SOVIET PHYSICS JETP VOLUME 21, NUMBER 3 SEPTEMBER, 1965

OPTICAL PROPERTIES OF LEAD AT LOW TEMPERATURES

A. I. GOLOVASHKIN

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor October 15, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 48, 825-832 (March, 1965)

The optical constants of lead were measured at temperatures of 4.2, 78 and 293°K in the spectral region 0.9—12 μ. The temperature dependence of the electron-phonon collision frequency was determined. The experimental results were compared with the theory.

THE results of quantum-mechanical calculations[1-3] predict that at low temperatures the mechanism of the absorption of infrared radiation by metals differs from the mechanism of the absorption of long-wavelength radiation. The difference is particularly strong in the case $\hbar\omega \gg k\Theta \gg kT$; here, ω is the angular frequency of light, Θ is the Debye temperature, T is absolute temperature, $\hbar$ is Planck's constant, and k is Boltzmann's constant. In this case, an electron which has absorbed a high-energy quantum travels far outside the Fermi surface. Although such an electron cannot absorb high-energy phonons, which are absent, it may create phonons throughout the spectrum because it has high energy. Therefore, in contrast to weak classical absorption (which applies in the case when $\hbar\omega < kT$), the "quantum" absorption may reach a considerable value and give rise to the "residual" absorption, which is retained down to absolute zero.[3] (We note that we are dealing with the volume absorption, associated with the electron-phonon interaction.)

Earlier[4], the present author and Motulevich measured the temperature dependence of the electron-phonon collision frequency for tin (this quantity governs the volume absorption in pure metals) and found good agreement with the theoretical calculations.

In the present study, similar investigations were carried out on lead. Like tin, lead is very suitable for the verification of the predicted temperature dependence of the collision frequency. Our earlier[5] measurements of the optical constants of lead at T = 293°K and T = 78°K showed that the surface losses in this metal are small compared with the volume losses. Moreover, it is easy to prepare lead samples whose absorption associated with impurities and defects can be neglected.

In the present study, the optical constants of lead were measured at T = 4.2°K in the spectral region 0.9—12 μ. Since it was desirable to carry out an investigation of the optical constants over the whole range of temperatures under the same conditions, the measurements at T = 78°K and T = 293°K were repeated. Moreover, an investigation was made of some electrical and other properties of the samples. The Hall effect was measured, the temperature of the transition to the superconducting state was determined, the conductivity was measured in the temperature range 7—20°K, and the measurements of the conductivity and the density were repeated at higher temperatures.

1. The method of measuring the optical constants, the arrangement of the apparatus and the experimental details were fully described earlier.[4] The same method was used in the present study, practically without any modifications. It should be mentioned, however, that the optical arrangement used differed from that used in the earlier study of the optical properties of lead.[5] In particular, in the present investigation, we eliminated the stray signal associated with the temperature drop between the test mirrors and other parts of the apparatus. Moreover, it was not necessary to allow for the polarization introduced by the monochromator, and this made it easier to obtain and analyze the data.

The investigation was carried out on lead films prepared by evaporation in vacuum onto polished glass. The lead used had a purity of 99.999%. The thickness of the investigated films was 0.6—1.2 μ. The method of preparing the samples was described in[5].

The conductivity, density and the Hall effect were measured on the samples. The method of preparing the samples for these measurements and the measurements themselves were described in detail earlier.[5,6] The superconducting properties of lead were investigated on samples located in helium vapor in a special chamber fitted with a

Table I. Optical constants of lead

λ, μ	T = 293° K		T = 78° K		T = 4,2° K	
	n	ϰ	n	ϰ	n	ϰ
0.7	1.68	3,67	—	—	—	—
0,8	1.51	4.24	1,05	3.90	—	—
0,9	1.44	4,85	0,97	4.50	0,81	4.30
1,0	1.41	5,40	0,87	5,12	0,68	4,94
1.1	1.42	5.97	0,82	5,73	0,58	5,60
1,2	1,46	6.53	0.69	6.35	0.465	6,24
1,3	1,51	7.12	0.63	7,02	0.345	6,90
1,4	1.59	7,67	0,595	7.69	0,28	7.54
1.5	1,67	8,24	0,595	8,35	0,27	8.19
1,7	1.90	9,37	0,65	9,64	0,29	9.51
2,0	2.28	11.1	0,78	11.4	0,34	11,5
2.5	3,20	13.7	1.09	14,3	0,53	14,3
3,0	4.27	16,4	1.53	17,3	0.81	17.3
3,5	5.39	18,6	2,01	20.4	1,10	20.1
4,0	6.58	20.8	2.48	22,9	1,49	23,1
5.0	9.04	24.8	3,99	28,7	2.15	28,6
6.0	11,7	28,1	5.41	33.9	2.95	34,4
7,0	14,1	30.9	7,16	38,7	3.75	39,9
8,0	16.4	33,6	8.82	43,9	4,50	45.5
9.0	18.7	35,8	10,5	49,1	5,56	50,6
10,0	21.0	37.4	12,3	54,4	6.70	55,9
11,0	23,2	39.2	14.4	59,1	7.90	61,3
12.0	24,6	40.5	16.3	63,5	9,20	66,5

heater to provide temperatures higher than 4.2°K. The temperature dependence of the resistance of lead was obtained near the critical temperature. From this dependence, the critical temperature and the range of temperatures in which the transition occurred were determined. Temperature was measured with a carbon thermometer.

2. The results of the optical measurements are presented in Table I and in Figs. 1 and 2. The values of the optical constants n and κ (n − iκ is the complex refractive index) were obtained by averaging the results of several series of measurements. The error in the determination of the optical constants amounted to 1—2% at room temperature and 2—4% at helium temperature.[1] The main error occurred in the averaging of the different series of measurements. The error in the determination of κ was considerably less than the error in the determination of n over the whole spectral range at all temperatures.

The optical constants n and κ, listed in Table I, were obtained allowing for the dependence of the surface impedance on the angle of incidence of light on a mirror. This dependence was allowed for using formulas (1)—(4) from[4].

The optical constants of lead obtained at room temperature agreed with the constants determined earlier by the present author and Motulevich.[5] [2] At nitrogen temperature, there was some discrepancy between the two sets of results at the long-wave end. This discrepancy was probably due to the insufficiently complete compensation of the stray signal in[5].

The results of the determinations of the other properties of the lead films are listed in Table II, which includes the tabulated values for the bulk metal. The first row lists the film thickness d. For this range of thicknesses, the investigated parameters did not depend on the thickness. Other rows in Table 2 list the density ρ, and the conductivity σ_0 (at T = 293°K), σ_N (at T = 78°K) and σ_H (at T = 20.3°K). The accuracy of the determination of these quantities was 3—4%. The resistance

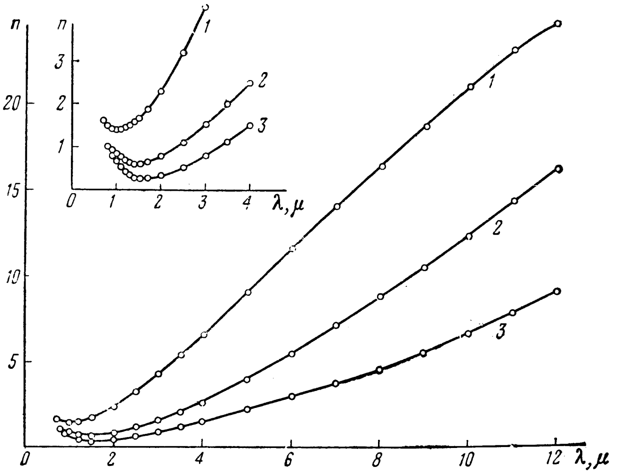

FIG. 1. Dependence of the refractive index of lead on the wavelength at temperatures of: 1) 293°K; 2) 78°K; 3) 4.2°K.

---

[1] The maximum error in the determination of n at helium temperatures was 5% at λ ≈ 1.5 μ.

[2] In [5], the optical constants of lead were given without a correction for the dependence of the surface impedance on the angle of incidence.

Table II. Some properties of the investigated lead films

	Investigated films	Bulk metal		Investigated films	Bulk metal
d, μ	0,6—1,2	∞	R_n/R_0	$6\cdot10^{-3}$	$1\cdot10^{-3}$
ρ, g/cm^3	11,3	11.3	R_{res}/R_0	$\sim5\cdot10^{-3}$	$\sim10^{-3}$
σ_0, cgs esu	$0.41\cdot10^{17}$	$0.41\cdot10^{17}$	T_c, °K	7.3	7.2
σ_N, cgs esu	$1,70\cdot10^{17}$	$1,70\cdot10^{17}$	E_H/jH, $\mu\Omega\cdot$cm/kOe	$+1,5\cdot10^{-8}$	$+1.0\cdot10^{-8}$
σ_H, cgs esu	$1.4\cdot10^{18}$	$1,5\cdot10^{18}$	Θ, °K	85	86

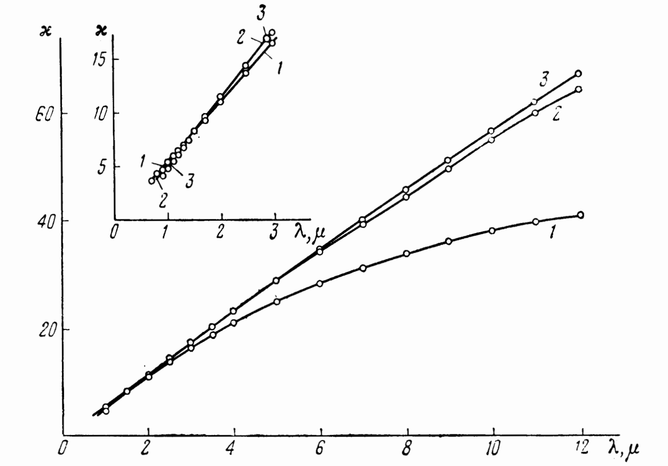

FIG. 2. Dependence of the absorption coefficient of lead on the wavelength at temperatures of: 1) 293°K; 2) 78°K; 3) 4.2°K.

of the films in the normal state R_n close to the critical temperature was $6 \times 10^{-3}R_0$ (R_0 is the resistance at T = 293°K). At this temperature, the resistance still had not reached its residual value. The residual resistance R_{res} was estimated from the temperature dependence of a sample's resistance. The critical temperature T_c was determined from the resistance discontinuity in the absence of a magnetic field. The accuracy in the determination of T_c was about 3%. The transition to the superconducting state was quite sharp, extending over about 0.2 deg. The penultimate row of Table II lists the value of the Hall field, per unit current and unit magnetic field; it gives the Hall field E_H for a current of density j = 1 A/cm^2 and a magnetic field H = 1 kOe. The "+" sign indicates the p-type nature of the Hall effect.

The value of the Hall coefficient of lead was 20 times smaller than that of copper or gold. This indicated considerable compensation of the electron and "hole" effects. The accuracy in determination of the Hall emf was 10% and was governed by scatter of the emf values for different samples. The value of E_H/jH was constant for values of H up to 8 kOe and j up to 70 A/mm^2. The last row of Table II gives the Debye temperature Θ, determined from the temperature dependence of the conductivity of lead. The determination was carried out in the same way as in[6] using the values of the "ideal" resistance tabulated by Grüneisen.[7,8] The accuracy in the determination of Θ was about 10%. The values of the properties of bulk lead were taken from handbooks.[8,9]

Taken as a whole, the results indicate that the properties of the investigated lead films were identical with the properties of the bulk metal. Since the thickness of the films exceeded, by a factor of several tens, the depth of the penetration of light, these films represented bulk metal from the optical point of view. Therefore, all the optical results and the conclusions refer to the bulk metal.

Table III

λ, μ	T = 293° K				T = 78° K				T = 4,2° K			
	$N\cdot10^{-22}$	$\nu_{opt}\cdot10^{-14}$	$10^2\beta_1$	$10^2\beta_2$	$N\cdot10^{-22}$	$\nu_{opt}\cdot10^{-14}$	$10^2\beta_1$	$10^2\beta_2$	$N\cdot10^{-22}$	$\nu_{opt}\cdot10^{-44}$	$10^2\beta_1$	$10^2\beta_2$
1,5	3,85	5.12	0.18	2.2	3.56	1,64	0.06	8.0	3,38	0.73	0.02	11.2
1,7	3,87	4,52	0,20	2,6	3.67	1,35	0.07	9.7	3,54	0.58	0,02	14.0
2,0	3.90	3.91	0.24	2.9	3.68	1,15	0.08	11.2	3.70	0.46	0,02	17.3
2,5	3,95	3.61	0.34	3.2	3.72	1.02	0.10	12,5	3,66	0.46	0.03	17,1
3,0	4.09	3.40	0,46	3.3	3.80	0.98	0,15	13.0	3.73	0.49	0.05	16,4
3,5	4.06	3.30	0.59	3.4	3,91	0,94	0.19	13,8	3.71	0.50	0.07	16.3
4,0	4.08	3,22	0.72	3,4	3,79	0.90	0.24	14.1	3,77	0,51	0,09	15,9
5,0	4.10	3.09	0,99	3,4	3,89	0.92	0,40	13,9	3.72	0,47	0.13	16,9
6.0	4,13	3,10	1,3	3.2	3,86	0.89	0,56	14,2	3,75	0.45	0.17	17,8
7.0	4,07	3.06	1.6	3.0	3.80	0,90	0,74	13,8	3,73	0.42	0.21	18,8
8,0	4,03	2.98	1,8	3.0	3,82	0,86	0.91	14,4	3.72	0,38	0.24	20,4
9,0	4,02	2.98	2.1	2,8	3.84	0,81	1.1	15.1	3.66	0,37	0.30	20,4
10.0	4,04	3,08	2,4	2.6	3.89	0,77	1.3	15,8	3,65	0,36	0,36	20,7
11,0	4.08	3,13	2.6	2.4	3,90	0,78	1.5	16.0	3,66	0,36	0,42	21,1
12,0	3.88	3,04	2,8	2,4	3,86	0.75	1.7	16.3	3,64	0,35	0,49	21,3

3. An analysis of the optical properties showed the presence of a weakly anomalous skin effect in lead in the investigated spectral range at all three temperatures. The surface losses were 2—3% at room temperature, 10—15% at nitrogen temperature, and even at helium temperature they did not exceed 22% of the total losses. Therefore, the experimental data were analyzed using the formulas for the weakly anomalous skin effect.[10]

The values of the conduction electron density N, the electron collision frequency ν_{opt} and the correction coefficients β_1 and β_2 (the coefficient β_2 represents the fraction of the surface absorption) are listed in Table III for various wavelengths.

The analysis was carried out by the method of successive approximations according to a procedure given in[4,6]. The approximations were continued until the value of a given quantity differed by less than 0.1% from the value in the preceding stage. Usually 2—3 approximations were sufficient. The complete dependence N(λ) for the three temperatures is given in Fig. 3. Figure 4 shows the dependence on the wavelength of the electron-phonon collision frequency $\nu^{\text{ef}} = \nu_{\text{opt}} - \nu^{\text{ed}}$; here ν^{ed} is the frequency of the collisions of electrons with impurities and defects.[3]

The nature of the dependences of N and ν on λ (Table 3 and Figs. 3 and 4) indicates the influence of the internal photoeffect on the optical constants of lead in the short-wavelength part of the spectrum. It follows from the figures that the limit of the influence of the internal photoeffect becomes more defined at lower temperatures. This makes it possible to separate the influence of various groups of electrons on the optical properties.

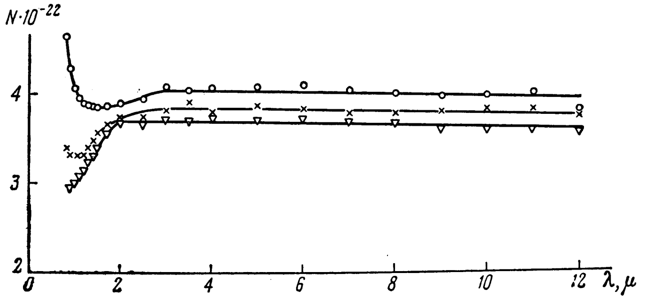

FIG. 3. Conduction electron density in lead: o – T = 293°K x – T = 78°K; ∇ – T = 4.2°K.

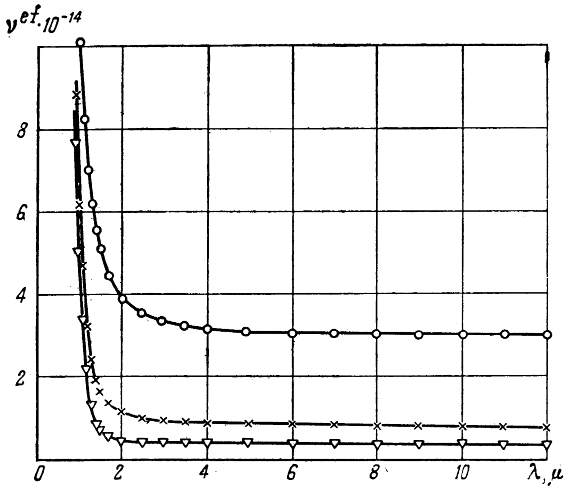

FIG. 4. Dependence of the electron–phonon collision frequency on the wavelength: o – T = 293°K; x – T = 78°K; ∇ – T = 4.2°K.

In the region λ ≳ 2—3 μ, the values of N and ν are constant. In this region of the spectrum, the microproperties are governed by the conduction electrons. The average values of some microproperties, obtained for this range, are listed in Table IV. The first and second rows of Table IV list the conduction electron density N and its ratio to the atomic concentration N_a. The third row gives the frequency of the electron-phonon collisions. The fourth row gives the classical frequency for the collisions of electrons with phonons $\nu^{\text{ef}}_{\text{cl}}$, which occurs in the expression for the static conductivity. The next row gives the value of the electron velocity v on the Fermi surface. The velocity v at T = 293°K and T = 78°K was determined from our optical data. In determining v, the least-squares method was used, as in[6]. Since the skin effect in lead is weakly anomalous, the role of the surface losses is small and the velocity v is found quite roughly (to within ≈20%), therefore the obtained values of v may be regarded as identical. Owing to the smallness of the coefficients β_1, it was difficult to determine v reliably from the optical constants at T = 4.2°K, and therefore we found v at this temperature using the data of Eisenstein[11] on the electronic specific heat of lead and our value of the conduction electron density.

The last two rows of Table IV give the electron mean free path l and the depth of penetration δ. These quantities depend relatively weakly on λ. The listed values refer to the region λ ≈ 4—6 μ. The ratio l/δ, which determines the nature of the skin effect, was less than unity at all temperatures.

---

[3] The value of ν^{ed} is ≈1.0 × 10^{12} and amounts to about 3% of ν^{ef} at T = 4.2°K. The introduction of the frequency of the electron–impurity collisions ν^{ed} at T = 4.2°K, when lead is in the superconducting state, is due to the fact that on the absorption of a high-energy photon, a pair dissociation takes place. The electron which has absorbed a quantum then interacts with impurities and phonons as if the metal were in the normal state. At T = 293°K and T = 78°K, the quantity ν^{ed} may be ignored completely compared with ν^{ef}, so that ν^{ef} coincides with ν_{opt}.

Table IV. Microproperties of lead

	$T = 293°$ K	$T = 78°$ K	$T = 4{,}2°$ K		$T = 293°$ K	$T = 78°$ K	$T = 4{,}2°$ K
N^*, 10^{22}	4.0	3.8	3,7	v, 10^8	0.9	1.0	0.71
N/N_a	1.2	1.15	1,1	l, 10^{-6}	0,3	1,1	1,5
ν^{ef}, 10^{14}	3,05	0.85	0.40	δ, 10^{-6}	3.2	2.9	2,8
ν^{ef}_{cl}, 10^{14}	2,50	0.56	—				

*All values are given in cgs units.

At room temperature, $l/\delta \approx 1/10$ and the skin effect was practically normal. At T = 4.2°K, $l/\delta = \frac{1}{2}$ and the surface absorption was already noticeable.

4. The values obtained for the electron—phonon collision frequency show that the temperature dependence of this frequency for lead confirms qualitatively the results obtained from the quantum transport equation. At T = 4.2°K, the value of ν^{ef} remains large and is equal to 0.40×10^{14} sec^{-1}. For comparison, we would state that the classical frequency of the electron-phonon collisions at this temperature should be 10^{10} sec^{-1}, which is 4×10^3 smaller than the observed frequency.

For quantitative comparison of the experimental and theoretical results, it is best to use the ratios of the collision frequencies at different temperatures. This eliminates the coefficients whose theoretical values are not very accurate. These ratios are as follows:

$$\nu^{ef}\,(293°\text{ K}) : \nu^{ef}\,(78°\text{ K}) : \nu^{ef}\,(4.2°\text{ K}) = \begin{cases} 1 : 0.28 : 0.13 \text{ (experiment)} \\ 1 : 0.29 : 0.12 \text{ (theory [3]).} \end{cases}$$

The experimental and theoretical results agree to within 10%. Such agreement may be regarded as good.[4)]

In the case of lead, in the same way as for tin,[4] the observed effect cannot be explained by the surface losses. These losses are allowed for in the formulas used for the weakly anomalous skin effect.[4,10] A possible error in this allowance is related to the fact that the formulas refer to a spherical Fermi surface and only the first-order correction terms are used in them, but this does not basically alter the results. This is because the skin effect in lead is "almost" normal even at helium temperatures and the neglect of the surface losses leads to an error not greater than 15—20% in the determination of the collision frequency.

The presence of the "residual" absorption could not be explained by the presence of impurities, defects, etc., since the losses due to their presence did not exceed 2% of the total losses even at T = 4.2°K.

The data in Table IV and Fig. 3 show that cooling reduces slightly the conduction electron density, ΔN. Although this reduction is at the limit of observation, the sign of ΔN has been found to be the same for all measurements at all wavelengths. This certainly indicates an effect associated with a change of the band overlap as the temperature drops. In lead, this effect is much weaker than in tin.[4]

In the short-wavelength region of the spectrum, the collision frequency ν_{opt} of lead depends on the frequency of light ω. This dependence is approximately quadratic in the region $\lambda \approx 1$—$4\,\mu$. The coefficient of ω^2 decreases as the temperature decreases by a factor of 9. However, even at T = 4.2°K, there is still a small quadratic tail. Assuming that at this temperature the whole quadratic dependence of the frequency ν_{opt} is due to electron—electron collisions,[12] we find that $\nu_{cl}^{ee} \approx 2 \times 10^8$ sec^{-1} (ν_{cl}^{ee} is the classical frequency of the electron-electron collisions, proportional to T^2). The corresponding theoretical value of this quantity for lead has been calculated by Ginzburg and Silin[13] and found to be 1.7×10^8 sec^{-1}. The agreement between the theory and experiment is surprisingly good. Obviously, a study of the electron-electron collisions, which affect considerably the optical properties of metals only in the near infrared region, is possible only at sufficiently low temperatures.

The present investigation of the optical properties of lead and the earlier study of tin at low temperatures[4] show that the absorption by these metals in the infrared region of the spectrum is mainly due to the interaction between electrons and phonons. The anomalous skin effect and the

---

[4)] The data obtained, taken as a whole, may also be used to compare the absolute values of the experimentally determined frequencies with those predicted theoretically. The theoretical values are found to be smaller than the experimental ones by about 30%. The additional contribution to the collision frequency is evidently associated with the influence of the interaction between electrons and high-frequency lattice vibrations.

associated surface losses are of secondary importance and do not govern the optical properties of these metals. The temperature dependence of the electron-phonon collision frequency in the presence of high-energy quanta does not agree with the classical formula but gives rise to the "residual" absorption at low temperatures. From this point of view, it would be interesting to carry out a detailed study of the optical properties of other metals at sufficiently low temperatures.

In conclusion, the author expresses his gratitude to G. P. Motulevich for constant interest and valuable comments.

---

[1] T. Holstein, Phys. Rev. **96**, 535 (1954).

[2] V. L. Ginzburg and G. P. Motulevich, UFN **55**, 469 (1955).

[3] R. N. Gurzhi, JETP **33**, 451, 660 (1957), Soviet Phys. JETP **6**, 352, 506 (1958); Dissertation presented at the Physico-technical Institute of the U.S.S.R. Academy of Sciences, 1958.

[4] A. I. Golovashkin and G. P. Motulevich, JETP **47**, 64 (1964), Soviet Phys. JETP **20**, 44 (1965).

[5] A. I. Golovashkin and G. P. Motulevich, JETP **44**, 398 (1963), Soviet Phys. **17**, 271 (1963).

[6] A. I. Golovashkin and G. P. Motulevich, JETP **46**, 460 (1964), Soviet Phys. JETP **19**, 310 (1964).

[7] E. Grüneisen, Ann. Physik **16**, 530 (1933).

[8] Collection: Low-Temperature Physics (Russ. Transl., ed. by A. I. Shal'nikov, IIL, 1959).

[9] Handbook of Chemistry and Physics, Cleveland, Ohio; Sbornik fizicheskikh konstant (Handbook of Physical Constants), ed. by Ya. G. Dorfman and S. É. Frish, ONTI, 37-th edition, C. J. Smithells, Metals Reference Book, 1955; J. M. Ziman, Electrons and Phonons (Russ. Transl. IIL, 1962).

[10] G. P. Motulevich, JETP **46**, 287 (1964), Soviet Phys. JETP **19**, 199 (1964).

[11] J. Eisenstein, Revs. Modern Phys. **26**, 277 (1954).

[12] R. N. Gurzhi, JETP **35**, 965 (1958), Soviet Phys. JETP 8, 673 (1959); L. P. Pitaevskiĭ, JETP 34, 942 (1958), Soviet Phys. JETP **7**, 652 (1958).

[13] V. L. Ginzburg and V. P. Silin, JETP **29**, 64 (1955), Soviet Phys. JETP **2**, 46 (1956).

Translated by A. Tybulewicz
118