SOVIET PHYSICS JETP VOLUME 22, NUMBER 5 MAY, 1966

THE EFFECT OF PERIODIC STRUCTURE ON THE OPTICAL PROPERTIES OF ALUMINUM

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Submitted to JETP editor June 12, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) 49, 1431-1434 (November, 1965)

The optical constants n and κ ($n - i\kappa$ being the complex index of refraction) of crystalline and amorphous aluminum have been measured in the infrared region. It was found that with the destruction of the periodic structure the number of conduction electrons, as determined optically, varies from one electron per atom to three electrons per atom.

1. The structure of a crystal lattice has a considerable influence upon the optical properties of a metal. This has been shown by measurements of the optical properties of a number of multivalent metals in the crystalline and liquid states (see, for example, [1-3]). In this study we set out to investigate the optical constants of crystalline and amorphous aluminum. In both cases the investigated layers were prepared by evaporation of 99.99% pure aluminum in vacuo from a tungsten wire onto a glass substrate. When sufficiently thick layers were deposited rapidly and then annealed in vacuum to a temperature of 300°C over a period of several hours, we obtained polycrystalline samples with a density $\rho = 2.7\ \mathrm{g/cm^3}$, corresponding to the density of the solid metal. In order to study the optical properties of these samples, we reflected light from the outer aluminum-air interface. By slowly depositing thin layers on glass having an amorphous structure, we obtained samples with a mean density of $2.2\ \mathrm{g/cm^3}$, somewhat less than the density of the liquid metal, $2.4\ \mathrm{g/cm^3}$. To measure the optical constants of these samples, we reflected light from the inner glass-aluminum interface.

We have measured n and κ ($n - i\kappa$ being the complex index of refraction) in the infrared region of the spectrum. The measurements were accomplished by a polarization technique, using fourfold reflection of light from the studied surface. (For a description of this technique see [4]).

The optical constants of polycrystalline samples of aluminum were also investigated in [5]. However, unlike the present study, the layers examined were not annealed. The effect of annealing upon the optical properties of various metals was studied in [6,7]. In the case of metals with a high melting temperature, the annealing process has a significant effect upon the optical constants. For Al the effect of annealing upon the optical constants is noticeable, though not too great.

Table I. Optical constants of annealed polycrystalline aluminum layers

λ, μ	n	$\varkappa$	$N_{\mathrm{opt}}^{\mathrm{cryst}} \cdot 10^{-22}$, $\mathrm{cm^{-3}}$	$\nu_{\mathrm{eff}}^{\mathrm{cryst}} \cdot 10^{-14}$, $\mathrm{sec^{-1}}$
1.00	0.98	7.65	6.86	4.71
1.50	1.14	11.6	6.87	2.29
2.00	1.67	15.2	6.69	1.90
2.50	2.50	18.8	6.67	1.85
3.00	3.48	22.6	6.82	1.79
4.00	5.58	29.4	6.75	1.67
5.00	7.84	35.7	6.62	1.56
6.00	10.4	41.3	6.46	1.52
8.00	16.2	52.2	6.44	1.47
10.0	25.5	60.9	7.17	1.81

Table II. Optical constants of amorphous aluminum layers

λ, μ	n	$\varkappa$	$N_{\mathrm{opt}}^{\mathrm{amor}} \cdot 10^{-22}$, $\mathrm{cm^{-3}}$	$\nu_{\mathrm{eff}}^{\mathrm{amor}} \cdot 10^{-14}$, $\mathrm{sec^{-1}}$
0.96	2.0	10.2	14.2	7.9
1.15	1.8	12.0	13.1	5.0
1.46	3.1	16.4	15.7	5.0
1.82	4.9	19.9	16.0	5.4

The optical properties of aluminum on the inner glass-aluminum interface were also studied in [8]. However, the measurements in that study were carried out in the visible portion of the spectrum. Since we are interested in those optical properties which are determined by the conduction electrons, it is necessary for us to carry out our measurements in the infrared region, where the energy of a light quantum is less than the threshold energy of the internal photoeffect.

2. The results of our measurements of the optical constants of crystalline and amorphous aluminum are shown in Tables I and II (λ is the wavelength of the light). This experiment has shown that the skin-effect exhibits a slightly anomalous character in crystalline aluminum at room temperature. Therefore, analysis of the results of these measurements was with the formulas obtained in[9] [Eqs. (11)—(14)]. The skin-effect in amorphous aluminum is almost normal (the correction terms

in Eqs. (11)—(14) of [9] are less than the experimental error). Results of the analysis of the experimental data are given in these same tables where the concentration of conduction electrons obtained by the optical measurements, $N_{\rm opt}$, and the effective collisions frequency of the electrons $\nu_{\rm eff}$ are given in the last two columns. Both of these quantities are related to the complex dielectric constant $\epsilon = (n - i\kappa)^2$ in the following manner:

a) in the case of normal skin effect for the infrared spectral region

$$\varepsilon \approx -\frac{4\pi e^2}{\omega(\omega - i\nu_{\rm eff})}\frac{N_{\rm opt}}{m}; \qquad (1)$$

b) in the case of weak anomalous skin effect for the infrared spectral region

$$\varepsilon \approx -\frac{4\pi e^2}{\omega(\omega - i\nu_{\rm eff})}\frac{N_{\rm opt}}{m}(1-\gamma). \qquad (2)$$

In both cases it is assured that $|\epsilon| \gg 1$, which is well satisfied for aluminum. Here m is the mass of the free electron, e the charge of the electron, ω the angular frequency of the radiation, and γ a correction term 1) determined from Eqs. (9)—(10) of [9].

It follows from Table I that annealed layers of aluminum have $N_{\rm opt}^{\rm cryst} = (6.74 \pm 0.1) \times 10^{22}$ cm^{-3}. If we take into account the fact that the density of these layers $\rho_{\rm cryst} = 2.70 \pm 0.16$ g/cm^3 and thus the concentration of atoms $N_{\rm a}^{\rm cryst} = (6.03 \pm 0.36) \times 10^{22}$ cm^{-3}, we find that the number of conduction electrons per atom is equal to

$$N_{\rm opt}^{\rm cryst}/N_{\rm a} = 1.12 \pm 0.07.$$

Not only does this ratio not increase upon annealing, but it even decreases somewhat (see the results of [5]). It is considerably less than the number of valence electrons per atom, which is equal to three.

From Table II it follows that for amorphous aluminum layers $N_{\rm opt}^{\rm amor} = (14.8 \pm 1.1) \times 10^{22}$ cm^{-3}. The density of these layers is $\rho_{\rm amor} = 2.2 \pm 0.2$ gm/cm^3, giving for the concentration of atoms the magnitude $N_{\rm a}^{\rm amor} = (4.9 \pm 0.45) \times 10^{22}$ cm^{-3}. From this the number of conduction electrons per atom is equal to $N_{\rm opt}^{\rm amor}/N_{\rm a}^{\rm amor} = 3.0 \pm 0.5$, agreeing with the number of valence electrons.

Thus, during the transition from crystalline aluminum layers to amorphous layers the concentration of conduction electrons changes from approximately one electron per atom to three electrons per atom. The effective frequency of electron collisions also changes considerably, increasing during the transition to the amorphous state. The important point is that the mean free path $l = v/\nu_{\rm eff}$ for the layers under consideration is equal to 6×10^{-7} cm for crystalline aluminum and 2×10^{-7} cm for amorphous aluminum, being thus 15 and 5 times greater than the interatomic distance. Similar variations are observed during the transition from the crystalline to the liquid state for tin and lead (see [1-3]). In all cases of multivalent metals mentioned, when the periodic structure is disrupted the number of conduction electrons, as determined by optical means, agrees with the number of valence electrons. When a periodic structure does exist, this number decreases to approximately one electron per atom. This strong effect of a periodic structure upon the optical properties of multivalent metals can be explained by the difference in the interelectron interaction in the crystalline and amorphous states. This topic is discussed further in [11].

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1) We used the value of the electron velocity on the Fermi surface, $v = 1.1 \times 10^8$ cm/sec, in calculating γ of Eq. (2). This value was obtained from the value of the electron specific heat $C_e = 1.27 \times 10^{-3}$ T (Joule/g-mole-deg) cited in [10], and from the value of $N_{\rm opt} = 6.74 \times 10^{22}$ cm^{-3} deduced in the present work. In this case the correction term for $N_{\rm opt}$ is about 3% in the 10 μ and about 4% in the 1 μ region. The smallness of the correction terms attests to the slight anomaly of the skin effect.

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Translated by L. Marquet
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