

## **The Thermal Diffusivity of Argon in the Critical Region**

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In this paper experimental results on the thermal diffusivity of argon in the supercritical region are reported. Five isotherms were investigated at 150.90, 153.16, 163.15, 173.14, and 188.14 K, in the pressure range from 2 to 13 MPa, corresponding to density variations from 90 to 800 kg · m<sup>-3</sup>. The experimental thermal diffusivity data are compared with theoretical predictions. The corresponding thermal conductivity coefficients are calculated and correlated with respect to the spinodal curve.

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**KEY WORDS:** argon; thermal conductivity; thermal diffusivity; supercritical region.

### **1. EXPERIMENTAL METHOD**

The measurements of the thermal diffusivity of argon were carried out with the same type of interferometer [1] which was used by Bach and Grigull [2]. The light-scattering interferometer which has been developed is based on the shadow apparatus Tepler IAB—451. It operates with a He—Ne laser light source at a wavelength of 6328 Å.

Initially, the first experimental apparatus built was expected to be used for simultaneous measurements of the thermal diffusivity and the thermal conductivity in the supercritical region. For this purpose, the cell used by Bach and Grigull was modified to measure the heat flux distribution between two semiinfinite media. It should be recalled that in this configura-

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tion of two semiinfinite media, with the heat flux source constituted by a thin layer of chromium ( $0.01 \mu\text{m}$ ) located at their boundary, a nonstationary method to measure thermal conductivity is used in one of them. The second semiinfinite medium was used as the standard with known and stable thermophysical properties ( $a$ ,  $\lambda$ ,  $\rho$ ,  $C_p$ ). It was made of quartz. A platinum resistance thermometer was located in the quartz at a finite distance from the chromium layer. However, this type of cell and the changes brought later, which included the replacement of the chromium layer by a platinum one on an aluminium substrate which was used at the same time as the heating source and resistance thermometer, did not allow us to obtain the values of the thermal conductivity with small enough errors.

## 2. EXPERIMENTAL RESULTS

The experiments were carried out in the supercritical region along five isotherms, 150.90, 153.16, 163.15, 173.14, and 188.14 K. The uncertainty in the thermal diffusivity measurements was estimated to vary from 1 to 2.5% with the exception of a small pressure range around the extremum of

Table I. Thermal Diffusivity of Argon

$T$ (K)	$P$ (MPa)	$\rho$ ( $\text{kg} \cdot \text{m}^{-3}$ )	$10^7 a$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )
188.12	3.153	91.12	2.587
188.11	3.369	98.27	2.333
188.15	4.701	145.81	1.471
188.16	5.996	198.46	1.050
188.07	7.256	257.26	0.730
188.14	8.455	319.82	0.545
188.15	10.208	420.93	0.414
188.15	11.239	480.81	0.376
188.19	11.380	488.40	0.379
188.16	12.512	549.16	0.365
188.15	14.326	630.82	0.376
173.19	3.025	98.59	1.892
173.21	3.367	112.17	1.597
173.15	4.466	161.07	1.065
173.14	4.793	177.45	0.902
173.14	5.602	222.59	0.673
173.16	6.085	253.27	0.550
173.15	7.039	324.86	0.401
172.99	7.684	384.02	0.321
173.01	7.959	410.22	0.303

Table I. (Continued)

$T$ (K)	$P$ (MPa)	$\rho$ ( $\text{kg} \cdot \text{m}^{-3}$ )	$10^7 a$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )
173.15	8.551	466.59	0.262
173.15	8.722	483.37	0.258
173.15	9.016	511.61	0.254
173.17	9.941	591.35	0.250
173.18	11.174	672.68	0.282
173.15	12.769	745.91	0.334
163.14	2.590	90.28	1.883
163.15	3.931	154.92	0.894
163.15	4.284	175.61	0.758
163.15	5.472	265.69	0.399
163.18	5.997	322.38	0.283
163.10	6.596	411.34	0.196
163.15	6.955	473.34	0.167
163.14	7.006	482.85	0.165
163.14	7.080	496.19	0.166
163.15	7.175	512.85	0.163
163.17	7.307	535.14	0.158
163.12	7.642	590.15	0.163
163.15	7.771	607.37	0.169
163.19	8.878	716.30	0.229
163.16	9.701	766.16	0.281
153.19	2.513	97.11	1.477
153.25	3.727	169.62	0.633
153.17	4.499	243.44	0.374
153.15	4.708	273.28	0.310
153.15	4.885	305.92	0.218
153.16	4.984	329.13	0.179
153.15	5.046	347.31	0.154
153.15	5.147	384.90	0.110
153.17	5.190	404.43	0.087
153.15	5.268	456.76	0.058
153.16	5.320	500.55	0.052
153.15	5.335	516.77	0.050
153.15	5.351	532.54	0.049
153.15	5.408	585.36	0.050
153.17	5.422	593.03	0.050
153.15	5.479	633.01	0.056
153.18	5.552	662.10	0.077
153.17	5.567	668.45	0.075
153.18	5.955	746.07	0.174
153.15	6.838	819.73	0.262
153.16	7.643	857.36	0.308

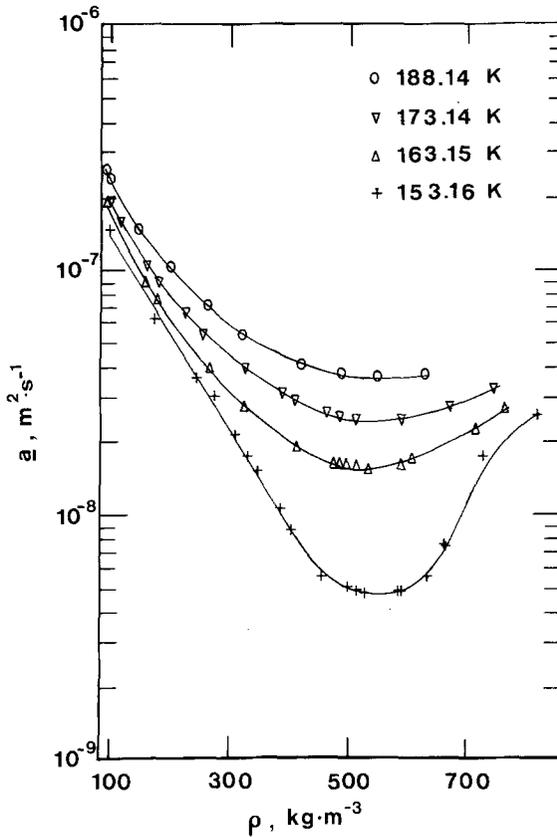


Fig. 1. Variation of the thermal diffusivity of argon as a function of density.

150.90 and 153.16 K isotherms, where they can reach, respectively, 17 and 6.5%. Due to large errors in the experimental results of the 150.90 K isotherm, these data were not used in the analysis. The experimental data for the thermal diffusivity of argon are reported in Table I and plotted as a function of density in Fig. 1.

### 3. ANALYSIS OF THE RESULT ON THERMAL DIFFUSIVITY

#### 3.1. Along the Critical Isochore

To determine the experimental values of the thermal diffusivity on the critical isochore, the data along each isotherm were fitted following polyno-

mials by a least-squares method. The best fit which leads to the minimum error was obtained for a polynomial of degree 7.

$$10^7 a = \sum_{i=0}^7 K_i (\rho - \rho_c)^i \quad (1)$$

where  $a$  is in  $\text{m}^2 \cdot \text{s}^{-1}$ .

The coefficients of these polynomials for the isotherms which were studied are given in Table II.

For the critical parameters of argon, the following values were selected [3]:

$$T_c = 150.66 \text{ K}$$

$$\rho_c = 535.1 \text{ kg} \cdot \text{m}^{-3}$$

$$P_c = 4.850 \text{ MPa}$$

Along the critical isochore, thermal diffusivity values were calculated by Eq. (1) and were fitted by the following equation:

$$a_{\rho = \rho_c} = 10^{-7} (1.01 \pm 0.01) (t)^{0.739 \pm 0.005} \quad (2)$$

where  $a$  is in  $\text{m}^2 \cdot \text{s}^{-1}$  and  $t = (T - T_c)/T_c$ .

The results are shown in Fig. 2 and are compared with the data of thermal diffusivity of other noble gases along the critical isochore [4-7].

### 3.2. Outside the Critical Isochore

Following the mode-mode coupling theory [8, 9], the thermal diffusivity in the supercritical region is calculated in first approximation by

$$a_{\text{cal}} = a_{\text{reg}} + \Delta a(\rho, T) \quad (3)$$

Table II. Coefficients of Eq. (1)

$K_i$	$T$ (K)			
	188.14	173.14	163.15	153.16
$K_0$	$3.5976290 \times 10^{-1}$	$2.4869832 \times 10^{-1}$	$1.6267476 \times 10^{-1}$	$4.8737480 \times 10^{-2}$
$K_1$	$3.0555299 \times 10^{-4}$	$-1.5824172 \times 10^{-4}$	$-2.3000260 \times 10^{-5}$	$-9.5198377 \times 10^{-5}$
$K_2$	$1.4653982 \times 10^{-5}$	$1.7134644 \times 10^{-6}$	$5.2921772 \times 10^{-7}$	$7.7379333 \times 10^{-7}$
$K_3$	$2.0133434 \times 10^{-8}$	$5.2173430 \times 10^{-9}$	$-1.8883936 \times 10^{-9}$	$8.8343393 \times 10^{-9}$
$K_4$	$-1.0263790 \times 10^{-9}$	$5.5003418 \times 10^{-11}$	$7.4490810 \times 10^{-11}$	$1.0958613 \times 10^{-10}$
$K_5$	$-7.7373675 \times 10^{-12}$	$-5.2662321 \times 10^{-14}$	$1.0017631 \times 10^{-13}$	$-2.6865459 \times 10^{-13}$
$K_6$	$-2.0073882 \times 10^{-14}$	$-7.7215088 \times 10^{-16}$	$-6.8014500 \times 10^{-16}$	$-2.6134222 \times 10^{-15}$
$K_7$	$-1.8049261 \times 10^{-17}$	$-1.3823182 \times 10^{-18}$	$-1.6064264 \times 10^{-18}$	$-3.9026014 \times 10^{-18}$

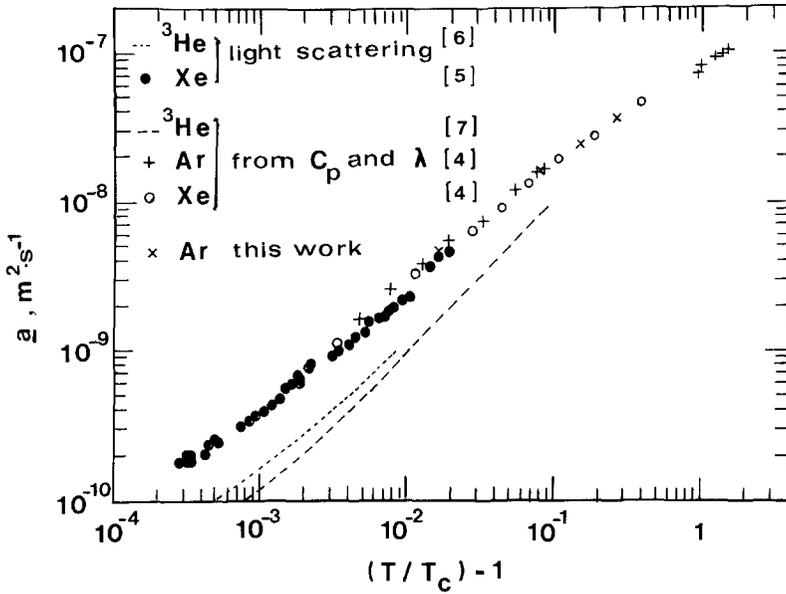


Fig. 2. Variation of the thermal diffusivity of noble gases along their critical isochore.

where

$$a_{\text{reg}} = \frac{\lambda_{\text{reg}}}{\rho C_p} \quad (4)$$

and

$$\Delta a(\rho, T) = \frac{\Delta \lambda(\rho, T)}{\rho C_p} = (1.01 \pm 0.04) \frac{k_B T}{6\pi\eta(\rho, T) \xi(\rho, T)} \quad (5)$$

where  $k_B$  is the Boltzmann constant,  $\eta$  the viscosity, and  $\xi$  the correlation length.

The correlation length (in meters) can be approximated by the Ornstein-Zernike theory:

$$\xi = R(nk_B T K_T)^{1/2} \quad (6)$$

where  $R = R_0(n/k_B T)^{1/2}$ .

Then

$$\xi = R_0(n^2 K_T)^{1/2} \quad (7)$$

The values of the isothermal compressibilities were calculated by the NBS equation of state [13]. The singular part of the viscosity [11, 12] was

neglected and the correlation length defined by Eq. (7) was used to calculate the thermal diffusivity by Eq. (3). A reasonable agreement was found between experimental and calculated values as can be seen in Table III, where a comparison is given for two isotherms.

**Table III.** Comparison Between Experimental and Calculated Data on the Thermal Diffusivity of Argon

$T$ (K)	$\rho$ ( $\text{kg} \cdot \text{m}^{-3}$ )	$10^7 a_{\text{exp}}$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )	$10^7 \Delta a(\rho, T)$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )	$10^7 a_{\text{cal}}$ ( $\text{m}^2 \cdot \text{s}^{-1}$ )	$[(a_{\text{exp}} - a_{\text{cal}})/a_{\text{cal}}] \times 100$ (%)
163.14	90.28	1.883	0.500	2.304	18.3
163.15	154.92	0.894	0.309	1.189	24.8
163.15	175.61	0.758	0.268	0.994	23.7
163.15	265.69	0.399	0.155	0.505	21.0
163.18	322.38	0.283	0.112	0.349	18.9
163.10	411.34	0.169	0.074	0.224	12.5
163.15	473.34	0.167	0.060	0.188	11.2
163.14	482.85	0.165	0.058	0.186	11.3
163.14	496.19	0.166	0.056	0.181	8.3
163.15	512.85	0.163	0.054	0.178	8.4
163.17	535.14	0.158	0.052	0.176	10.2
163.12	590.15	0.163	0.047	0.180	9.4
163.15	607.37	0.169	0.047	0.184	8.2
163.19	716.30	0.229	0.048	0.247	7.3
153.19	97.11	1.477	0.440	1.913	22.8
153.25	169.62	0.633	0.246	0.879	28.0
153.17	243.44	0.374	0.143	0.436	14.2
153.15	273.28	0.310	0.113	0.328	5.2
153.15	305.92	0.218	0.089	0.240	9.2
153.16	329.13	0.179	0.074	0.192	6.8
153.15	347.31	0.154	0.065	0.161	4.3
153.15	384.90	0.110	0.048	0.113	2.7
153.17	404.43	0.087	0.041	0.094	7.4
153.15	456.76	0.058	0.029	0.063	7.9
153.16	500.55	0.052	0.023	0.050	4.0
153.15	516.77	0.050	0.022	0.048	4.3
153.15	532.54	0.049	0.021	0.046	6.5
153.15	585.36	0.050	0.020	0.048	4.2
153.17	593.03	0.050	0.020	0.050	0.0
153.15	633.01	0.056	0.021	0.061	8.2
153.18	662.10	0.077	0.023	0.077	0.0
153.17	668.45	0.075	0.024	0.081	7.4
153.18	746.07	0.174	0.032	0.154	13.0
153.15	819.73	0.262	0.039	0.249	5.2
153.16	857.36	0.308	0.042	0.303	1.7
153.16	766.16	0.281	0.051	0.294	4.4

#### 4. DETERMINATION OF THE THERMAL CONDUCTIVITY OF ARGON

The values of the density and the specific heat at constant pressure are needed to calculate the values of the thermal conductivity following the relation

$$\lambda = a\rho C_p \quad (8)$$

These quantities were determined by the equation of state of NBS for argon [13]. The singular part of the thermal conductivity is given by

$$\Delta\lambda = \lambda - \lambda_{\text{reg}} \quad (9)$$

where the regular part  $\lambda_{\text{reg}}$  was estimated by an analysis of the experimental results reported in Ref. 14. The regular part of the thermal conductivity was approximated by the following equation:

$$\lambda_{\text{reg}} = (a_1 + a_2 T + a_3 T^2) T^{1/2} + (a_4 + a_5 T) \rho + a_6 \rho^2 + a_7 \rho^3 + a_8 \rho^4 + a_9 \rho^5 \quad (10)$$

where  $\rho$  is the amagat density. (One amagat unit of density corresponds for argon to  $1.7834 \text{ kg} \cdot \text{m}^{-3}$ .) The coefficients of Eq. (10) are reported in Table IV.

The values of thermal conductivities and singular parts of thermal conductivities are shown in Table V.

Table IV. Coefficients of Eq. (10)

$a_i$	Numerical value
$a_1$	$0.3850 \times 10^{-3}$
$a_2$	$0.2949 \times 10^{-5}$
$a_3$	$-0.278 \times 10^{-8}$
$a_4$	$0.3537 \times 10^{-4}$
$a_5$	$0.235 \times 10^{-7}$
$a_6$	$0.4465 \times 10^{-7}$
$a_7$	$0.9416 \times 10^{-10}$
$a_8$	$-0.1217 \times 10^{-12}$
$a_9$	$0.2510 \times 10^{-15}$

Table V. Thermal Conductivity Coefficient of Argon

$T$ (K)	$\rho$ ( $\text{kg} \cdot \text{m}^{-3}$ )	$C_p$ ( $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ )	$10^3 \lambda$ ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )	$10^3 \Delta \lambda$ ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )
188.12	91.12	683.4	16.11	2.41
188.11	98.27	698.4	16.01	2.13
188.15	145.81	811.1	17.40	2.26
188.16	198.46	961.2	20.03	3.39
188.07	257.26	1151.5	21.63	3.18
188.14	319.82	1351.8	23.74	3.16
188.15	420.93	1647.1	28.70	4.24
188.15	480.81	1737.3	31.41	4.34
188.19	488.40	1742.3	32.25	4.83
188.16	549.16	1747.3	35.02	4.64
188.15	630.82	1657.2	39.31	4.46
173.19	98.59	747.0	13.82	0.80
173.21	112.17	781.0	13.99	0.62
173.15	161.07	951.2	16.32	1.64
173.14	177.45	1018.8	16.31	1.17
173.14	222.59	1231.6	18.45	1.98
173.16	253.27	1396.8	19.46	2.04
173.15	324.86	1837.4	23.94	4.10
172.99	384.02	2210.4	27.25	5.22
173.01	410.22	2345.5	29.15	6.07
173.15	466.59	2533.3	30.97	5.48
173.15	483.37	2565.8	32.01	5.76
173.15	511.61	2588.4	33.64	6.08
173.17	591.35	2470.7	36.53	4.89
173.18	672.68	2157.8	40.93	4.51
173.15	745.91	1834.9	45.71	4.34
163.14	90.28	751.0	12.77	0.54
163.15	154.92	1021.3	14.15	0.23
163.15	175.61	1136.5	15.13	0.64
163.15	265.69	1850.0	19.61	2.39
163.18	322.38	2500.8	22.82	3.68
163.10	411.34	2639.7	29.34	6.83
163.15	473.34	4137.9	32.71	7.56
163.14	482.85	4182.9	33.33	7.75
163.14	496.19	4223.0	34.78	8.50
163.15	512.85	4243.0	35.47	8.49
163.17	535.14	4218.0	35.66	7.59
163.12	590.15	3967.7	38.17	7.25
163.15	607.37	3822.5	39.24	7.36
163.19	716.30	2713.5	44.51	5.89
163.16	766.16	2265.4	48.77	6.58
153.19	97.11	826.1	11.85	0.03
153.25	169.62	1281.7	13.76	0.02

Table V. (Continued)

$T$ (K)	$\rho$ ( $\text{kg} \cdot \text{m}^{-3}$ )	$C_p$ ( $J \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ )	$10^3 \lambda$ ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )	$10^3 \Delta\lambda$ ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )
153.17	243.44	2227.9	20.28	4.37
153.15	273.28	2878.7	24.39	7.53
153.15	305.92	3882.5	25.89	7.94
153.16	329.13	4846.3	28.55	9.79
153.15	347.31	5795.0	30.99	11.57
153.15	384.90	8365.9	35.42	14.58
153.17	404.43	10000.5	35.19	13.58
153.15	456.76	15212.3	40.30	16.51
153.16	500.55	18897.1	49.19	23.43
153.15	516.77	19840.8	51.27	24.74
153.15	532.54	20268.8	52.89	25.59
153.15	585.36	18093.5	52.96	22.94
153.17	593.03	17214.9	51.04	20.60
153.15	633.01	12816.7	45.43	12.74
153.18	662.10	9642.5	49.16	14.72
153.17	668.45	9079.3	45.52	10.68
153.18	746.07	4413.2	57.29	17.25
153.15	819.73	2653.4	56.99	11.24
153.16	857.36	2177.8	57.51	8.51

## 5. ANALYSIS OF THE RESULTS OF THE THERMAL CONDUCTIVITY

### 5.1. Along the Critical Isochore

In order to determine the singular part of the thermal conductivity along the critical isochore, the calculated data on each isotherm were approximated by a polynomial, following the least-squares method:

$$10^3 \Delta\lambda = \sum_{i=0}^6 N_i (\rho - \rho_c)^i \quad (11)$$

where  $\Delta\lambda$  is in  $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ , and the density in amagat units. The coefficients of Eq. (11) are reported in Table VI.

Along the critical isochore the data were represented by

$$\Delta\lambda_{\rho=\rho_c} = (1.9 \pm 0.2) 10^{-3} t^{0.62 \pm 0.04} \quad (12)$$

The comparison between the results of this present work and the experimental thermal conductivity data of argon, in the supercritical

Table VI. Coefficients of Eq. (11)

$N_i$	$T$ (K)			
	188.14	173.14	163.15	153.16
$N_1$	4.7552817	5.7299590	8.0248816	24.463139
$N_2$	$2.7262346 \times 10^{-3}$	$-7.0301596 \times 10^{-3}$	$-4.7698441 \times 10^{-3}$	$-1.9611018 \times 10^{-4}$
$N_3$	$-4.3791825 \times 10^{-5}$	$-6.2034623 \times 10^{-5}$	$-1.1627018 \times 10^{-4}$	$-1.0474730 \times 10^{-3}$
$N_4$	$-1.8462172 \times 10^{-7}$	$1.5830671 \times 10^{-7}$	$2.1399244 \times 10^{-7}$	$-2.1362698 \times 10^{-6}$
$N_5$	$-2.4948889 \times 10^{-10}$	$6.4428307 \times 10^{-10}$	$1.1926806 \times 10^{-9}$	$2.0867450 \times 10^{-8}$
$N_6$	$-4.7464798 \times 10^{-14}$	$4.0245951 \times 10^{-13}$	$1.2082196 \times 10^{-13}$	$9.4206411 \times 10^{-11}$
$N_7$	0	0	$-1.5985122 \times 10^{-15}$	$1.0589428 \times 10^{-13}$

region, measured by the parallel plate method [14] is shown in Fig. 3 for two quasi-isotherms. The density is expressed in amagat units. The agreement is within the experimental uncertainty.

## 5.2. Outside the Critical Isochore

To perform the analysis of the values of the singular part of the thermal conductivity along isochores different from the critical isochore, we used the concept of the spinodal curve. Following this concept, a thermodynamic property for instance  $C_v$ , which is considered on isochores different from the critical isochore, shows a similar singularity when the temperature approaches an asymptotic value which is a characteristic of this isochore  $T_s(\rho)$ . This concept was suggested by Benedeck [15], who assumes that the singularity on the noncritical isochore, when  $T \rightarrow T_s(\rho)$ , is the same as the singularity along the critical isochore, when  $T \rightarrow T_c$ . The curve  $T_s(\rho)$  is called the spinodal curve. Several developments of this concept were used for the description of properties near the critical point and near the range of stability of the monophase [16, 17].

With the concept of the spinodal curve, the singular part of the thermal conductivity is described by the relation

$$\Delta\lambda_s(\rho, T) = \lambda_s \left| \frac{T - T_s(\rho)}{T_c} \right|^{-\Psi} \quad (13)$$

where  $\lambda_s$  is the amplitude of the divergence of the singular part of the thermal conductivity,  $\Psi$  is the corresponding critical exponent, and  $T_s(\rho)$  is the

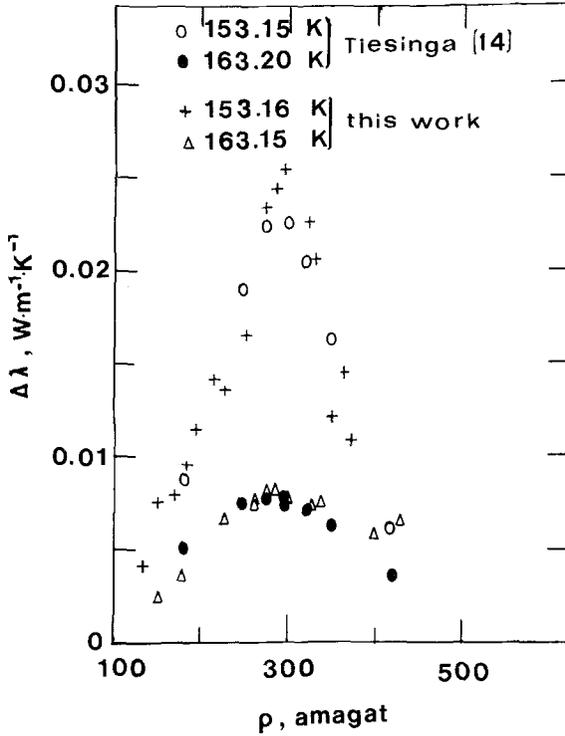


Fig. 3. Comparison of calculated thermal conductivity data with experimental ones obtained by the flat plate apparatus.

equation of the spinodal curve. A correlation for the spinodal curve was reported in Ref. 16:

$$T_s(\rho) = T_c \left( 1 - X \left| \frac{\rho - \rho_c}{\rho_c} \right|^{1/\beta} \right) \quad (14)$$

where  $X = B_s^{-1/\beta}$  and  $B_s$  is the amplitude of the spinodal curve.

As shown in Refs. 18 and 19,  $B_s$  is related to  $B_c$  by

$$\left( \frac{B_c}{B_s} \right)^{1/\beta} = \left( 1 - \left( \frac{\gamma - 2\beta}{\gamma(1 - 2\beta)} \right)^{-1/2\beta} \right)^{-1} \quad (15)$$

where  $B_c$  is the amplitude of the coexistence curve. Along the critical isochore, when  $\rho = \rho_c$ ,  $T_s(\rho) = T_c$  and Eq. (13) is identical to the equation of the scaling law along the critical isochore. We must remark that Eq. (14) is limited to a range close to the critical points. In Ref. 17, the experimental

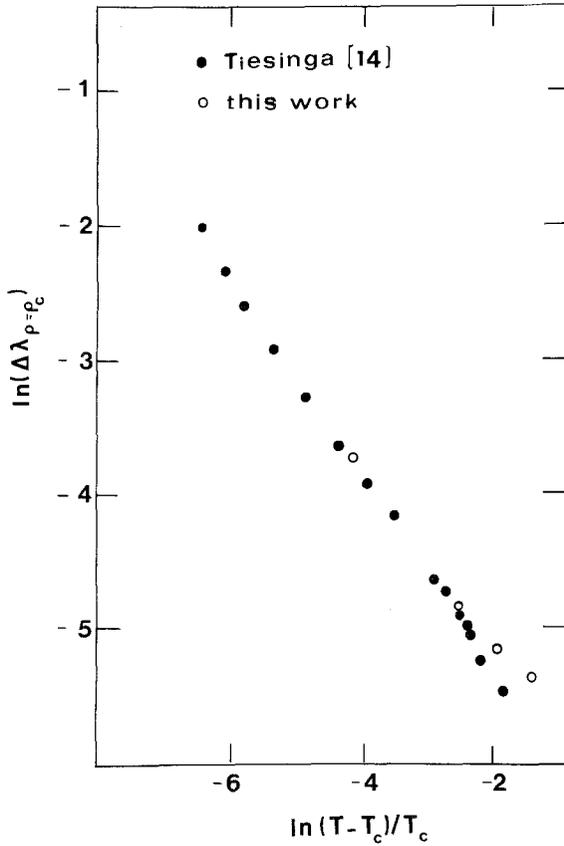


Fig. 4. Variation of the thermal conductivity (in  $W \cdot m^{-1} \cdot K^{-1}$ ) of argon along the critical isochore.

data on viscosity were analyzed over larger ranges of temperature and pressure by using another relation for the spinodal. In this case the equation of the spinodal curve is very similar to that of the coexistence curve. The treatment of the data of the singular part of the thermal conductivity by the method of the spinodal was carried along 10 isochores in the density range from  $485.1$  to  $585.1 \text{ kg} \cdot \text{m}^{-3}$ . The coefficients which have been used in Eq. (15) are the following:

$$B_c = 1.63 \quad [20]$$

$$\beta = 0.340 \quad [20]$$

$$\gamma = 1.208 \quad [3]$$

The values of the singular part of the thermal conductivity along the following isochores—485.1, 495.1, 505.1, 515.1, 525.1, 535.1, 545.1, 555.1, 565.1, 575.1, and 585.1  $\text{kg} \cdot \text{m}^{-3}$ —and the following isotherms—188.14, 173.14, 163.15, and 153.16 K—were represented by a single equation:

$$10^3 \Delta\lambda(\rho, T) = 1.858 \left| \frac{T - T_s(\rho)}{T_c} \right|^{-0.61} \quad (16)$$

These results are plotted in Fig. 5, which shows that almost all the isochores fall on a single curve. The small deviations which are observed between the experimental points and the curve can be explained by the errors in the estimation of the singular part of the thermal conductivity and by the presence of a slight asymmetry of the critical isochore. If we take into account the asymmetry factor by multiplying the left member of Eq. (13) by  $(\rho/\rho_c)^{a_x}$ , we can decrease the deviations with respect to the experimental data, but they are not entirely eliminated. The value of  $a_x$  which gives the best agreement with the experimental data was found to be 0.33.

Another method which is also based on the equation of the spinodal and allows description of the singularity in large intervals of pressure and

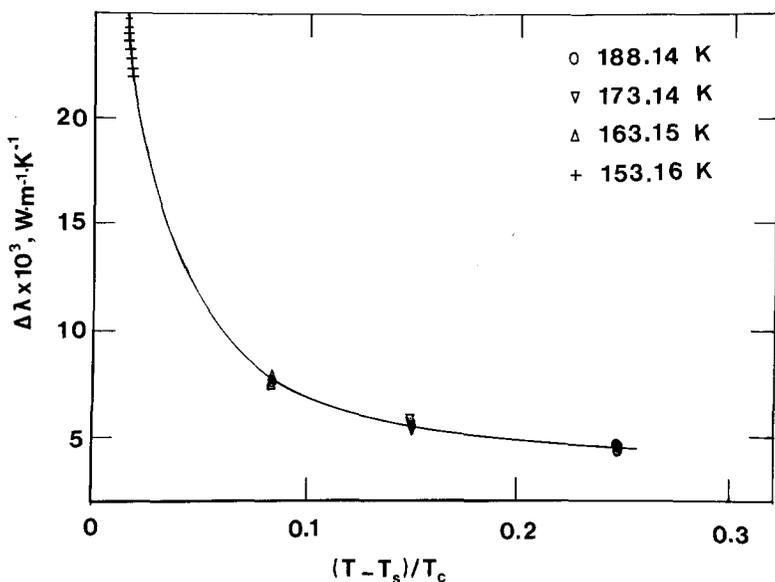


Fig. 5. Variation of the singular part of thermal conductivity as a function of the quantity  $(T - T_s)/T_c$ .

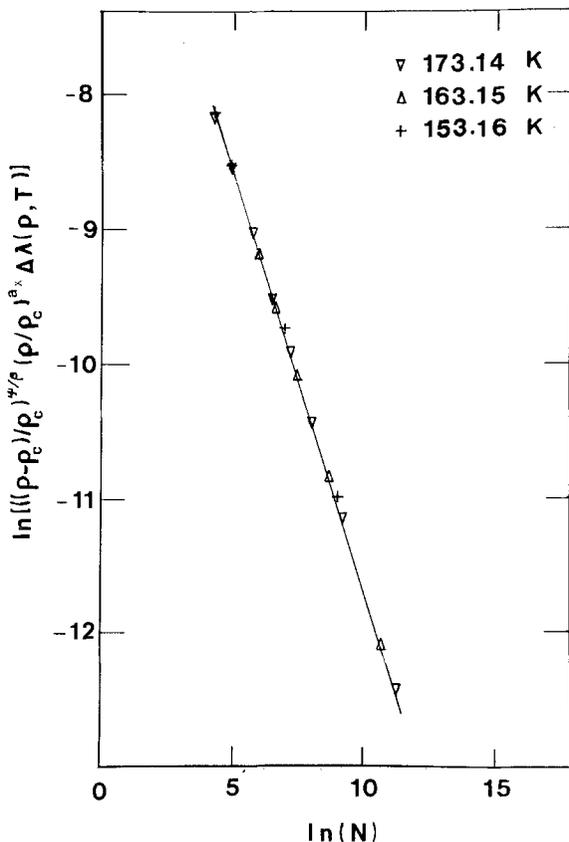


Fig. 6. Variation of the reduced singular part of thermal conductivity as a function of the quantity  $\ln N$ , where  $N = (Z + Z_0)/Z_0$ .

temperature around the critical point, takes into account the singularity on the spinodal [21]. The main relation is

$$\Delta\lambda(\rho, T) = \lambda_s Z^{-\phi} \tag{17}$$

where  $Z = \left(\frac{T - T_c}{T_c}\right) + K \left(\left(\frac{\rho - \rho_c}{\rho_c}\right) + (B_c - 1) \left(\frac{T - T_c}{T_c}\right)\right)^{1/\beta}$  (18)

$$K = \left(\frac{F}{(2 - F) B_c}\right)^{1/\beta} \tag{19}$$

$$F = \frac{\rho_1^c - \rho_v}{\rho_1^s - \rho_v} = \text{const.} \tag{20}$$

where  $\lambda_s$  is the amplitude of the singular part of the thermal conductivity,  $\phi$  the corresponding critical exponent,  $B_c$  the amplitude of the coexistence curve,  $\rho_1^c$  the density of the liquid on the coexistence curve,  $\rho_1^s$  the density of the liquid on the spinodal curve, and  $\rho_v$  the density of the vapor on the coexistence curve. The analysis of the results by this method leads to practically the same conclusion as in the previous case [16, 17]. In the density interval from 485.1 to 585.1 kg · m<sup>-3</sup>, Eq. (17) reads

$$10^3 \Delta\lambda(\rho, T) = 1.887Z^{-0.61} \quad (21)$$

In the determination of the singular part of the thermal conductivity, which was analyzed by various methods [16, 17, 21], the value of the scaling law of the thermal conductivity which takes into account the asymmetry factor was determined. The results are shown in Fig. 6. The curve plotted in this figure was approximated by the least-squares method.

$$\ln \left( \left| \frac{\rho - \rho_c}{\rho_c} \right|^{\phi/\beta} \left( \frac{\rho}{\rho_c} \right)^{a_x} \Delta\lambda(\rho, T) \right) = A + B \ln N + C(\ln N)^2 + D(\ln N)^3 \\ + E(\ln N)^4 + F(\ln N)^5 + G(\ln N)^6 \quad (22)$$

where  $N = (Z + Z_0)/Z_0$  and  $Z_0$  is the value of  $Z$  on the coexistence curve. For  $Z_0$  we select the value 0.2423 [3]. The coefficients of Eq. (22) are the following:

$$\Psi = 0.62$$

$$a_x = 0.33$$

$$\beta = 0.34$$

$$A = -6.567317$$

$$B = -1.334088$$

$$C = 0.850742$$

$$D = -0.265166$$

$$E = 0.037421$$

$$F = -0.002505$$

$$G = 6.462425 \cdot 10^{-5}$$

## 6. CONCLUSION

Experimental data on the thermal diffusivity of argon were reported along five isotherms. Reasonable agreements were found between

experimental and calculated thermal diffusivity and thermal conductivity data. However, the comparison between experimental and calculated data is hindered by large uncertainties on the quantities which are used in the different approximations.

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