International Journal of Thermophysics, Vol. 10, No. 4, 1989

Thermal Conductivity of Lead in the Range −180 to 500°C

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Received August 1, 1988

The thermal conductivity of lead (99.99%) has been measured in the range −180 to 500°C using four measuring devices (steady-state method). The thermal conductivity of both solid and liquid lead can be represented as a linear function of the temperature. The uncertainty of the measured values is estimated at 2.5% (solid) and 3% (liquid). Between the melting point and 500°C, the thermal conductivity increases by 14%. The ratio of the thermal conductivity of solid to liquid lead at the melting point corresponds to the ratio of the electric conductivities. The Lorenz function for liquid lead is approximately 1% above the ideal value at the melting point and some 3% lower than the ideal value at 500°C.

KEY WORDS: lead; molten metal; thermal conductivity.

1. INTRODUCTION

For the thermal conductivity of solid metals, far fewer values are available between −180°C and the ambient temperature than in the adjacent temperature ranges. Very few values are available for the molten state and these are generally more than 20 years old. The determination of the thermal conductivity of a metal over a large temperature range on the basis of various papers, each of which covers only a limited range, is always affected by uncertainties, due to different materials (purities) and different methods and apparatuses being used. This makes it very difficult to separate the differences and uncertainties caused by the material from those caused by the instrument.

The objective of the work described here was to measure the thermal conductivity of specimens of a single sample of lead over a large tem-

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0195-928X/89/0700-0765$06.00/0

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perature range. For this purpose, four different apparatuses (based on the same principle of measurement) were used, whose working ranges partly overlapped, so that instrument-specific differences—and thus systematic uncertainties—could be identified.

2. MATERIAL

The sample used had a purity of at least 99.994% (Preussag AG, Goslar, F.R.G.), with 0.005% bismuth representing the greatest impurity. According to the specifications of the suppliers, the total proportion of the 10 impurities determined analytically amounted to less than 0.006%.

Two cylinders were cast from a melt to provide two specimens for measurements on solid lead (50 mm in diameter, 90 mm high) and one specimen cylinder for measurements on liquid lead (49 mm in diameter, 45 mm high).

The density as determined from the mass and the dimensions of the cylinders was 11.34 $g \cdot cm^{-3}$.

3. METHOD OF MEASUREMENT AND MEASURING INSTRUMENTS

The measurements were carried out under steady-state conditions with axial heat flow through the cylindrical specimen using four different measuring arrangements, each designed for one particular temperature range. The heat flux density is determined from the electrical heating power of a heater and the cylinder cross section; the temperature difference is measured in the specimen, along the axis of the cylinder, using radially inserted thermocouples. Two cylinders were used for the measurements between -180 and 70°C. For the other measurements on solid lead one of these specimens was used.

3.1. Temperature Range -180 to 70°C

Figure 1 shows the device [1]. The two cylinders with a transverse heater arranged between them are installed in a copper casing, which can be evacuated. Silicone grease is applied to couple the end faces of the cylinders to the heater and to cold plates of the casing. The cylinders can be loaded with adjustable spring load to improve thermal contact. The copper casing is suspended in a cooling chamber, the temperature of which is kept constant by the controlled injection of liquid nitrogen. Temperature variations with time inside the cooling chamber are minimized due to the thick-walled copper casing. At the temperature measuring points at the extreme

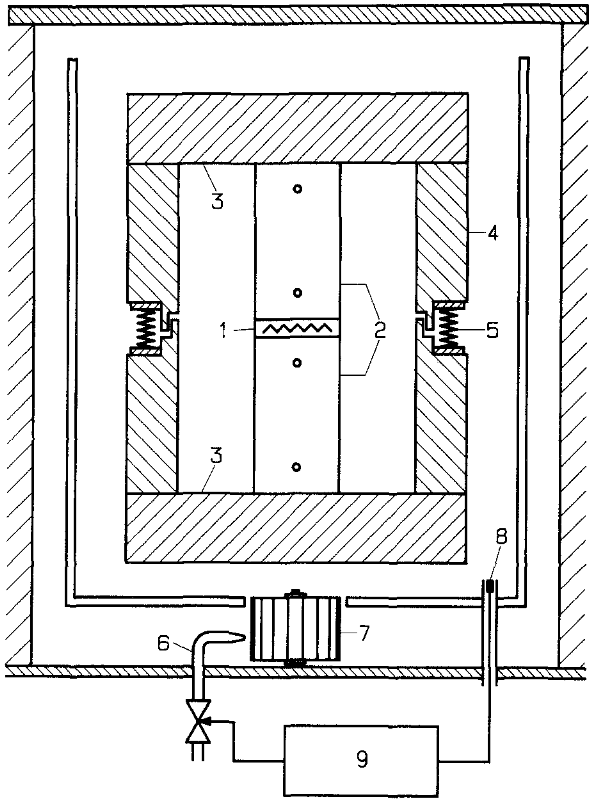

Fig. 1. Device for measuring the thermal conductivity between −180 and 70°C. (1) Heater; (2) specimen cylinders; (3) cold plates (screwed with 4); (4) copper casing; (5) spring bellows; (6) injection nozzle for liquid nitrogen; (7) turbine to spray and swirl the liquid nitrogen; (8) platinum resistance thermometer; (9) control unit; (○) sites of temperature measurement.

ends of the specimens, the maximum variations amount to about ±0.05 K. The heating power of the heater is adjusted to provide a temperature difference of approximately 10 K between the two thermocouples (which are 60 mm apart). As the contact resistances between the two cylinders and the adjacent plates are not exactly the same, somewhat different temperature differences are measured inside the two specimens (from 0.05 to 0.35 K; mean deviation, 0.2 K). Distribution of the total heating power (approximately 20 W) to the heat fluxes flowing through the specimens is proportional to the respective temperature difference. According to the slightly scattering temperature differences mentioned, the heat flux in the two pieces shows a difference between 0.5 and 3.5% (mean, 2%). The thermal conductivity is calculated from the heat flux density and the temperature difference (the thermal conductivity is assigned to the average value of the two mean temperatures). The influence of the Joule effect in the input leads to the heater is determined and taken into account as a correction (see Section 4). The heat transfer in the evacuated circular gap between the specimens and the casing, which is filled with cotton wool, can be neglected [1].

3.2. Temperature Range 20 to 85°C

Figure 2 shows the device [2]. The specimen cylinder is coupled to a heater and cold plate with silicone oil. The heater is shielded by a guard heater, and the temperature difference between these two components is kept as small as possible to avoid heat losses. The specimen is surrounded by a heated guard cylinder and the temperature of the upper end is adjusted as closely as possible to the heater temperature. The temperatures of the guard heater and heated guard cylinder are adjusted with the aid of a water thermostat; another thermostat serves to adjust and control the temperature of the cold plate and thus also the temperature of the lower end of the heated guard cylinder. A temperature difference of about 10 K is maintained in the specimen cylinder between the thermocouples, which

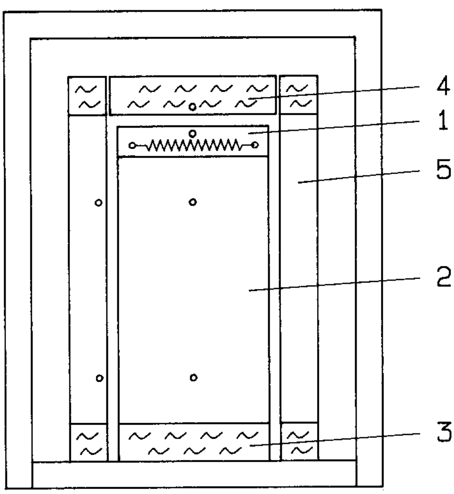

Fig. 2. Device for measuring the thermal conductivity between 20 and 85°C. (1) Heater; (2) specimen cylinder; (3) cold plate; (4) guard heater; (5) heated guard cylinder; (○) sites of temperature measurement.

are 60 mm apart. The heating power used is approximately 10 W. Heat losses of the heater due to temperature differences between heater and guard heater are taken into account as corrections.

3.3. Temperature Range 100 to 250°C

Figure 3 shows the device [3]. The arrangement corresponds to that described in Section 3.2, with electrical heaters used to adjust and control the temperature of the guard heater and heated guard cylinder instead of water thermostats.

By separate heating of the upper and lower end of the guard cylinder the temperature gradient is matched as closely as possible to the temperature gradient along the test cylinder. The highest operating temperature of the instrument is 500°C. In the case of solid lead measurements were carried out only up to 250°C. Preliminary tests at higher temperatures had shown that the material quickly deformed under the (low) contacting pressure applied. With a heating power of 10 W, the temperature difference in the cylinder was approximately 10 K. Heat losses due to mismatched guard heater temperatures are taken into account as correction.

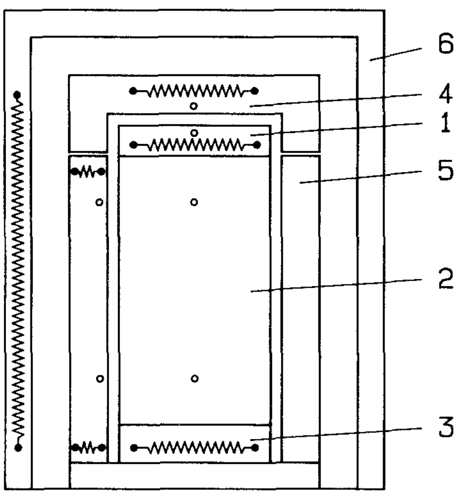

Fig. 3. Device for measuring the thermal conductivity between 100 and 250°C. (1) Heater; (2) specimen cylinder; (3) cold plate; (4) guard heater; (5) heated guard cylinder; (6) heated casing; (○) sites of temperature measurement.

3.4. Temperature Above the Melting Point of Lead

In order to measure the thermal conductivity of the liquid lead a steel setup [4] (shown in Fig. 4) was installed in the device described in Section 3.3. The still solid specimen (49 mm in diameter, 45 mm high) is placed into the thin-walled container (0.5-mm wall thickness), the upper steel part of which is equipped with a concentric groove to receive the expanding melt. Two thermocouples 30 mm apart in thin-walled steel tubes serve to measure the temperature difference in the melt. Another thermocouple measures the axial temperature in the upper steel part and, at the same time, serves to adjust the temperature of the upper heated guard cylinder to the preset value. In this case, a heating power of approximately 6 W produces a temperature difference of about 5 K and the highest temperature attained is about 500°C.

4. RESULTS, CORRECTIONS, AND UNCERTAINTIES

4.1. Results

Table I contains details of all of the experimental results.

Figure 5 shows all measured values, together with a curve fit for all three sets of data for the solid.

For ϑ between -180 and 327°C,

$$\lambda(\vartheta) = 35.49 - 1.82 \times 10^{-2}\, \vartheta \tag{1}$$

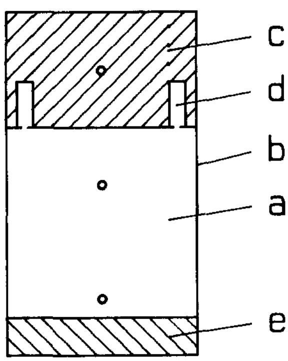

Fig. 4. Setup for the device according to Fig. 3 for measuring the thermal conductivity of liquid lead up to 500°C. (a) Specimen; (b) container wall (steel); (c) upper steel part; (d) concentric groove; (e) base plate; (○) sites of temperature measurement.

Table I. Thermal Conductivity λ of Lead, Measured with Four Different Devices (the Thermal Conductivity Is Assigned to the Mean Sample Temperature): ϑ in °C, λ in $W \cdot m^{-1} \cdot K^{-1}$

				Range, −180 to 70°C					
ϑ	−181.3	−177.9	−159.5	−142.7	−117.0	−98.2	−90.8	−80.3	
λ	39.01	38.85	38.48	38.03	37.48	37.14	36.95	36.74	
ϑ	−65.4	−59.5	−39.8	−20.5	−14.6	−6.3	11.0	40.0	71.5
λ	36.50	36.40	36.08	35.79	35.70	35.57	35.31	34.91	34.37
				Range, 20 to 85°C					
ϑ	20.0	22.5	40.0	60.0	85.0	85.0			
λ	35.29	35.49	35.18	34.70	34.15	34.25			
				Range, 100 to 250°C					
ϑ	100.0	100.8	150.0	200.0	250.0				
λ	33.39	33.09	32.45	31.80	31.01				
				Range, 327 to 500°C					
ϑ	343.6	401.2	424.7	447.4	499.7				
λ	15.82	16.57	16.87	17.15	17.845				

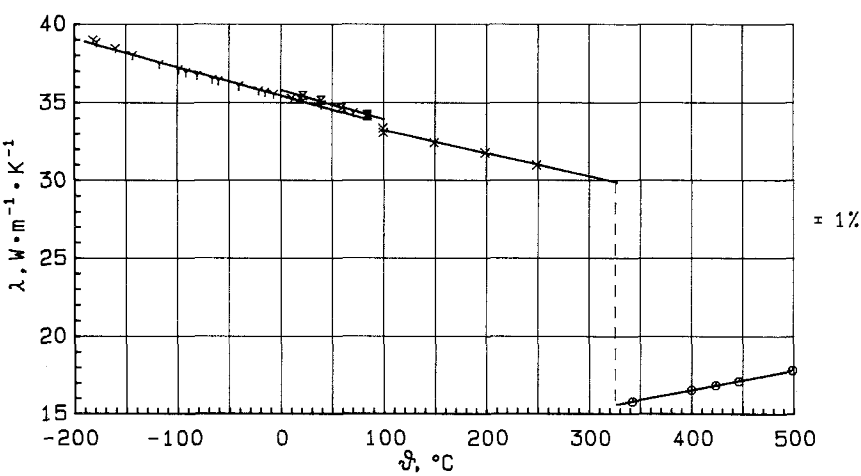

Fig. 5. Thermal conductivity λ of lead as a function of the temperature ϑ. Measured values and fitted lines. (Y) Low-temperature apparatus; (X̄) medium-temperature apparatus; (X) high-temperature apparatus; (①) setup for molten lead.

with deviations of the measured values from the fitted line ranging from -1.2 to 1.7% at the most.

The data for the melt were similarly fitted.

For ϑ between 327 and 500°C,

$$\lambda(\vartheta) = 11.37 + 1.29 \times 10^{-2}\,\vartheta \tag{2}$$

with deviations of the measured values from the fitted line of less than 0.1%.

4.2. Corrections and Uncertainties

The experimental results include the following corrections.

(a) The thermal expansion of the specimens. This leads to corrections of between -0.9 and 0.5%. The uncertainty of these corrections is estimated at 0.1% at the most. Thus their contribution to the total uncertainty is negligible.

(b) The influence of the Joule effect in the input leads to the heater of the low-temperature device. This correction has been determined quantitatively [1] and amounts to about 1% with an uncertainty of approximately 20%. Its contribution to the total uncertainty of the measured values is, therefore, about 0.2%.

(c) The proportion of heat passing through the steel wall of the liquid cell depends on the ratio area × thermal conductivity for steel wall and specimen. It amounts to about 5% of the total heat flux, with the uncertainty of this correction estimated at 10%. The total uncertainty affecting the results amounts to approximately 0.5%.

(d) The temperature profile along the surface of the container for the melt (Fig. 4) deviates from the temperature profile of the heated guard cylinder. The temperature profiles of both surfaces in the longitudinal direction can be approximated on the basis of the temperatures measured. The heat transfer between both surfaces has been estimated to be 0.15% at the most. Its determination is, however, affected by such an uncertainty that its contribution to the uncertainty of the measured values is estimated to be 0.1%.

The following uncertainties must also be added to the corrections.

(a) The uncertainty of the determination of the heating power (maximally 0.1%)

(b) The uncertainty of the determination of the temperature difference along the specimen (maximally 0.5% for solid lead, approximately 0.7% for the melt)

(c) The uncertainty of the determination of the specimen area (approximately 0.1%).

For repeat measurements carried out with the solid specimen mounted in the device, changes of the measured values ranged from −0.6 to 0.9%, when the temperature of the device was reduced from the maximum to the minimum value as shown in Fig. 6. That is why a scatter of ±0.5% is taken into account as a systematic uncertainty of the results. Moreover, the results given by the different devices show systematic differences in the overlapping range. The low-temperature values lie about 1% below the intermediate-temperature values, which in turn, lie about 2% above the high-temperature results.

It is therefore assumed that the thermal conductivity values according to the fitted lines for solid lead (Fig. 5) are affected by a systematic uncertainty of ±1%. Consequently, an additive total uncertainty of 2.5% results for solid lead. Systematic uncertainties totaling 1.5% are assumed for the measured values of liquid lead so that the additive total uncertainty amounts to 3%.

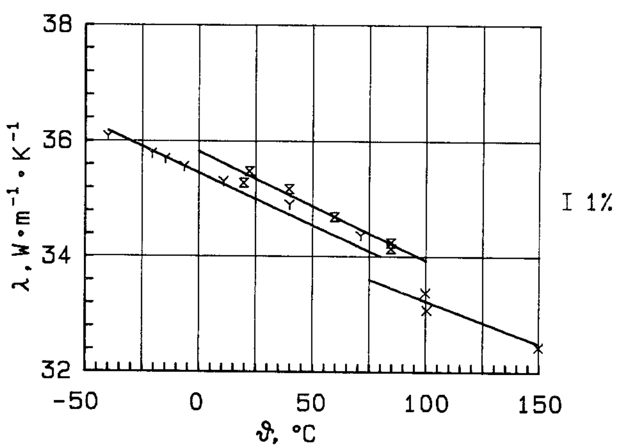

Fig. 6. Results of the three devices in the overlapping temperature range with fitted lines (λ, thermal conductivity; ϑ, temperature). (Y) Low-temperature apparatus; (X̄) medium-temperature apparatus; (X) high-temperature apparatus.

5. DISCUSSION

Figure 7 shows the fitted line of the values measured within the scope of this work for solid and liquid lead, together with the recommended values [5] and results of other investigations [6, 7]. The recommended values decrease less strongly with increasing temperature than the values found in the present measurements. The recommended values for higher temperatures are based on a few papers and the values given in these papers are higher than those obtained by Cook et al. [6]. The latter values are higher by up to 2.5% than the present values up to the melting point. The difference from the recommended values increases from 2% at 80°C to about 5% at the melting point.

For liquid lead, in the vicinity of the melting point, agreement of the values measured at the PTB with the recommended values is very good. With increasing temperature the recommended values increase more strongly than the PTB values, and at 500°C they show a difference of about 2%. Agreement with the experimental values of Powell and Tye [7] and Powell [8] is somewhat better and no evidence was found to show a decrease in the thermal conductivity with increasing temperature as found by Konno [9] or the temperature independence according to Filippov [10]. The recommended value for the thermal conductivity at the melting point is 15.5 $W \cdot m^{-1} \cdot K^{-1}$; we obtained 15.6 $W \cdot m^{-1} \cdot K^{-1}$.

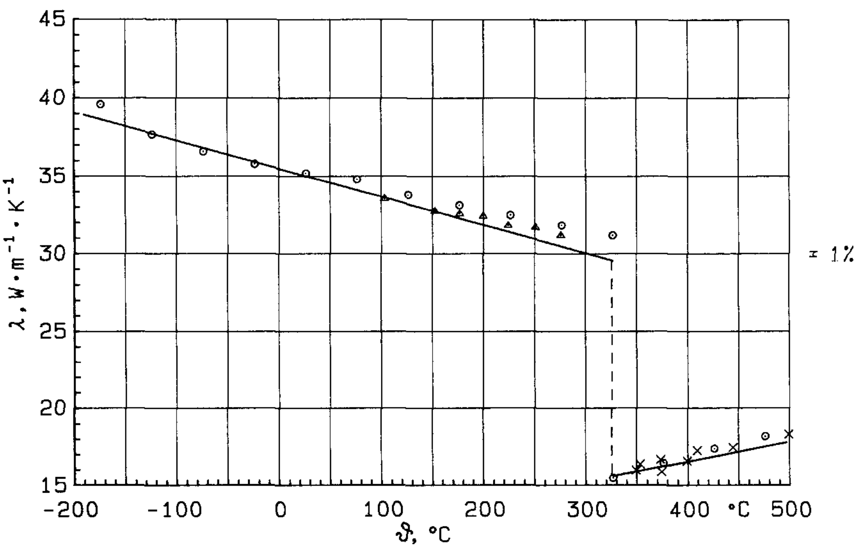

Fig. 7. Fitted line for the dependence of the thermal conductivity (PTB results) on the temperature ϑ together with values taken from the literature. (⊙) From Ref. 5; (△) from Ref. 6; (X) from Ref. 7.

As electron movement is the dominating mechanism of thermal conductivity in metals, the ratio of the thermal conductivity of solid (λ_s) and liquid lead (λ_l) at the phase transition should correspond to the respective ratio of the electrical conductivities. We obtained $\lambda_s/\lambda_l = 1.89$ for the thermal conductivity, which is in good agreement with the recommended value [5] of 2.01. Roll and Motz [11] found the ratio of the electrical conductivities to be 1.94. Table II shows the material properties at the melting point.

As it is very difficult to measure the thermal conductivity λ of liquid metals, it is generally determined from the specific electric resistivity ρ, which is easier to measure. The Lorenz function L is applied for this purpose: ($L = \lambda\rho/T$). When the deviation of the Lorenz function from the ideal value $L_0 = 2.443 \times 10^{-8}\ \mathrm{V}^2 \cdot \mathrm{K}^{-2}$ remains within the range of an acceptable uncertainty, the thermal conductivity can be calculated with $L \approx L_0$.

For most of the measurements of ρ and λ on liquid metals, the relative deviation $(L - L_0)/L_0$ lies in the range of $\pm 10\%$ [8]. Figure 8 shows $(L - L_0)/L_0$ for liquid lead; the deviation decreases from 1 to -3%.

Although λ and ρ have not been determined on the same material, the represented curve should be correct, as the reliable values for ρ taken from the literature differ only very slightly from one another (measured with an accuracy of 1% by means of a rotating-field method without electrodes [1]; using capillaries with liquid metal threads and a dc four-point probe technique; estimated accuracy, 0.15% [12]). In the case of tin, $(L - L_0)/L_0$ also decreased by about 5% in all, a minimum being found at a temperature of 150 to 200 K above the melting point [4]. Should there also be a minimum for lead, it has not yet been reached in the present temperature range.

A Russian group working at the University of Moscow has postulated that the temperature independence of the thermal conductivity and the increasing decrease in the Lorenz function with increasing temperature are the characteristic features of liquid metals, and they have tried to prove this

Table II. Properties of Lead at the Melting Point (Thermal Conductivity λ, Specific Electric Resistivity ρ [11], Lorenz Function $L = \lambda\rho/T$ and Its Relative Deviation from the Ideal Value $L_0 = 2.443 \cdot 10^{-8}\ \mathrm{V}^2 \cdot \mathrm{K}^{-2}$)

	λ $(\mathrm{W} \cdot \mathrm{m}^{-1} \cdot \mathrm{K}^{-1})$	ρ $(10^{-8}\ \Omega \cdot \mathrm{m})$	L $(10^{-8}\ \mathrm{V}^2 \cdot \mathrm{K}^{-2})$	$(L - L_0)/L_0$ (%)
Solid at 327.5°C	29.55	49	2.41	−1.35
Liquid at 327.5°C	15.61	95	2.47	0.98
	$\lambda_s/\lambda_l = 1.89$	$\rho_l/\rho_s = 1.94$		

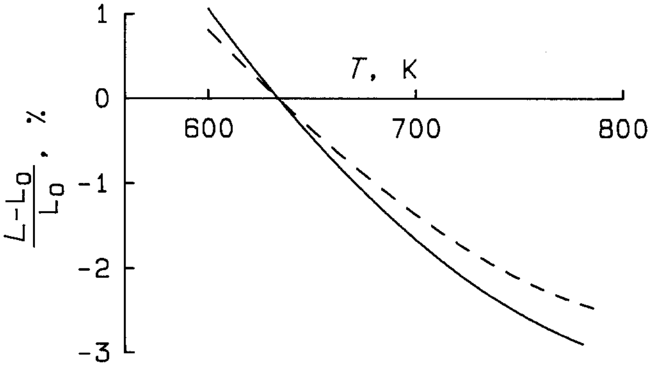

Fig. 8. Relative deviation of the Lorenz function L from the ideal value $L_0 = 2.443 \times 10^{-8}\ V^2 \cdot K^{-2}$ for liquid lead as a function of temperature. (——) With electric conductivity according to Ref. 11; (-----) with electric conductivity according to Ref. 12.

assumption for lead and tin on the basis of experiments [13, 14]. The present results do not confirm this.

Although the Lorenz function for lead (measured up to 500°C) decreases according to Fig. 8, the decrease is considerably lower than suggested by Filippov [13]. The thermal conductivity is not temperature independent but increases with increasing temperature by about 14% between the melting point and 500°C. A more or less constant thermal conductivity should result in a deviation $(L - L_0)/L_0$ of about -14% but we find a change of only -3% at 500°C. Consequently, the temperature independence of the thermal conductivity is not a distinctive feature of liquid metals, and the main argument for the conclusion that the Lorenz function of all liquid metals continuously decreases with increasing temperature is thus demolished. The reasons for the relatively small decrease in the Lorenz function above the melting point and for a possible new increase (in the case of tin) have not yet been clarified.

ACKNOWLEDGMENTS

The author wishes to thank Mr. W. Stein und Mr. H.-W. Krupke, who carried out the difficult and time-consuming measurements.

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