.11 eV and 0.17 eV in the band spectrum of nickel was already noted earlier in optical[9] and magneto-optical[10] experiments. The structure found at 0.25, 0.32 and 0.4 eV were also reported by Kirillova[1] either from her optical studies on nickel single crystal or from magneto-optical and temperature-modulation measurements. The frequency dependence equatorial Kerr effect reveals similar results[11,12]. The energy gap determined from the thermal reflection spectra by Hanus et al.[5] is 0.25 eV. In the present work a structure was found around this value of energy.

The different structure found in the present work are explained in terms of the band structure of nickel near the L — point of the Brillouin zone.

The model of the energy bands of ferromagnetic nickel proposed by Ehrenreich et al.[2] assumed the arrangements of the energy bands at L — point such that $E(L_2'^{\downarrow}) - E(L_{32}^{\downarrow}) > 0$. Hodges et al.[13] explained many experimental data for nickel using this model. Krinchik[4] and Hanus et al.[5] proposed a reversed model in which they assumed $E(L_{32}^{\downarrow}) - E(L_2'^{\downarrow}) > 0$. Later on Zenberg[14] arrived to the conclusion that the model proposed[4,5,15] is the most suitable for both spin subbands as shown in Fig. 6. Hanus et al.[5] interpreted the spectrum of the thermal reflection and suggested that the peak at 0.25 eV corresponds to the transition $E(Q_-^{\uparrow}) - E(Q_+^{\uparrow})$ and the peak at 0.4 eV to the transition $E(L_{32}^{\downarrow}) - E(L_2'^{\downarrow})$. Such arrangements of the levels corresponds to d-exchange splitting of the bands at the point $L \sim 0.4$ eV. At the same time Zenberg[14] suggests the opposite to ascribe the two structure. This leads to a change in the position of the d-like band relative to the Fermi-level and 0.57 eV for the value of the exchange splitting.

The present results which agree with those of Kirillova[1] may be explained in terms of the energy gap model given by Hanus et al.[5]. The complex structure of the absorption band is the result of the splitting the L_{32} band by spin orbit

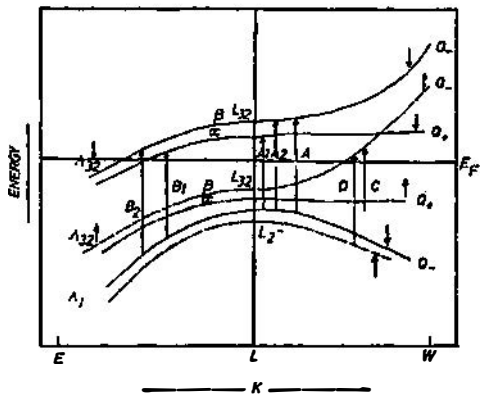

Fig. 6. Energy bands near L — point

$E(L_{32}\downarrow) - E(L_2'\downarrow) > 0$

$E(Q_-\uparrow) - E(Q_+\uparrow) \approx 0.25$ eV $\quad \uparrow C$

$E(L_{32}\downarrow) - E(L_2'\downarrow) > 0.4$ eV $\quad \uparrow A$

$E(L_{32\alpha}\downarrow) - E(L_2') \approx 0.33$ eV $\quad \uparrow A_1$

$E(L_{32\beta}\downarrow) - E(L_2'\downarrow) \approx 0.43$ eV $\quad \uparrow A_2$

With a difference of peak energies $\Delta E_{\alpha 13} \sim 0.1$ eV

$E(\Lambda_{32\alpha}\downarrow) - E(\Lambda_1\downarrow) \approx 0.69$ eV $\quad \uparrow B_1$

$E(\Lambda_{32\beta}\downarrow) - E(\Lambda_1\downarrow) \approx 0.87$ eV $\quad \uparrow B_2$.

interaction[1]. The structure at 0.33 eV is ascribed to the transition $E(L_{32\alpha}\downarrow) - E(L_2'\downarrow)$ and 0.42 eV to the transition $E(L_{32\beta}\downarrow) - E(L_{2\downarrow}')$ with a difference of 0.09 eV between the peak energies corresponding to the magnitude of the spin-orbit splitting of the $L_{32}\downarrow$ level at the point (L).

The structure at 0.18 eV is ascribed to the transition between the split sublevels $\Lambda_{32\alpha}\downarrow$ and $\Lambda_{32\beta}\downarrow$. This estimation is based on the variant that $E(\Lambda_{32\beta}\downarrow) - E(\Lambda_1\downarrow) = 0.87$ eV and $E(\Lambda_{32\alpha}\downarrow) - E(\Lambda_1\downarrow) = 0.69$ eV with splitting difference of 0.18 eV. The other variant is based on the assumption that:

$E(\Lambda_{32\alpha}\downarrow) - E(\Lambda_1\downarrow) = 0.57$ eV and

$E(\Lambda_{32\beta}\downarrow) - E(\Lambda_1\downarrow) = 0.69$ eV with splitting difference of 0.12 eV.

The present work shows that the conductivity decreases with increasing frequency as in Fig. 5 for the nearly continuous film, which implies that the relaxation effect is dominating. Harris et al.[16] derived a relaxation connecting the inductance and wavelength in the form:

$$\frac{\mu C\, \sigma_{d.c}\, a}{\mu C \sigma(\omega)\, a} = 1 + \tau^2 \frac{(2\pi C)^2}{\lambda},$$

where $\sigma_{d.c}$ is the direct current optical conductivity. Fig. 8 shows the relation between $\frac{1}{\sigma(\omega)}$ and $\frac{C^2}{\lambda^2}$ for two nickel films of 45 nm and 40 nm thick on KBr substrate,

held at 160 °C during deposition in the spectral region between 8 μm and 20 μm. For the thicker film the data was fitted to a linear relation with a correlation factor of 99.98%. The intercept is 0.0883 which yields $\sigma_{d.c} = 1.14 \times 10^{16} (\Omega\text{m})^{-1}$. The slope is 0.3896×10^{-43} units which yields a relaxation time $\tau = 3.36 \times \times 10^{-15}$s. Using the calculated values of τ and $\sigma_{d.c.}$, one will get N from the relation:

$$N = \sigma_{d.c} \frac{m}{e^2 \tau} = 1.34 \times 10^{22} \text{ cm}^{-3},$$

where m was taken as the electron rest mass. This value of N is in good agreement with the value of 1.3×10^{22} cm^{-3} calculated by Beattie et al.[3].

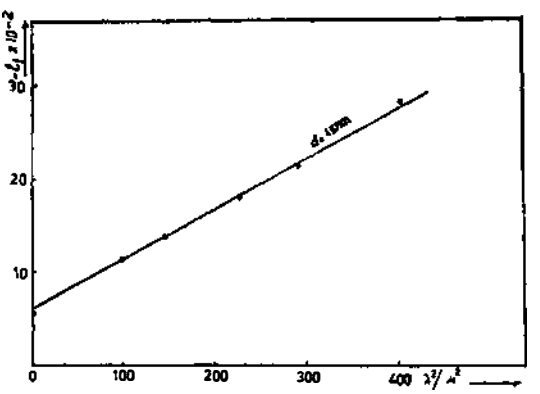

Fig. 7. The variation of ε_1 with the square of wavelength.

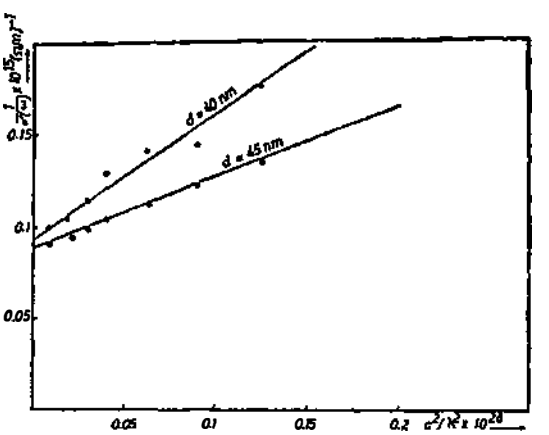

Fig. 8. The variation of $1/\sigma(\omega)$ with $(c/\lambda)^2$ in the I. R. region on KBr substrate.

As a matter of fact if m is replaced by m^*, the effective mass of the electron, N will increase by a factor of $\frac{m^*}{m}$ which approximates to 3.4 — 3.5. Using this value for m, the value of N will jump to $4.6 - 4.7 \times 10^{22}\,\mathrm{cm}^{-3}$. This value is in close agreement with the value of $5.5 \times 10^{22}\,\mathrm{cm}^{-3}$ estimated from the data of Manfred et al.[17] using the magnetic moment value of 0.6 electron per atom.

The last figure shows as well the previous relation for a film 40 nm thick. The data are scattered about the line because the film is thinner and the condenser effect may be important in the film of island structure as it was confirmed by electron microscopy. However the calculations yield values of

$$\sigma_{d.c} = 0.52 \times 10^{16}\,(\Omega\mathrm{m})^{-1}.$$

This value is reasonable compared with $1.4 \times 10^{16}\,(\Omega\mathrm{m})^{-1}$ units for the thicker film 45 nm thick. Accordingly, N was estimated to be $0.5 \times 10^{22}\,\mathrm{cm}^{-3}$ using the electron rest mass. The agreement between the values of N and $\sigma_{d.c}$ obtained by using different approaches is good.

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MJERENJA OPTIČKIH KONSTANTI NIKLA U INFRACRVENOM PODRUČJU

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U radu se proučavaju optička svojstva filmova nikla nataloženih na razne podloge pod različitim uvjetima, i nalaze se vrijednosti za n i k. Proučavaju se promjene realnih i imaginarnih dijelova dielektrične konstante i optičke vodljivosti. Opažene strukture su objašnjene pomoću prijelaza među vrpcama na višim energijama. Na energijama manjim od 0.18 eV mehanizam relaksacije i ovisnost ε_1 o λ^2 ukazuje da je međuvrpčano ubrzanje vodljivih elektrona važnije od prijelaza među vrpcama.