DETERMINATION OF MICROSCOPIC PARAMETERS OF ALUMINUM FROM ITS OPTICAL CONSTANTS AND ELECTRIC CONDUCTIVITY

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The optical constants of aluminum were measured in the $0.8 - 9\,\mu$ range at room temperature and at liquid-nitrogen temperature. The static conductivity and density of the same specimens were also measured. The data obtained were evaluated by using a theory based on the quantum kinetic equations, which takes into account the electron-electron collisions. The concentration of the conduction electrons, the electron velocity on the Fermi surface, and the electron collision frequency were determined.

IN the present investigation, which should be considered as a continuation of our research on metal optics,[1-3] we measured the optical constants of aluminum in the spectral region $0.8 - 9\,\mu$ at room temperature and at liquid-nitrogen temperature.

The present status of the theory, which relates the microscopic parameters of the metals with their optical constants,[4-6] calls for accounting for the electron-electron collisions and quantum corrections for the frequency of electron-phonon collisions. The method for reduction of the experimental data is detailed in a paper by one of the authors.[7] According to this method, the measurements of the optical constants must be supplemented by measurements of the specific static conductivity σ_0 at various temperatures.

The optical constants were determined at room temperature by the polarization method, described previously,[1] and at liquid-nitrogen temperature with the apparatus described in reference 8. Measurements were made on an aluminum layer obtained by evaporation in vacuum from a tungsten helix on polished glass. The initial aluminum was 99.99% pure. The measurements were made on mirrors with external reflection within the first few hours after sputtering. Specimens for the measurement of σ_0 and the density ρ were sputtered simultaneously with the mirrors.*

In measuring the quantities σ_0 and ρ, the thickness of the layer was determined by an interference method accurate to 1.5×10^{-6} cm. It has been found that by evaporation in vacuum it is easy to obtain aluminum layers with $\sigma_0 \approx 1.6 \times 10^{17}$ cgs esu* and $\rho \approx 2.0$ g/cm^3. With special choice of the distance from the sputterer to the specimen, of the temperature, and of the sputtering rate, it is possible to obtain mirrors with $\sigma_0 \approx 2.2 \times 10^{17}$ cgs esu and $\rho \approx 2.4$ g/cm^3. For bulk metal $\sigma_0 = 3.1 \times 10^{17}$ cgs esu and $\rho = 2.7$ g/cm^3.

The measurement of the optical constants of the mirrors with different σ_0 and ρ have shown that a reduction in σ_0 and ρ leads to a reduction in κ and to an increase in n, where $n - i\kappa$ is the complex index of refraction; the change in κ is small, but the change of n is considerable.†

Table I shows the results obtained for the optical constants of aluminum mirrors, characterized by values $\sigma_0 = 2.2 \times 10^{17}$ cgs esu and $\rho = 2.4$ g/cm^3. The table also lists, for comparison, the data of Beattie,[9] obtained at room temperature. A comparison of these data shows that our values of n are lower than those of Beattie. Apparently this discrepancy is explained by the lower value of the conductivity of the mirrors used by Beattie ($\sigma_0 = 1.5 \times 10^{17}$ cgs esu). A comparison of our values of κ with those of Beattie[9] shows that these data are in good agreement up to $5\,\mu$. In the further region Beattie's values of κ are somewhat lower, and they still can be explained by the lower values of the conductivity. Unfortunately we could not

*The density ρ does not enter into the calculation formulas. It is needed, however, for evaluating the number of conduction electrons per atom.

*The low value of σ_0 is not due to the small thickness of the aluminum layers. The layers used for the measurements were of the order of 0.45μ, which is 30 times greater than the mean free path of the electrons. In all probability, this is due to the more finely dispersed structure of the sputtered layers compared with the ordinary polycrystalline materials.

†Aluminum exhibits an anomalous effect in the region of the spectrum we have investigated. In this case n and $\varkappa$ must be taken to mean their effective values;[3] we shall leave out the subscripts "eff."

TABLE I. Optical constants of aluminum*

λ_μ	n			$\varkappa$			$10^2 \cdot 4n/(n^2+\varkappa^2)$		$10^2 \cdot 4\varkappa/(n^2+\varkappa^2)$		$\lambda \cdot 4\varkappa/(n^2+\varkappa^2)$	
	T=295		78	295		78	T=295	78	295	78	295	78
	P.W.	B	P.W.	P.W.	B	P.W.						
0.8	1.12	—	0.83	6.0	—	6.0	12.0	10.9	64.2	65.5	0.514	0.524
0.9	1.05	—	0.75	7.0	—	7.0	8.4	6.0	55.8	56.4	0.504	0.507
1.2	0.95	—	0.63	9.6	—	9.6	4.09	2.72	41.4	41.5	0.495	0.500
1.5	1.14	—	0.78	12.1	—	12.1	3.09	2.12	32.8	32.9	0.492	0.494
2.0	1.75	2.30	1.30	16.1	16.5	16.1	2.67	2.00	24.5	24.6	0.490	0.492
2.5	2.4	3.22	1.7	19.8	20.3	19.8	2.41	1.72	19.9	20.0	0.497	0.500
3.0	3.2	4.41	2.2	23.5	24.2	23.5	2.28	1.58	16.7	16.8	0.500	0.506
4.0	4.8	5.97	3.2	30.0	30.3	30.1	2.08	1.40	13.0	13.1_5	0.520	0.526
5.0	6.7	8.19	4.4	37.6	36.8	37.8	1.84	1.22	10.3_5	10.4	0.520	0.520
6.0	9.5	11.0	6.5	44.4	42.4	44.9	1.84	1.26	8.6	8.7	0.516	0.522
7.0	12.6	14.6	9.1	51.0	49.0	52.0	1.83	1.31	7.4	7.5	0.518	0.524
8.0	15.6	17.0	—	58.1	55.0	—	1.73	—	6.4	—	0.515	—
9.0	21.1	21.1	—	62.1	61.3	—	1.95	—	5.7_5	—	0.517	

*The symbols used here are: λ – wavelength of light in microns, P.W. – present work, B – data of Beattie.[9] The temperature is in degrees Kelvin.

make an analogous comparison with the results of Hodgson,[10] since it does not contain numerical values of n and κ.

A comparison of the data at room temperature and temperature of liquid nitrogen shows, as expected, that κ remains almost constant with temperature, whereas n is substantially reduced.

The reduction of the experimental data, obtained both at room temperature and at liquid-nitrogen temperature, was based on formulas (3) and (11) of reference 7 and the method of successive approximations. It was found here that the third approximation differed from the second by less than 1%.

TABLE II. Microscopic characteristics of aluminum

T, °K	295	78
$N \cdot 10^{-22}$, cm^{-3}	7.4	7.1
N/N_a	1.3_8	1.3_3
$\nu_0^{ee} \cdot 10^{-12}$, sec^{-1}	3.8	0.19
$\nu_0^{ef} \cdot 10^{-13}$, sec^{-1}	6.3	1.2
$\nu^{ef} \cdot 10^{-13}$, sec^{-1}	7.7	3.9
$\nu^{imp} \cdot 10^{-13}$, sec^{-1}	1.3	1.3
$v \cdot 10^{-8}$, cm/sec	2.8	2.5

The results of the reduction of our experimental data are listed in Table II. Here N is the concentration of the conduction electrons, N_a the concentration of the aluminum atoms, ν_0^{ee} the classical frequency of the electron-electron collisions for $\hbar\omega \ll kT$, ν_0^{ef} the classical frequency of the electron-phonon collisions for $\hbar\omega \ll kT$, ν^{ef} the frequency of collisions between electrons and phonons for the near infrared region, ν^{imp} the frequency of electron-impurity collisions, and v the electron velocity on the Fermi surface.

According to the results obtained by Gurzhi[4] $\nu^{ef} = \nu_0^{ef}\varphi(T)$. The function $\varphi(T)$ was also calculated by Gurzhi.[11] For aluminum, the Debye temperature is $\Theta = 398°K$,[12] and therefore $\varphi(T) = 1.22$ when $T = 295°K$. When $T = 78°K$, the value of ν^{ef} reaches its practical limit, 1.2×10^{13} sec^{-1}. Since a further reduction in temperature does not change ν^{ef} for aluminum, we did not deem it necessary to perform measurements at lower temperatures.

The error in the determination of microscopic characteristics of aluminum was 5% for N, 10% for ν_0^{ee}, 30% for v at room temperature, and 20% for v at liquid-nitrogen temperature. The great error in v is due to the fact that aluminum is subject to a weakly pronounced anomalous skin effect and the role of surface losses is small compared with that of volume losses. It must be noted that v should be determined from the value of σ_0 of the investigated mirrors. If the tabulated values are used, the resultant values of v are greatly overestimated.*

Comparison of the results for the microscopic parameters of aluminum, obtained at room temperature and at liquid-nitrogen temperature, shows that N and v hardly change with temperature. The temperature variation of ν_0^{ee} is much faster than the theoretically expected dependence, which is proportional to T^2.

As shown by Gurzhi,[5] the frequency of electron-electron collisions for a frequency ω is

$$\nu^{ee} = \nu_0^{ee}[1 + (\hbar\omega/2\pi kT)^2],$$

*In our earlier investigations[1,2] we did not measure the σ_0 of the sputtered specimens, but used the tabulated values. The values of v given in reference 2 are therefore overestimated.

and therefore the effective electron-collision frequency $\nu_{eff} = \nu^{ef} + \nu^{ee} + \nu^{imp}$ depends on the wavelength of the incident light λ. For $\lambda \approx 5\mu$ we have $\nu_{eff} \approx 0.9 \times 10^{14}$ sec^{-1} at T = 295°K and $\nu_{eff} \approx 0.6 \times 10^{14}$ sec^{-1} at T = 78°K.

Using the values obtained for ν_{eff} and v, we can estimate the mean free path of the electron l and the relaxation time τ.

For T = 295°K

$$l = v/\nu_{eff} \approx 3 \cdot 10^{-6} \text{ cm}, \quad \tau = 1/\nu_{eff} \approx 1.1 \cdot 10^{-14} \text{ sec},$$

for T = 78°K

$$l \approx 4 \cdot 10^{-6} \text{cm}, \qquad \tau \approx 1.7 \cdot 10^{-14} \text{ sec}.$$

The depth of the skin layer for aluminum, in the wavelength interval used by us, is $\delta = \lambda/2\pi\kappa \approx 2 \times 10^{-6}$ cm for both temperatures. We see that $l \sim \delta$, i.e., at both temperatures there is a weakly pronounced anomalous skin effect for aluminum.*

The expressions we employed to relate the optical constants of the metal with its microscopic parameters hold if the inequalities $v/\omega\delta \ll 1$, $\nu_{eff}/\omega \ll 1$, and $\omega \ll \omega_0$ are satisfied, where ω_0 is the limit of the internal photoeffect. That the first two inequalities are satisfied is readily verified. For $\lambda = 5\mu$ we have $v/\omega\delta \approx 0.37$ and $\nu_{eff}/\omega \approx 0.24$ at T = 295°K; and $v/\omega\delta \approx 0.33$ and $\nu_{eff}/\omega \approx 0.16$ at T = 78°K. That the third inequality is satisfied can be verified from the character of the λ-dependence of the quantities

$$N = 1{,}79 \cdot 10^{22} (1 + p^2 G)^2 f \lambda^2 \left(\frac{4\varkappa}{n^2 + \varkappa^2}\right)^2,$$

$$M = \frac{4n}{n^2 + \varkappa^2} + p^2 \beta D.$$

Figures 1 and 2 show the dependence of these quantities on λ in the interval $1.2 - 9\mu$ (the notation of reference 7 is used here). It is seen from the figures that N is independent of λ, and that the dependence of M in this interval coincides with the calculated curve, obtained when the electron-electron collisions are taken into account.

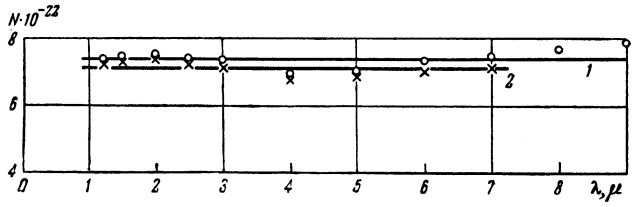

FIG. 1. Plot of N(λ): o – for T = 295°K, × – for T = 78°K.

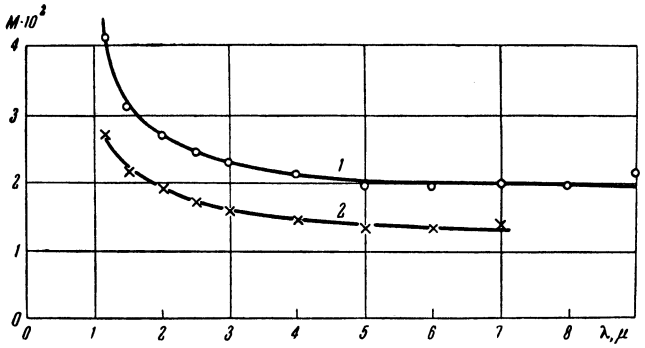

FIG. 2. Plot of M(λ): o – for T = 295°K, × – for T = 78°K.

We can therefore say that the measurements were carried out in the region where the third inequality is also satisfied.*

In the aforementioned paper by Beattie[9] no account was taken of the theory that allows for electron-electron collisions in the processing of the experimental data. Therefore Beattie reaches the erroneous conclusion that aluminum has in the region $2 - 5\mu$ an additional absorption, connected with the internal photoeffect. This conclusion has led Beattie to determine the microscopic parameters only from the results obtained in the $6 - 12\mu$ region. In this region the correction terms already become substantial: at $10 - 12\mu$ the correction terms reach 20 – 30% (the values of $v/\omega\delta$ and ν_{eff}/ω are close to unity). Therefore the accuracy of the corresponding formulas is reduced. Our own measurements and their reduction with allowance for the electron-electron collisions show that there are no additional absorption sources in this region, with the exception of those indicated above.

The value of ν_0^{ee} given above was obtained from optical measurements. This quantity cannot be determined with sufficient accuracy from the temperature dependence of σ_0, since the condition $T \gg \Theta$ cannot be realized.

Although there are no data at present on measurements of the electronic specific heat and surface impedance at radio frequencies, carried out on specimens identical with those used in the optical methods, there is nevertheless a certain interest in the comparison of the results known to us.

Measurements of the electron specific heat yields[13]

$$v/\sqrt{N} = (3{,}7 \cdot 10^{-3} - 4{,}1 \cdot 10^{-3}) \text{ cm}^{5/2} \text{ sec}^{-1}.$$

From optical measurements we obtain

$$v/\sqrt{N} = 9{,}8 \cdot 10^{-3} \text{ cm}^{5/2} \text{ sec}^{-1}.$$

*The values of v and ν^{imp} allow us to estimate the dimensions of the individual crystals of aluminum, obtained in thermal evaporation of the metal: $L \approx v\nu^{imp} \approx 3 \times 10^{-6}$ cm.

*As can be seen from Table I, M increases sharply in the region 0.8 – 0.9 μ. The corresponding absorption band is probably connected with the internal photoeffect.

Measurements of the surface impedance in the radio-frequency range yield[14]

$$v/N = 4.5 \cdot 10^{-15} \text{ cm}^4 \text{ sec}^{-1}.$$

Optical measurements for the same quantity yield

$$v/N = 3.7 \cdot 10^{-15} \text{cm}^4 \text{ sec}^{-1}.$$

In conclusion, the authors are deeply grateful to I. L. Fabelinskiĭ for a discussion of the present work.

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