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Thermal Conductivity of Argon in the Temperature Range 107 to 423 K[1]

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The thermal conductivity of argon between 107 and 425 K has been measured in a transient hot-wire instrument. The results in the limit zero density have been employed to assess the accuracy of the instrument using exact kinetic theory expressions and has been found to be better than $\pm 0.5\%$. The data at elevated densities are employed to examine the applicability of the modified Enskog theory in the gaseous phase and the hard-sphere theory in the liquid phase.

KEY WORDS: argon; hard-sphere theory; modified Enskog theory; thermal conductivity; transient hot-wire technique.

1. INTRODUCTION

As part of a continuing programme of high-precision measurements of the thermal conductivity of dense fluids, we have developed a new instrument for operation over the temperature range 80–450 K at pressures up to 30 MPa. The present paper describes the first application of the new instrument for measurements of the thermal conductivity of argon.

Despite the fact that fluid argon has been studied before over much of this temperature range, there are several reasons why this new study has been performed. First, the monatomic nature of argon and the existence of accurate viscosity data for the gas above room temperature enable us to use exact kinetic theory results to confirm the accuracy of the measurements. Second, the accuracy thereby confirmed ($\pm 0.5\%$) is

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superior to that achieved in earlier work at low temperature [1–3]. Third, the accuracy of measurements in the gas phase below room temperature is better than that achieved in the viscosity [4] so that the thermal conductivity data for the dilute gas provide a more stringent test of intermolecular pair potentials for argon. Finally, the results for elevated densities enable us to assess various semiempirical methods for the calculation of the thermal conductivity.

2. EXPERIMENTAL

The transient hot-wire instrument employed for the measurements has been described in detail elsewhere [5]. The only difference between the arrangement employed here and that described earlier is the replacement of the 10-μm-diameter platinum wires by a set with a nominal diameter of 7 μm, in order to improve the signal-to-noise ratio of the electrical measuring circuit. The characteristics of the wires employed for the present measurements are collected in Table I. The argon used for the measurements was supplied by Ar Liquido (Lisbon) and had a purity in excess of 99.999%. The density of argon and its heat capacity required in the reduction of the experimental data were taken from the IUPAC equation of state [6]. It is important to record here [7, 8] that in none of the experimental runs leading to the results reported here was any curvature observed in the line of temperature rise against the logarithm of time at the level of $\pm 0.06\%$. On this basis, it can be asserted that radiation absorption in the fluid makes a negligible contribution to the reported thermal conductivity [8] and that accounting for other random errors of measurement the estimated accuracy of the data is one of $\pm 0.5\%$.

4. RESULTS

Tables II–VII list the experimental data for the six isotherms studied at 107.15, 174.15, 225.00, 308.15, 378.15, and 429.15K.

At the lowest isotherm the measurements extend into the liquid phase. The tables include the thermal conductivity at the reference state $\lambda(T_r,\rho_r)$ [8] as well as the thermal conductivity corrected to a nominal temperature T_{nom}.

Table I. Characteristics of the Thermal Conductivity Cells

Cell diameter	10.00 ± 0.011 mm
Platinium-wire radius	6.80 ± 0.05 μm
Long-wire length at 293 K	169.13 ± 0.02 mm
Short-wire length at 293 K	68.72 ± 0.02 mm

Table II. Thermal Conductivity of Gaseous Argon at $T_{nom} = 174.15$ K

P (MPa)	ρ_r (kg·m^{-3})	T_r (K)	$\lambda(T_r, \rho_r)$ (mW·m^{-1}·K^{-1})	$\lambda(T_{nom}, \rho_r)$ (mW·m^{-1}·K^{-1})
0.3768	10.646	173.014	11.255	11.328
1.2380	36.595	172.509	11.856	11.961
1.5136	45.513	172.114	12.122	12.252
1.8240	55.836	171.954	12.264	12.405
2.0307	62.832	172.049	12.397	12.532
2.3058	72.854	171.415	12.003	12.778
2.5130	80.055	171.890	12.846	12.991
2.8571	92.678	172.202	13.015	13.140
3.1188	103.077	171.970	13.440	13.580
3.4082	115.049	171.767	13.628	13.781

The correction was applied in the form

$$\lambda(T_{nom},\rho_r) = \lambda(T_r,\rho_r) + \left(\frac{\partial\lambda}{\partial T}\right)_{\rho_r} (T_{nom} - T_r) \tag{1}$$

In the case of gaseous isotherms $(\partial\lambda/\partial T)_{\rho_r}$ has been deduced from the correlations given by Kestin et al [13] with the additional assumption that

$$\left(\frac{\partial\lambda}{\partial T}\right)_{\rho_r} = \left(\frac{\partial\lambda}{\partial T}\right)_{\rho=0} \tag{2}$$

Table III. Thermal Conductivity of Gaseous Argon at $T_{nom} = 225.00$ K

P (MPa)	ρ_r (kg·m^{-3})	T_r (K)	$\lambda(T_r, \rho_r)$ (mW·m^{-1}·K^{-1})	$\lambda(T_{nom}, \rho_r)$ (mW·m^{-1}·K^{-1})
0.7200	15.842	221.624	14.066	14.258
0.9956	22.063	221.317	14.255	14.465
1.3401	29.927	221.204	14.323	14.539
1.7545	39.560	221.010	14.638	14.866
2.2014	49.980	221.476	14.817	15.018
2.3401	53.618	220.299	14.969	15.237
2.7880	64.876	219.228	15.220	15.549
3.2358	75.776	219.815	15.413	15.709
3.6151	84.415	221.707	15.657	15.845
4.0285	94.970	221.548	16.039	16.236
4.4419	105.599	221.584	16.505	16.700
4.8898	117.396	221.507	16.661	16.860
5.3377	129.106	221.800	17.083	17.266

Table IV. Thermal Conductivity of Gaseous Argon at $T_{nom} = 308.15$ K

P (MPa)	ρ_r (kg·m^{-3})	T_r (K)	$\lambda(T_r, \rho_r)$ (mW·m^{-1}·K^{-1})	$\lambda(T_{nom}, \rho_r)$ (mW·m^{-1}·K^{-1})
1.1334	17.756	308.519	18.539	18.520
1.8224	28.671	308.285	18.769	18.762
2.7179	42.979	308.032	19.118	19.124
3.3725	53.487	308.041	19.351	19.357
4.1302	65.748	307.903	19.652	19.664
4.7851	76.419	307.749	19.958	19.977
5.6119	89.864	307.874	20.273	20.287
6.3560	102.018	307.949	20.612	20.622
6.9210	111.319	307.887	20.838	20.851
7.6307	123.065	307.754	21.163	21.183
8.0926	130.709	307.712	21.354	21.375
8.6440	139.852	307.656	21.714	21.738
8.6439	139.860	307.639	21.657	21.683
9.2162	149.304	307.685	21.997	22.020
9.2163	149.319	307.662	22.002	22.026

For the liquid isotherms $(\partial\lambda/\partial T)_{\rho_r}$ has been estimated from the wide-ranging correlation given by Hanley et al [10].

For any event none of these corrections amounts to more than $\pm 1\%$ so that the contribution of uncertainties in the correction to the error in the reported thermal conductivity is estimated to be $\pm 0.1\%$ in the worst case.

Table V. Thermal Conductivity of Gaseous Argon at $T_{nom} = 378.15$ K

P (MPa)	ρ_r (kg·m^{-3})	T_r (K)	$\lambda(T_r, \rho_r)$ (mW·m^{-1}·K^{-1})	$\lambda(T_{nom}, \rho_r)$ (mW·m^{-1}·K^{-1})
0.9624	12.109	382.207	21.961	21.779
1.5825	19.933	381.977	22.112	21.940
2.2163	27.942	381.795	22.371	22.207
2.7882	35.160	381.826	22.516	22.350
3.6139	45.626	381.516	22.787	22.635
4.2351	53.432	381.832	23.047	22.881
4.9241	62.157	381.681	23.371	23.212
5.6131	72.408	374.072	23.065	23.248
6.4399	81.319	381.524	23.699	23.548
7.1288	90.038	381.382	23.950	23.804
8.0246	101.341	381.291	24.140	23.998
8.9204	112.655	381.104	24.351	24.218
9.3681	118.380	380.790	24.523	24.405
10.057	127.023	380.763	24.763	24.655

Table VI. Thermal Conductivity of Gaseous Argon at $T_{nom} = 429.15$ K

P (MPa)	ρ_r (kg · m^{-3})	T_r (K)	$\lambda(T_r, \rho_r)$ (mW · m^{-1} · K^{-1})	$\lambda(T_{nom}, \rho_r)$ (mW · m^{-1} · K^{-1})
0.6173	6.949	426.707	24.051	24.156
1.0996	12.385	426.374	24.017	24.137
1.7197	19.370	426.224	24.268	24.394
2.3743	26.747	426.002	24.492	24.627
2.9255	32.952	425.895	24.502	24.642
3.6140	40.553	427.226	24.872	24.955
4.6136	51.923	425.647	25.062	25.213
5.3095	59.735	425.494	25.278	25.436
6.0053	67.519	425.453	25.405	25.564
6.6111	74.215	425.788	25.622	25.767
7.3010	81.872	425.863	25.902	26.044
8.0589	90.304	425.746	26.080	26.227
8.9890	100.702	425.295	26.182	26.345
9.5747	107.142	425.377	26.439	26.601

For the purpose of interpolation along an isotherm the experimental data have been represented by a finite polynomial of the form

$$\lambda = a_0 + a_1 \rho + a_2 \rho^2 \tag{3}$$

The values of the coefficients a_i which secure an optimum representation of the data are listed in Table IX. The maximum deviation form these correlations is 0.9%, whereas the maximum standard deviation is

Table VII. Thermal Conductivity of Liquid Argon at $T_{nom} = 107.15$ K

P (MPa)	T_r (K)	ρ_r (kg · m^{-3})	$\lambda(T_r, \rho_r)$ (mW · m^{-1} · K^{-1})	$\lambda(T_{nom}, \rho_r)$ (mW · m^{-1} · K^{-1})
0.7907	107.145	1262.31	100.07	100.07
0.7907	107.066	1262.90	100.94	100.95
1.6589	107.500	1264.18	101.32	101.30
2.4097	107.403	1268.69	101.89	101.87
3.1332	107.430	1272.07	102.54	102.52
3.7808	107.502	1274.70	103.32	103.30
4.4075	107.175	1279.95	103.82	103.82
5.1987	107.937	1278.41	104.28	104.23
5.8199	107.937	1285.09	105.13	105.08
6.6604	107.207	1289.97	105.58	105.58
8.0246	107.815	1291.93	106.52	106.48

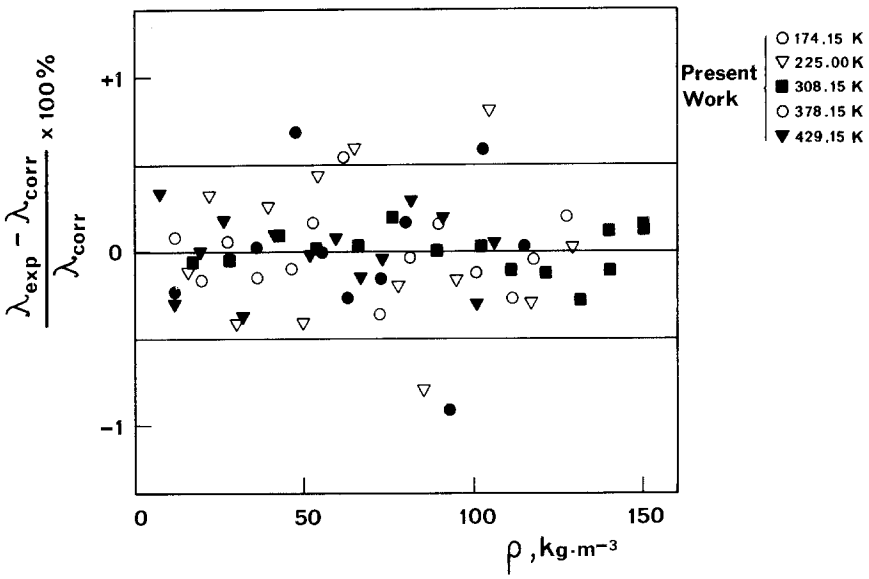

Fig. 1. Departures of the experimental points from Eq. (3). (●) 174.15 K; (▽) 225.00 K; (■) 308.15K; (○) 378.15 K; (▼) 429.15 K.

$\pm 0.5\%$, which is commensurate with the estimated precision of the experimental results. Figure 1 contains plots of the deviation of the present data from the correlation of Eq. (3) and Table VIII. Figure 2 includes a comparison with the results of earlier work [11]. The comparison with earlier work is limited to studies at essentially the same temperatures as those studied here in order to obviate substantial temperature corrections. An analysis of Figs. 1 and 2 shows that the agreement between the two sets of data is consistent with their mutual uncertainty.

Table VIII. Coefficients of Eq. (3)

T_{nom} (K)	$a_0 \pm \sigma_{a0}$ (mW·m^{-1}·K^{-1})	$a_1 \pm \sigma_{a1}$ (μW·m^2·kg^{-1}·K^{-1})	$a_2 \pm \sigma_{a2}$ (nW·m^5·kg^{-2}·K^{-1})	σ (mW·m^{-1}·K^{-1})
174.15	11.11 ± 0.09	23.3 ± 2.8	—	0.067
225.00	13.94 ± 0.09	21.0 ± 2.9	36 ± 20	0.080
308.15	18.16 ± 0.03	20.6 ± 0.9	34 ± 5	0.030
378.15	21.42 ± 0.06	28.4 ± 2.0	-26 ± 14	0.059
429.15	23.90 ± 0.05	25.6 ± 2.2	—	0.063

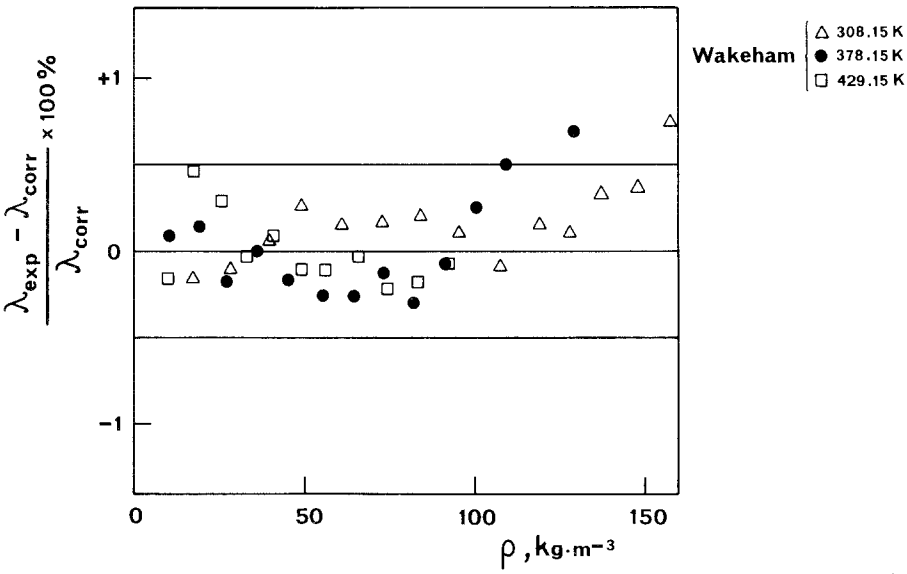

Fig. 2. Departures of data obtained by Haran *et al.* [11] from Eq. (3). (△) 308.15 K; (●) 378.15 K; (□) 429.15 K.

4. ANALYSIS OF THE DATA

4.1. The Zero-Density Limit

To the experimental data in the gaseous phase along each isotherm we have applied the statistical analysis described in detail elsewhere [12] in order to determine best estimates of the first two coefficients in the density expansion of the thermal conductivity

$$\lambda = \lambda_0 + C_1\rho + C_2\rho^2 + \cdots \tag{4}$$

The derived values, together with their statistical uncertainty, are listed in Table IX. The same table includes zero-density viscosity data η_0 at the same temperatures. Above room temperature we have taken the viscosity from the work of Kestin et al. [13] (which have estimated an uncertainty of $\pm 0.2\%$), whereas below room temperature we have employed the data of Clarke and Smith [4] (which have an associated uncertainty of $\pm 1\%$).

These data have been used to calculate experimental values of the Eucken factor:

$$\mathrm{Eu} = \frac{2\lambda_0 M}{3R\eta_0 F} \tag{5}$$

Table IX. Data Analysis of the Thermal Conductivity of Gaseous Argon

T_{nom} (K)	$\lambda_0 + \sigma_{\lambda 0}$ (mW·m^{-1}·K^{-1})	$C_1 \pm \sigma_{C1}$ (μW·m^2·kg^{-1}·K^{-1})	E_u	% deviation from 2.500	η_0 (μPa·s)	λ_0^{AC} (mW·m^{-1}·K^{-1})	%[a]	C_1^{MET} (μW·m^2·kg^{-1}·K^{-1})
174.15	11.10 ± 0.05	23.7 ± 0.6	2.542	+1.71	13.99 [4]	11.13	+0.31	17.1
225.00	13.82 ± 0.05	26.6 ± 0.6	2.484	−0.65	17.78 [4]	13.82	$+0.00_4$	17.8
308.15	18.03 ± 0.02	25.4 ± 0.3	2.486	−0.56	23.25_8 [13]	18.00	−0.16	18.3
378.15	21.51 ± 0.03	24.5 ± 0.4	2.499	−0.05	27.53_1 [13]	21.57	+0.30	24.8
429.15	23.91 ± 0.03	25.3 ± 0.5	2.497	−0.13	30.61_5 [13]	23.86	−0.22	26.1

[a] Defined as $(\lambda_0^{AC} - \lambda_0^{exp})/\lambda_0^{exp} \times 100\,\%$, where λ_0^{AC} is the theoretical value of the thermal conductivity calculated from the Aziz–Chen potential [15].

for which the kinetic theory predicts a value

$$\mathrm{Eu} = 2.500 \text{ (exactly)} \tag{6}$$

In Eq. (5), F is a higher-order correction factor of the kinetic theory which is readily estimated [14]. Table IX indicates that the experimental values for the Eucken factor include within their range of uncertainty the theoretical result and, thereby, confirms the accuracy of the present measurements as one of $\pm 0.5\%$.

Below room temperature, the present thermal conductivity data have an estimated accuracy superior to that of the reported viscosity data [4]. It is therefore of some interest to examine just how well the latest intermolecular pair potential proposed for argon, which has been partly based on viscosity data, reproduces the present results. Accordingly we have employed the latest potential proposed by Aziz and Chen [15] to evaluate the thermal conductivity by standard methods [14]. Table IX includes the calculated values of λ_0. It can be seen that there is a very good agreement with the experimental values of the thermal conductivity quoted in this paper as well as with the high-temperature viscosity data [13]. The deviation for low-temperature viscosity [4] seems to indicate that these results have an uncertainty of about 2%.

4.2. The Moderately Dense Gas

There exists no rigorous theory for the description of the behavior of the transport properties of fluids in the dense gaseous region.

However, an empirical modification of the Enskog theory of a dense hard-sphere fluid [16] has some acceptance as a useful predictive procedure in this regime. According to the modified Enskog theory

$$\lambda = \lambda_0(1/b\rho\chi + 1.20 + 0.755 b\rho\chi)\, b\rho \tag{7}$$

where

$$b = (B + T\, dB/dT)(1/M) \tag{8}$$

and

$$\chi = \left[\frac{1}{R}\left(\frac{\partial(PV)}{\partial T}\right) - 1\right] \Big/ b\rho \tag{9}$$

Here, M is the molar mass of the gas, B its second virial coefficient, V the molar volume of the gas, P the pressure, and R the universal gas con-

stant. For the range of densities of interest here it is sufficient to use the low-density limit of these expressions for which

$$\lambda = \lambda_0 + C_1 \rho \tag{10}$$

where the first density coefficient, C_1, is given by

$$C_1 = -[(\gamma/b) - 1.2\, b]\lambda_0 \tag{11}$$

in which

$$\gamma = \left(C + T\frac{dC}{dT}\right)\frac{1}{M^2} \tag{12}$$

We have evaluated the density coefficient C_1 at each of the temperatures studied experimentally using the second virial coefficients derived from the correlation of the extended law of corresponding states [9] and third virial coefficients from the work of Dymond and Alder [17].

The tabuled values are included in Table IX. It is clear that at the higher temperatures the prediction of the modified Enskog theory for C_1 is in rather close agreement with experiment. This must be contrasted with a molecular dynamics simulation test of the modified Enskog theory for a square-well potential which indicated that the method led to a poor prediction of the first density coefficient [18]. At low temperatures the modified Enskog theory is less successful but may be acceptable for some purposes.

A second feature of the behavior of moderately dense, real gases which has been reported in a number of occasions is the temperature independence of the excess thermal conductivity.

$$\Delta\lambda = \lambda(\rho, T) - \lambda_0(T) \tag{13}$$

The present experimental data permit us to examine this result with a higher degree of precision than was possible hitherto. Figure 3 displays a plot of excess thermal conductivity for the five isotherms studied in this work. It can be seen that although there is a very weak temperature dependence, over the wide range of temperatures studied in this work the effect assuming a temperature independent excess property upon the total thermal conductivity of the fluid amounts to, at most, 1 %. The assumption of temperature independence of $\Delta\lambda$ far from the critical point is therefore extremely valuable for predictive purposes.

It should be stated that the result described above is most certainly not consistent with either the original Enskog theory for hard spheres or the modified form of the theory. The former predicts that it is the ratio λ/λ_0 which is temperature independent, whereas the latter leads to the relatively

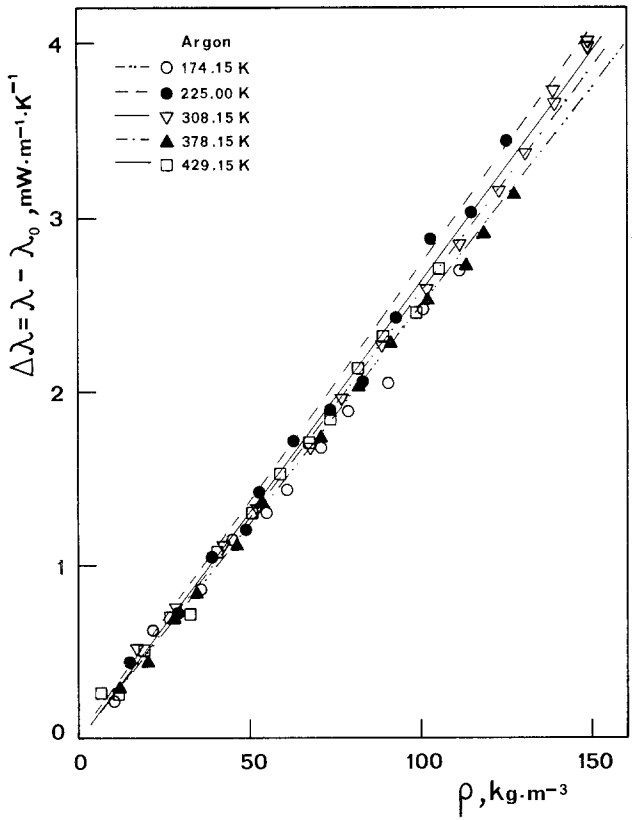

Fig. 3. The excess thermal conductivity of argon as a function of density for the isotherms. (○) 174.15 K; (●) 225.00 K; (▽) 308.15 K; (▲) 378.15 K; (□) 429.15 K.

strong temperature dependence of C_1 and therefore $\Delta\lambda$ revealed in Table IX. The reason for this widely observed phenomenon therefore remain obscure. The more rigorous treatment of the first density coefficient of transport properties given by Rainwater [19, 20] and Friend and Rainwater [21] has not yet been tested against the present data.

4.3. The Liquid Phase

Only one isotherm has been studied within the liquid phase of argon in this work, although further systematic studies in this region are planned. The present results do, however, allow us to carry out one limited test of the most successful theory of liquid phase transport which is based on the Van der Waals model of the fluid [22].

This model enables the properties of a real monatomic fluid to be represented by those of a rigid sphere system at sufficiently high temperatures and densities. Dymond [22] has composed the results of the Enskog theory of hard spheres with the results of computer simulations of dense fluids to yield expressions for the viscosity and thermal conductivity in the form

$$\eta^* = \frac{\eta}{\eta_0}\left(\frac{V}{V_0}\right)^{2/3} = F_\eta\left(\frac{V}{V_0}\right) \tag{14}$$

and

$$\lambda^* = \frac{\lambda}{\lambda_0}\left(\frac{V}{V_0}\right)^{2/3} = F_\lambda\left(\frac{V}{V_0}\right) \tag{15}$$

in which V_0 is the close packed volume of the hard-sphere system. Dymond [22] has given explicit equations for F_η and F_λ valid for $V/V_0 > 1.5$, below which the hard-sphere system reveals a metastable solid phase.

In applications to real monatomic fluids V_0 is allowed to be a disposable parameter which is weakly temperature dependent, reflecting the finite steepness of repulsive interaction in real fluids.

Easteal and Woolf [23, 24] have applied an analysis of this type to the viscosity data for argon and have derived values of V_0 from them. They concluded that the hard-sphere theory was not entirely adequate for a description of all the liquid phase properties of argon at the lowest temperatures.

The analysis of Easteal and Woolf could not consider the thermal conductivity of the liquid owing to the absence of data. We now take the opportunity to include this property in the analysis and have therefore deduced the value of V_0 which secures the best representation of a thermal conductivity along the isotherm $T = 107$ K using Dymond's form for F_λ. Figure 4 contains a plot of the deviations of the data from this equation when V_0 is assigned the value $V_0 = 16.18\ \text{cm}^3 \cdot \text{mol}^{-1}$. Within the range of densities covered by the present measurements the hard-sphere theory represents the data with an error of no more than $\pm 0.8\%$. However, there is some evidence that the deviation is systematic, indicating that the hard-sphere form of F_λ is not quite appropriate.

When the value of V_0 deduced from the analysis of the thermal conductivity data is employed to evaluate the viscosity of argon at the same temperature through Eq. (14) and the expressions of Dymond [22], the results deviate by some 20% from the experimental data reported by Haynes [25]. The only possible conclusion from these results is that the hard-sphere theory is inappropriate to describe consistently the behavior of

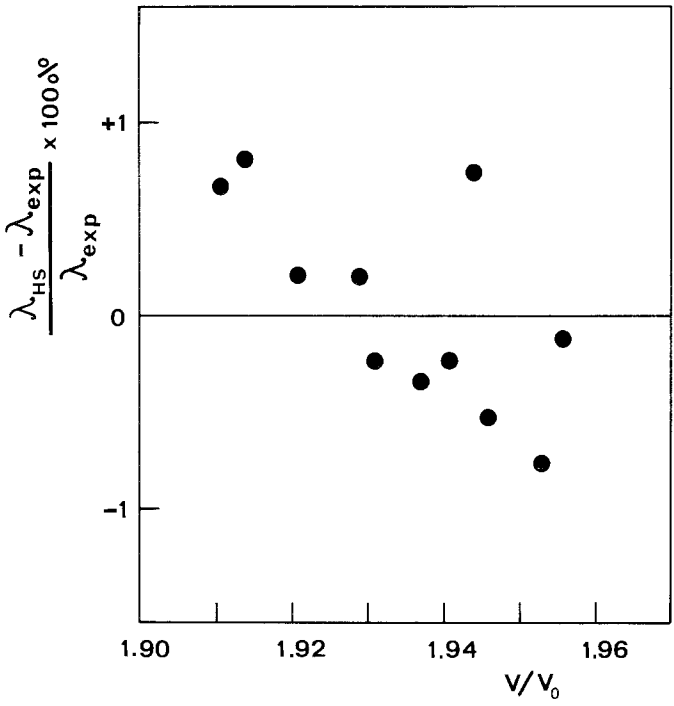

Fig. 4. Difference between the hard-sphere results and the experimental data for liquid argon thermal conductivity as a function of V/V_0.

the transport properties of real fluids at such low temperatures ($T/T_c < 0.71$) in accord with the findings of Easteal and Woolf [23, 24]. Presumably this is a consequence of the importance of attractive forces in this range of states since these are ignored in the rigid-sphere theory.

5. CONCLUSIONS

The new measurements of the thermal conductivity of argon reported in this paper extend the range of thermodynamic states for which accurate data are available. The accuracy of the data is confirmed by evaluation of the Eucken factor in the limit of zero density. In the same limit, the intermolecular pair potential for argon proposed by Aziz and Chen [15] provides a good description of the results.

At higher densities in the gaseous and liquid phases, the available semiempirical procedures for predicting the thermal conductivity prove less satisfactory. Further measurements on simple fluids such as argon are necessary in order to suggest means of working about their improvement.

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