

## THE OPTICAL PROPERTIES OF VACUUM EVAPORATED TIN IN THE VISIBLE REGION AT ROOM-TEMPERATURE

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Received 18 March 1985

UDC 538.958

Original scientific paper

The values of the refractive index  $n$  and the extinction coefficient  $k$  have been measured for evaporated films of tin at wavelengths between 300 to 800 nm by transmission methods at room temperature. The results are compared with those obtained by previous workers at shorter wavelength, mainly in the visible region. The Drude parameters were derived from the real part of the dielectric constant giving  $\tau = 14.99 \times 10^{-16}$  s for the relaxation time.

### 1. Introduction

It is well known that thin metallic layers are formations consisting of an accumulation of fine grains separated by gaps. These gaps frequently play a decisive role in the optical and electrical properties of such layers.

Optical constants for tin in the energy range 0.1 to 1.3 eV have been determined by Hodgson<sup>1)</sup> and by Golovashkin et al.<sup>2)</sup>. Both used evaporated tin films which were prepared outside, and then transferred into the reflectometer. Lenham et al.<sup>3)</sup> have determined  $\varepsilon_1(E)$  and  $\varepsilon_2(E)$  for the principal directions in crystalline tin from below 0.1 to 3.1 eV. However, it is found that the values of  $n(E)$  and  $k(E)$  calculated from their  $\varepsilon_1(E)$  and  $\varepsilon_2(E)$  do not agree with those for evaporated tin films. From 7.5 to 27.5 eV, Walker et al.<sup>4)</sup> have measured the reflectance at 20° angle of incidence from a glass backed tin film evaporated in situ, but the optical constants were not determined from these data. Hunter<sup>5)</sup> has obtained values of  $k(E)$  from 14.5 to 27.5 eV from transmission measurements made on tin films. However, Macrae et al.<sup>6)</sup> calculated the real and imaginary parts,  $n(E)$  and  $k(E)$ ,

respectively, of the energy-dependent complex refractive index over the energy range 0.1 to 27.5 eV. Finally, the optical constants of liquid tin at 261°C have been deduced from ellipsometric measurements for photon energies between 0.62 and 3.7 eV<sup>7)</sup>.

### 2. Experimental techniques

High purity tin 99.999% was evaporated from a molybdenum boat onto a fused silica substrate in a vacuum of approximately  $10^{-4}$  Pa at an evaporation rate of about 3 nm/s. The optical constants of these films were determined from the absorption curves obtained on a recording double beam spectrophotometer (Beckman 5260) in the range from 300 to 800 nm with accuracy to about 1%. Film thickness was measured by Tolansky interference method<sup>8)</sup>.

### 3. Results and discussion

If, as a first approximation, a thin metallic film is considered to be homogeneous and isotropic, and bounded by two plane-parallel faces, the optical properties of the film can be characterized by three parameters: the thickness  $d$ , the index of refraction  $n$ , and the index of extinction  $k$ . The later two properties are usually combined in the form of a complex index,  $\tilde{n} = n - i k$ . The measurement of the optical constants  $n$  and  $k$  of thin absorbing films has, for a long time, presented serious difficulties<sup>9)</sup>.

The optical absorption spectra of thin vacuum deposited tin films on a fused-silica substrate have been measured at room temperature. The spectral variation of absorbance ( $\log_{10}(1/\text{transmittance})$ ) for a thin films of tin of about 12.5, 16, 20 and 25 nm thick are shown in Fig. 1. The absorbance is measured from an arbitrary zero at 2.0 eV. In a region of intrinsic absorption, where multiple reflection are not important, the absorbance is proportional to the absorption coefficient. The

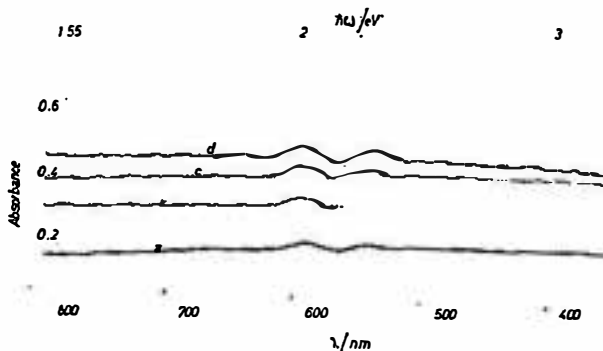


Fig. 1. The spectral variation of the absorbance ( $\log_{10}(1/\text{transmittance})$ ) for a thin film of tin.  
 (a)  $d = 12.5$  nm. (b)  $d = 16$  nm.  
 (c)  $d = 20$  nm. (d)  $d = 25$  nm.

absorbance in Fig. 1 decreases gradually as the wavelength increases, however, the absorbance increases with increasing film thickness.

Fig. 2 shows the variation of the calculated values of the real and imaginary part of the refractive index with the photon energy for layer of different thicknesses. The points for  $n$  and  $k$  obtained by the use of absorbance spectra are shown in the graph. As is seen from Fig. 2, the extinction coefficient  $k$  decreases with the increase of photon energy and thickness, but the refractive index  $n$  increases with the increase of photon energy and thickness.

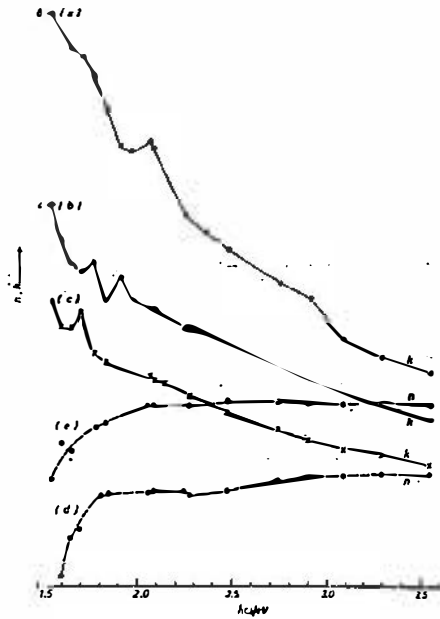


Fig. 2. The dependence of the extinction coefficient ( $k$ ) and refractive index ( $n$ ) on the photon energy.

- (a)  $d = 16$  nm.
- (b)  $d = 20$  nm.
- (c)  $d = 25$  nm.
- (d)  $d = 20$  nm.
- (e)  $d = 25$  nm.

Tin belongs to the fourth group in the periodic table and has four valence electrons per atom: two  $s$ -electrons and two  $p$ -electrons. Obviously, the  $p$ -electrons are on the Fermi surface and the internal photoeffect is due to the transfer of the  $s$ -electrons to the Fermi level.

To determine the microproperties related to free electrons, we used the wavelength range 300—800 nm. At shorter wavelengths, the optical constants were considerably affected by the internal photoeffect. The determination of the absorption coefficient for normal incidence  $A = 4n/[(n + 1)^2 + k^2]$  indicated the presence

of a sharp peak. In Fig. 3 is presented the dependence  $A = f(\lambda)$  for tin. From the position of the maximum  $A$  the quantity of the energy gap between bands, which corresponds to the quantum transition

$$\Delta E = \frac{hc}{\lambda}$$

where  $h$  is the Planck's constant,  $c$  is the speed of light and  $\lambda$  is the wavelength of the maximum absorption<sup>10)</sup>. The value of  $\Delta E$  equals 2.084, 2.275 eV.

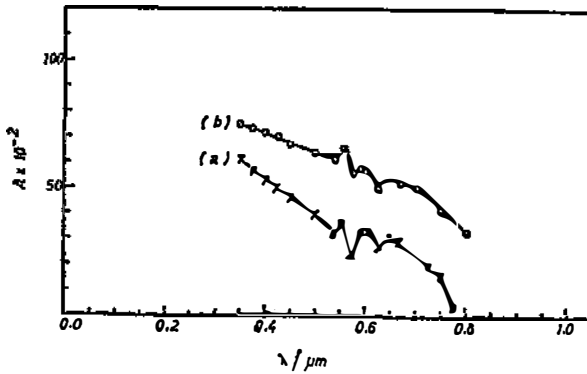


Fig. 3. Dependence of the absorption coefficient.

$$A = 4n[(n + 1)^2 + k^2] \text{ on } \lambda:$$

- (a)  $d = 20 \text{ nm}$ . (b)  $d = 25 \text{ nm}$ .

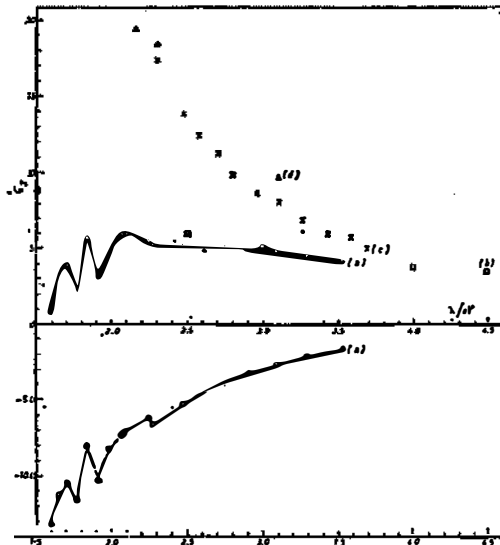


Fig. 4. Dielectric constants of tin as a function of incident photon energy.

- (a) present work (b) from Ref. 12  
(c) from Ref. 7 (d) from Ref. 6

Weisz<sup>11)</sup> has calculated the band structure for white tin without spin-orbit effects and then found spin-orbit corrections to this band structure at a few symmetry points in the Brillouin zone. Of the allowed direct transitions,  $V_4-V_3$  at 2.0 eV is the closest in energy to our observed interband transition at 2.084 eV. Furthermore on the basis of phase arguments, the transition has a relatively high probability. The real part  $\epsilon_1$  of the dielectric constant and the optical conductivity  $\sigma_1 = \epsilon_2 \omega / 4\pi$  are plotted in Fig. 4 together with the values of Macrae et al.<sup>6)</sup>, Kent<sup>12)</sup> and Petrakian et al.<sup>7)</sup>. For energies greater than 2.48 eV the deviations are too small to be shown. Difference in the degree of contamination of the surfaces studied by the various workers might account for some of the variation in the results. It has been shown both theoretically<sup>13)</sup> and experimentally<sup>14)</sup> that the presence of a dielectric layer on a metallic surface can reduce the apparent optical constant.

The Drude values for the present data were calculated from the equations

$$\epsilon_1(\omega) = 1 - \frac{4\pi e^2 n^*}{m} \frac{\tau^2}{1 + \omega^2 \tau^2} \quad (1)$$

and

$$\sigma(\omega) = \frac{e^2 n^*}{m} \frac{\omega}{1 + \omega^2 \tau^2}, \quad (2)$$

where  $\omega$  is the angular frequency of the incident light. The relaxation time  $\tau$  and the ratio of the effective density of free carries to the free electron mass  $n^*/m$  were deduced from the experimental values of  $\sigma_1$  by rearranging (1) as

$$(1 - \epsilon_1(\lambda))^{-1} = \left( \frac{\pi c^2 m}{e^2 n^*} \right) \frac{1}{\lambda^2} + \frac{m}{4\pi e^2 \tau^2 n^*} \quad (3)$$

where  $\lambda$  and  $c$  are the wavelength and velocity of the incident light, respectively, and then performing a least squares fit of the data to the linear relationship between  $(1 - \epsilon_1)^{-1}$  and  $1/\lambda^2$ . An excellent fit (correlation coefficient  $p^2 = 0.995$ ) was obtained, as shown in Fig. 5. The value of  $n^*/m$  was deduced from the slope of the line, and  $\tau$  was obtained from the intercept and equal to  $14.99 \times 10^{-16}$ s.

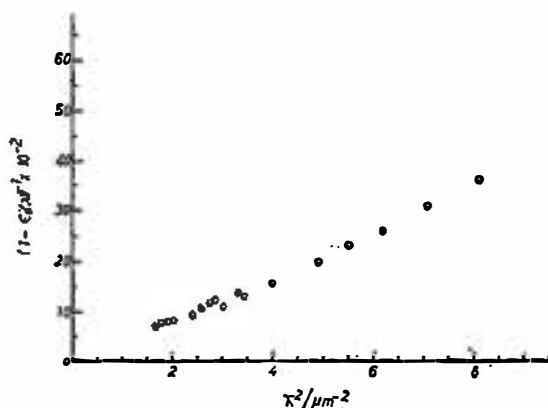


Fig. 5. The quantity  $(1 - \epsilon_1)^{-1}$  as a function of  $\lambda^{-2}$  for thin film of tin at room temperature, where  $\lambda$  is the wavelength of incident photons.

It is apparent in Table 1 that the relaxation time  $\tau$  in thin film is approximately an order of magnitude smaller than in the solid and large from liquid tin<sup>15)</sup>. In addition,  $n^*/n_v$  is smaller than in the solid.

TABLE 1.

	Temperature (°C)	Energy range	$\tau$ ( $10^{-16}$ s)	$n^*/n_v$
Present work	30	vis	14.99	0.192
Liquid tin				
Petrakian, Ref. 7 (1980)	261	uv-ir	4.90	$1.02 \pm 0.05$
Hodgson, Ref. 15 (1961)	446	vis-ir	3.90	1.17
	858	vis-ir	3.52	1.15
Comins, Ref. 16 (1972)	600	ir	4.11	$1.10 \pm 0.02$
	800	ir	3.88	$1.05 \pm 0.02$
Solid tin				
Hodgson, Ref. 15 (1961)	20	vis-ir	37.4	0.33
Golovashkin, Ref. 2 (1965)	20	ir	44.2	0.32
Macrae, Ref. 6 (1967)	20	uv-ir	42.0	0.325

Comparison between some properties of thin film, liquid and solid tin.

Comparison of the present results for the evaporated films of tin with those of previous workers is difficult because of the wide range of experimental conditions in the various studies.

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OPTIČKA SVOJSTVA VAKUUMSKI NAPAREN OG KOSITRA U  
VIDLJIVOM PODRUČJU PRI SOBNOJ TEMPERATURI

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UDK 538.95

Originalni znanstveni rad

Mjereni su realni i imaginarni dio indeksa loma vakuumski naparenih filmova kositra pomoću transmision e metode na sobnoj temperaturi i u području valnih duljina između 300 i 800 nm. Rezultati su uspoređeni sa onima dobivenim za kraće valne duljine, uglavnom u vidljivom području. Dobiveni su Drudeovi parametri iz realnog dijela dielektrične konstante što daje za relaksaciono vrijeme  $\tau = 14.99 \times 10^{-16}$  s.