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Thermal Transport Behavior of van der Waals Solids and Liquids in the Neighborhood of the Solid–Liquid Phase Transition[1]

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The thermal conductivity, λ, is one of the few transport coefficients which shows a relatively small change at the solid–liquid phase transition, and hence it is a property that can be used in comparing dynamic properties of both ordered and disordered systems. Although the discontinuity in λ can be accounted for largely by the difference in density, ρ, of solid and liquid at the phase transition, its volume dependence is examined more closely. The thermal diffusivity, which is known to dominate the dynamic structure factor of liquid argon, has been determined around the phase transition also; the sound velocity has been considered in addition. The results are discussed and a comparison is made with these properties in solid and liquid benzene and cyclohexane.

KEY WORDS: argon; benzene; cyclohexane; solid–liquid phase transition; thermal conductivity; thermal diffusivity; velocity of sound.

1. INTRODUCTION

The thermal conductivity, λ, and thermal diffusivity, $a = \lambda/\rho c_p$, are transport coefficients which change relatively little at the solid–liquid phase transition, and hence they might be useful in comparing dynamic properties of both ordered and disordered systems. More often, it has been suggested that the discontinuity in λ can be accounted for largely by the difference in density, ρ, of solid and liquid at the phase transition. This aspect is investigated by examining more closely the isothermal volume dependence of the thermal transport coefficients of solid and liquid argon,

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benzene, and cyclohexane, for which substances accurate and extensive thermodynamic and transport data along isotherms have become available up to very high pressures. In addition, the velocity of ultrasound, c, will be considered because the isothermal volume dependence of c in liquids closely resembles that of a [1].

In the next section the values of λ, a, and c of these substances in the solid and the liquid state at the triple point are compared, while in Section 3 the volume dependence of these properties of liquid and solid argon, benzene, and cyclohexane along isotherms is given. In the following section, the temperature dependence of a and λ in solid and liquid is treated, and some conclusions are given in the final section.

2. THERMAL TRANSPORT AND SOUND VELOCITY DATA AT THE TRIPLE POINT

The most straightforward way to investigate transport properties near the solid–liquid phase transition is to consider these properties of both solid and liquid at the triple point [2]. We have taken λ values from Slack's review on the thermal conductivity of nonmetallic substances, calculated the thermal diffusivity using ρ and c_p given by Rabinovitch et al. [3], and used the longitudinal sound velocity in the solidified and liquid rare gases [4, 5] to determine the ratios λ_1/λ_2, a_1/a_2, and c_1/c_2 for the latter, where the subscript 1 refers to the solid and the subscript 2 to the liquid near the triple point. The values of these ratios with the estimated uncertainties are given in table I.

The values of λ_1/λ_2 for benzene and cyclohexane at the triple point are from Slack [2], who also gives the densities, while c_p values for solid and liquid C_6H_6 and C_6H_{12} are listed in various handbooks [6, 7]. The heat capacity enables us to calculate the thermal diffusivity of these hydrocarbons and hence to determine a_1/a_2. The sound velocity in solid C_6H_6 at 279 K has been reported by Dobromyslov and Koshkin [8], and that in liquid C_6H_6 and C_6H_{12} at 280 K is obtained by linearly extrapolating the sound velocity measured by Takagi [9] in the temperature range 293 to 314 K. The extrapolated value for c of liquid C_6H_{12} at 280 K is in excellent agreement with that measured by Kreutzenbeck et al. [10]. These authors also measured the ultrasound velocity in solid C_6H_{12} near the melting point. The ratios c_1/c_2 for these hydrocarbons are also included in Table I, in the last column of which the percentage volume change at the triple point is given.

In general the ratios are substantially larger than one and of about the same size for both thermal conductivity and thermal diffusivity. The values given for argon are representative for krypton and xenon as well, but not

Table I. Ratio of the Thermal Conductivities and Diffusivities and Longitudinal Sound Velocities of Various Substances in the Solid (1) and in the Liquid (2) at the Triple Point[a]

Substance	T (K)	λ_1/λ_2	a_1/a_2	c_1/c_2	ΔV (%)
Ar [Kr, Xe]	84	1.6 ± 0.1 (1.2)	1.70 ± 0.15 (1.25)	1.58 ± 0.02 (1.1)	12.5
Ne	24.5	2.4 ± 0.3	3.1 ± 0.4	1.58	13.5
C_6H_6	279	1.7 ± 0.15 (1.15)	1.6 ± 0.2 (1.25)	1.8	11.2
C_6H_{12}	280	1.1 ± 0.1 (0.9)	1.1 ± 0.1 (1.0)	1.35	4.0

[a] The volume change upon melting is given also. The numbers in parentheses are the ratios of the physical quantity of the solid and that of a fictitious liquid which has the same density as the solid.

for neon, for which the ratios a_1/a_2 and λ_1/λ_2 are significantly larger. The author recently discussed these differences, and they have been ascribed to possible quantum effects in the thermal transport coefficients of neon [11]. The much smaller ratios for cyclohexane are an indication of the more liquid-like properties of the high-temperature plastic-crystalline phase of cyclohexane, C_6H_{12}–I. This is discussed in more detail in the next section.

3. VOLUME DEPENDENCE OF THE THERMAL TRANSPORT PROPERTIES AND THE SOUND VELOCITY IN SOLID AND LIQUID ALONG ISOTHERMS

Previously we reported [1, 12] that the thermal resistivity of a liquid depends linearly upon the relative expansion throughout a substantial fraction of the liquid range rather independently of the temperature. An example is given in Fig. 1, in which the λ^{-1} of compressed liquid argon at $T = 90$ and 100 K [3] is plotted as a function of the molar volume, V. We have used the linear λ^{-1} versus V relationship to determine the thermal conductivity of a fictitious liquid that has the same density as the solid argon near melting. The two arrows indicate the molar volumes at $T = 90$ K of solid and liquid argon on the melting and solidification curves, respectively [13]. From the result in Fig. 1 it follows that the value of λ of solid argon [14] is some 20% in excess of that of the fictitious liquid, contrary to Slack's findings [2], which indicated that the thermal conductivity of fictitious liquid argon near the triple point is very close to that of the solid. The numbers in parentheses in Table I are the ratio λ_1 to the thermal conductivity of fictitious liquid argon at 90 K with the same density as the solid upon melting.

In a similar way, we have determined the thermal diffusivity, a, and

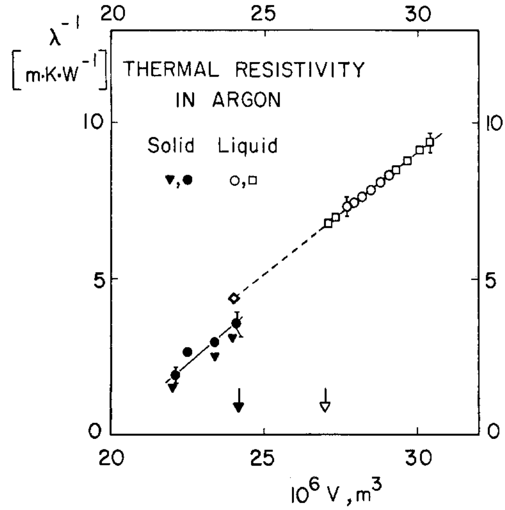

Fig. 1. Volume dependence of the thermal resistivity in solid and liquid argon along isotherms. The thermal conductivity and the density of solid and liquid argon at 75, 90, and 100 K have been reported in the literature at pressures up to 0.5 and 0.1 GPa, respectively [3, 14], at the temperatures indicated. The arrows refer to the molar volume of solid and liquid argon at $T = 90$ K [13]. The dotted line, which has the slope of the linear λ^{-1} vs V relationship observed in the liquid [1], has been used to determine the thermal resistivity of a fictitious liquid that has the same density as solid argon on the melting curve at $T = 90$ K (indicated by an open diamond).

the sound velocity, c, of fictitious liquid argon at $T = 90$ K, extrapolating the linear a vs V and c vs V relationships of liquid argon along the 90 K isotherm [1] as shown in Fig. 2. The longitudinal sound velocity in compressed solid argon at 90 K has been estimated from the limited amount of data available [4]; the sound velocities in liquid argon along the 90 K isotherm have been reported by Thoen et al. [15]. The values of c_1 and a_1 of solid argon on the melting curve are in excess of the corresponding quantities of a fictitious liquid by 10 and 25%, respectively (see also Table I). Hence the discontinuity in λ, a, and c at the solid–liquid phase transition of argon cannot be accounted for entirely by the difference in density of solid and liquid at the phase transition.

From the data presented in Fig. 1 it appears that the thermal resistivity of solid argon along an isotherm varies about linearly with the

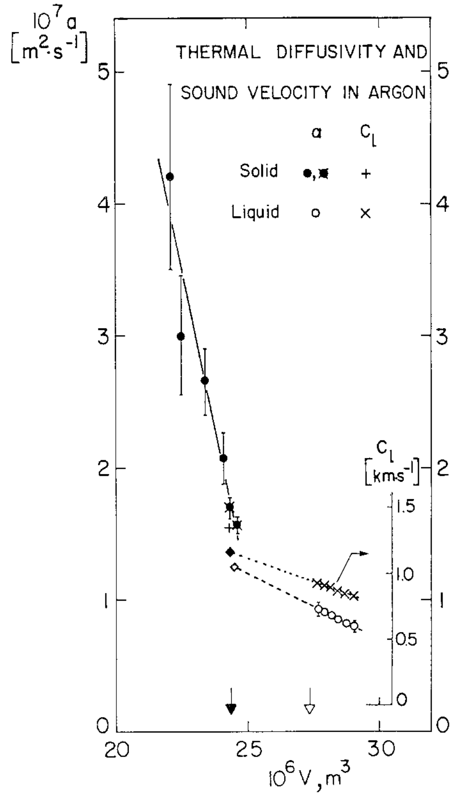

Fig. 2. Volume dependence of the thermal diffusivity and sound velocity in solid and liquid argon at $T = 90$ K. The thermal diffusivity, $a = \lambda/\rho c_p$, is obtained from λ and ρ used in Fig. 1 and the specific heat [3]. Values of c_p of solid argon at pressures exceeding 100 MPa have been estimated (see text). The velocity of sound of liquid argon along the 90 K isotherm has been reported [15]; the longitudinal sound velocity in solid argon has been estimated [4]. The arrows refer to the molar volume of solid and liquid argon at $T = 90$ K. The dotted lines, which are extensions of the linear a vs V and c vs V relationships observed in the liquid, enable us to determine the thermal diffusivity and the sound velocity in a fictitious liquid that has the same density as solid argon at $T = 90$ K, indicated by the open and the filled diamonds. The line through the data points of the thermal diffusivity of solid argon is drawn as a guide to the eye.

molar volume, with a slope that is somewhat steeper than that of the linear λ^{-1} vs V plot for liquid argon. Furthermore, λ^{-1} of solid argon at constant volume decreases at lower temperatures.

The corresponding states principle (CSP) has recently successfully been applied to the thermal conductivity of the monatomic liquids [16]; it is reasonable to expect that CSP will also be applicable to λ of the solidified heavy rare gases. Using the linear λ^{-1} vs V relationship of solid argon at 75 K in reduced units, the thermal conductivities of compressed hot Kr and Xe crystals can be predicted. To the best of our knowledge, these have not been measured, which in fact may be very difficult. Using the densities reported [3] and the Lennard-Jones parameters for these rare gases in order to obtain reduced quantities [16], values of $\lambda = 0.29$ and $0.25\ \mathrm{W \cdot m^{-1} \cdot K^{-1}}$ for solid krypton at 105 K and for solid xenon at 145 K, respectively, each at a pressure of 100 MPa, have been obtained with an estimated uncertainty of 15%. The thermal conductivity of compressed solid krypton is larger by about 40% than the λ of crystalline Kr at atmospheric pressure reported by Krupskii and Manzhelii [17].

The thermal diffusivity of solid argon at 90 K has been calculated using the values of ρ and λ reported at pressures up to 0.5 GPa [4, 14] and c_p at pressures up to 100 MPa [3]. By making use of the gradual change of the specific heat with volume at different temperatures of the solid, the values of c_p at higher pressures are obtained by extrapolation and they have been used to calculate the a values with the large error bars in Fig. 2. It is evident that the thermal diffusivity of solid argon along the 90 K isotherm increases very steeply upon contraction of the lattice; it should be noted, however, that in view of the rather large error bars, the volume dependence of the thermal diffusivity is not necessarily linear [11].

The thermal conductivity of liquid C_6H_6 and C_6H_{12} at pressures up to 0.33 GPa has been measured by Wakeham and co-workers [18], who also give the density. In Fig. 3 the thermal resistivity of liquid benzene at 310 and 320 K is plotted as a function of the molar volume, and the single linear relationship found has been used to obtain the value of λ for fictitious liquid C_6H_6 that has the same density as the solid near the melting curve. The double arrows in Fig. 3 indicate the molar volumes of solid and liquid benzene at 300 K and a pressure of about 70 MPa [19]. The thermal conductivity and heat capacity of solid C_6H_6 at pressures up to 2.4 GPa have been measured by Ross et al. [20], while densities have been reported at pressures up to 0.5 GPa [19]. The thermal resistivity of solid C_6H_6 at 300 K as a function of the molar volume is shown in Fig. 3, and within the limits of uncertainty the data satisfy a linear relationship between λ^{-1} and V, with a slope that is slightly steeper than that of the liquid, as was found in argon. It should be noted that the thermal conduc-

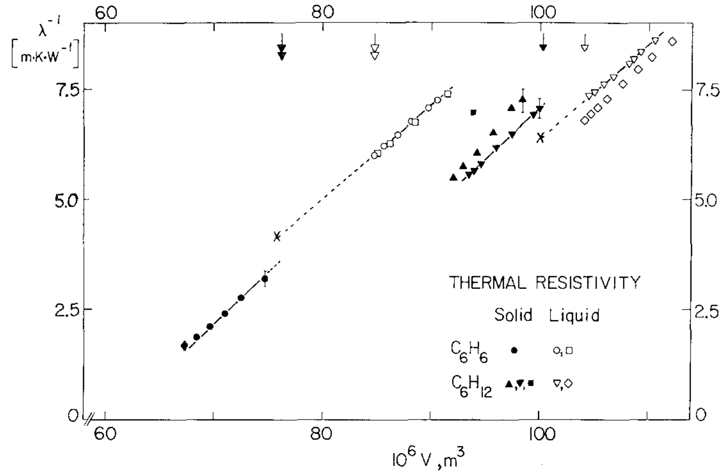

Fig. 3. Volume dependence of the thermal resistivity in solid and liquid benzene and cyclohexane along isotherms. The thermal conductivity and the density of solid and liquid C_6H_6 and C_6H_{12} have been reported at pressures up to 0.5 GPa [19–22]. The data on C_6H_6 are at $T = 300$ (●), 310 (○), and 320 K (□), respectively; the double arrows refer to the molar volume of solid and liquid C_6H_6 on the melting and the solidification curves at $T = 300$ K [19]. The data on cyclohexane are at $T = 186$ (■), 248 (▲), 323 (▼, ▽), and 361 K (◇), respectively; the arrows refer to the molar volume of C_6H_{12}-I and liquid C_6H_{12} on the melting and the solidification curves at $T = 323$ K. The crosses indicate the values of the thermal resistivity of a fictitious liquid that has the same density as solid C_6H_6 on the melting curve at 310 K and of C_6H_{12}-I on the melting curve at 323 K.

tivities of hot solid C_6H_6 and argon near the phase transition are of the same size indicating that the optical modes do not carry much heat. The (extrapolated) value of λ for solid C_6H_6 at the melting curve is in excess by about 15% of the thermal conductivity of fictitious liquid benzene at the same density (see Table I), similar to what has been found in argon.

Andersson [21] measured the thermal conductivity and heat capacity of C_6H_{12} at pressures up to 1.5 GPa. The thermal resistivity of C_6H_{12}-I at 248 and 323 K is given as a function of volume in Fig. 3 and the data satisfy a linear relationship between λ^{-1} and V. The thermal resistivity of C_6H_{12}-I at the I → II solid–solid phase transition at 186 K is shown also. The value of λ for C_6H_{12}-I near the melting point is smaller by more than a factor of two than that of solid C_6H_6 at similar thermodynamic conditions. The thermal resistivity of liquid cyclohexane at 324 and 361 K is plotted versus V and the linear relationship found at 324 K has been used to obtain the value of λ for fictitious liquid C_6H_{12} that has the same density as the solid near the melting curve. The arrows in Fig. 3 indicate the molar volumes of solid and liquid cyclohexane at 323 K [22] and a pressure of about 90 MPa. The ratio of λ for solid C_6H_{12}-I and fictitious liquid C_6H_{12} at the same density is smaller than one (see Table I).

The thermal diffusivity of solid and liquid benzene at temperatures of 300 and 310 K is given as a function of the molar volume in Fig. 4. The data for the solid are from Ross et al. [20]; the heat capacity of C_6H_6 around the phase transition and of liquid C_6H_6, taken from different handbooks [6, 18], together with the data for λ and ρ reported by Li et al. [18], enabled us to calculate the thermal diffusivity of liquid C_6H_6. From the results in Fig. 4 it is evident that the thermal diffusivity of solid C_6H_6 at the melting curve is by about 25% in excess of the value of a for fictitious liquid benzene with the same density as the solid near melting extracted from the linear relationship between a and V found for liquid C_6H_6 (see Table I). As in argon, the thermal diffusivity of solid C_6H_6 along the 300 K isotherm increases very steeply upon contraction of the lattice; the values of a are of the same magnitude as those of argon.

The thermal diffusivity of solid and liquid cyclohexane at 323 K is plotted vs V in Fig. 4 using the data reported by Andersson [21]. The most striking feature is the absence of any discontinuity in a by passing through the solid–liquid phase transition. Furthermore, the thermal diffusivity of both solid and liquid C_6H_{12} is substantially smaller than that of solid and liquid C_6H_6.

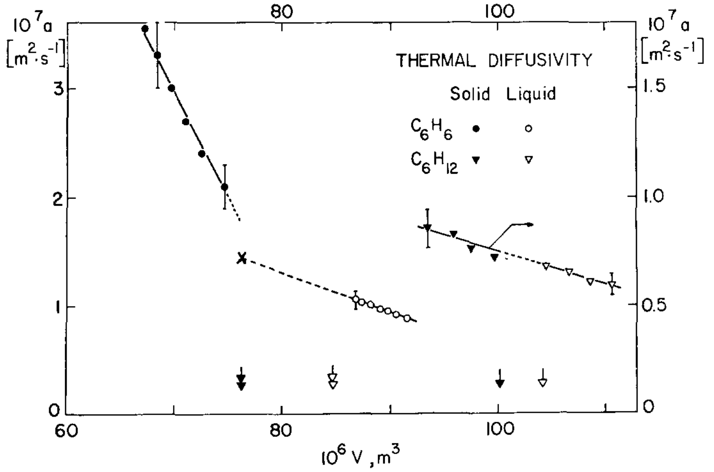

Fig. 4. Volume dependence of the thermal diffusivity in solid and liquid benzene and cyclohexane along isotherms. The thermal diffusivity is obtained from λ used in Fig. 3 and ρc_p reported by Andersson and co-workers [20, 21]. The symbols refer to the thermodynamic states as indicated in the legend to Fig. 3.

According to Takagi [9], the ultrasound velocities in liquid benzene and cyclohexane at any given pressure and temperature are different by only a few percent. Although c values for the compressed solids are not available, at atmospheric pressure and near the melting point the sound velocity in C_6H_{12}–I [10] is relatively low compared to that in C_6H_6 [8], as can be inferred from the much smaller value of c_1/c_2 in Table I.

The phonon mean free path in the plastic-crystalline solid is short and limited mainly by the disorder in molecular orientations. Such a picture is consistent with the small absolute values of λ and a of C_6H_{12}–I. According to Ross et al. [24] the presence of structural disorder is expected to provide an additional source of thermal resistivity, which in our opinion will affect the thermal diffusivity also. And it is known that the presence of lattice defects can be dominant in plastic crystals [25]. On the other hand, Mogilevskii et al. [26] found that the influence of impurities on the thermal conductivity of C_6H_{12}–I is very small. These authors and Andersson [21] also measured the λ of C_6H_{12} in the orientationally ordered low-temperature phase, C_6H_{12}–II. From their data we determined the ratios of the thermal conductivities and diffusivities, $\lambda_{\mathrm{II}}/\lambda_{\mathrm{I}} = 2.0 \pm 0.2$ and $a_{\mathrm{II}}/a_{\mathrm{I}} = 2.2 \pm 0.3$, respectively, at the II → I phase transition at 186 K, where the volume change $\Delta V/V = 8.5\%$. The discontinuity in the thermal transport properties is such that it cannot be accounted for by the change in density, similar to the results previously found for argon and benzene at melting. So the relatively small thermal transport coefficients, λ and a, and the low sound velocity of C_6H_{12}–I together with the virtual absence of a discontinuity in these quantities at the solid–liquid phase transition provide ample evidence for the liquid-like behavior of orientationally disordered cyclohexane with respect to these properties.

4. THE ISOCHORIC TEMPERATURE DEPENDENCE OF λ AND *a*

In this section, the isochoric temperature dependence of the thermal conductivity and diffusivity of solid and liquid argon, benzene, and cyclohexane are dealt with. For a normal crystal above the Debye temperature the phonon mean free path is limited by anharmonic processes and the thermal resistivity arises from a phonon interacting with other phonons, whose number density at high temperature is proportional to T. Hence $\lambda \approx T^{-1}$, as it follows from the expression for λ of a monatomic crystal under isochoric conditions [27]

$$\lambda = K m d \theta_{\mathrm{D}}^{3} / \gamma^{2} T \tag{1}$$

where K is a constant, m is the atomic mass, d is the lattice parameter, and θ_{D} is the Debye temperature. The anharmonicity is included through the

Grüneisen parameter γ. In the previous section it was shown that the thermal resistivity of solid argon, benzene, and cyclohexane along an isotherm increases linearly with the molar volume. This behavior is qualitatively consistent with the above expression for λ, since an isothermal increase in the pressure and thus in the density commonly results in an increase in θ_D and in a decrease in γ [24].

The isochoric temperature dependence of the thermal conductivity of a hot argon crystal and that of a hot complex crystal such as benzene are both found to be inversely proportional to T as shown in Table II. Evidently interactions between acoustic and optic phonons in C_6H_6 hardly affect λ as was already suggested in the previous section. The temperature dependence of λ of liquid Ar and C_6H_6 at constant densities well above ρ_c is weak and positive.

The strong increase in the thermal diffusivity of solid argon and benzene with density along an isotherm, shown in the previous section, is qualitatively consistent with the isothermal pressure dependence of λ, which if combined with that of ρ and c_p, leads to a decrease in the product ρc_p of the compressed solid. On the other hand, ρc_p of C_6H_{12}–I at 323 K tends to increase with pressure instead [20]; hence the thermal diffusivity of C_6H_{12}–I does not strongly increase with the density along an isotherm as shown in Fig. 4.

The isochoric temperature dependence of the thermal diffusivity of hot solid argon is somewhat more pronounced than that of λ; this is to be expected, as c_p at constant density decreases with lower temperatures. By analogy with the thermal conductivity, a is expressed in terms of T^n, and for solid argon this led to $n = -1.1$. The thermal diffusivity of liquid argon along an isochor is found to increase linearly with T^n [11], with an exponent $n = +0.7$ (Table II). The thermal diffusivity of solid and liquid benzene as a function of temperature is not available.

Table II. Simplified Summary of the Temperature Dependences of λ and a of Various Substances at Constant Volume in the Solid and in the Liquid State in the Neighborhood of the Phase Transition, Using Experimental Data, Reported in the Literature (see Text)[a]

	λ		a	
Substance	Solid	Liquid	Solid	Liquid
Ar	−1.0	+0.2	−1.1	+0.7
C_6H_6	−0.9	+0.3		
C_6H_{12}-I	+0.35	+0.4	−0.8	

[a] Values of the exponent n in the relations $\lambda \approx T^n$ and $a \approx T^n$, respectively, are given. The estimated uncertainty is about 10%.

In summary, the temperature dependences of a and λ in solid argon are about similar; the temperature effect on these transport coefficients of liquid argon is positive, but that of the former is much larger than that of the latter as shown in Table II. In this connection it should be noted that the thermal diffusivity strongly dominates the dynamic behavior of a simple dense liquid [28].

In the case of crystalline solids which are structurally imperfect, the thermal resistivity is determined not only by phonon interactions but also by a contribution associated with structural disorder that is added to the anharmonic term by analogy with Mathiessen's rule for electrical resistivity. The last contribution is temperature independent and relatively insensitive to pressure [24]. Such a description is consistent with the somewhat smaller isothermal volume dependence of the thermal resistivity observed in C_6H_{12}–I as compared to that in solid C_6H_6 (Fig. 3).

The temperature derivative of λ for both C_6H_{12}–I and liquid C_6H_{12} at constant density is weakly positive and of the same size. The positive temperature effect on λ of plastic-crystalline cyclohexane might be the consequence of the enhanced role which low-frequency modes play on the thermal conductivity; though being few in number, they have the larger phonon mean free path [20]. Generally, the thermal conductivity of glasses is found to increase modestly with temperature [24]. In contrast, the isochoric temperature dependence of the thermal diffusivity of C_6H_{12}–I is strongly negative, and this is due to the fact that the positive temperature effect on λ at constant density is much weaker than the decrease in heat capacity with increasing temperature [20, 24]. Data on the isochoric temperature dependence of the thermal diffusivity of liquid C_6H_{12} are not available. The exponents in the approximate relations $\lambda = T^n$ and $a = T^n$ are given in Table II.

5. CONCLUSIONS

By studying molecular dynamics on either side of the melting transition in the rare gases explored by inelastic light scattering, Fleury [30] observed no discernible difference between the liquid and the solid spectra at sufficiently high frequencies. This means that on the very short time- and length scales probed by these experiments, melting causes no discontinuous change in the substance's dynamics. The lower the frequency, the larger the discontinuity between solid and liquid behavior, and this seems to fit in with both the experimental results and Klemens' conjecture [29] of the enhanced role of the low-frequency modes in describing the lattice thermal conductivity.

On the other hand, the relatively small discontinuity in the thermal conductivity of van der Waals substances which has been observed in passing through the solid–liquid phase transition has led several authors to suggest that thermal transport in a hot solid and a liquid might be described by one and the same theory [2, 31, 32]. Such a viewpoint is neither supported by the former discussion nor consistent with the observed differences in isochoric temperature and isothermal volume dependences of the thermal conductivity and diffusivity between solid and liquid close to the phase transition, as shown in Sections 3 and 4. Moreover, the discontinuities in λ and a near the triple point cannot be accounted for by the jump in density of solid to liquid, as follows from using the thermal transport coefficients of a fictitious liquid that has the same density as the solid upon melting. In this respect, an exception should be made for the phase transition of a plastic-crystalline solid to the liquid, which does not indicate a discontinuity in the thermal transport coefficients as shown before [1]. However, it should be noted that the theory for the thermal transport properties of orientationally disordered crystals is still at an early stage of development [24].

It seems that the thermal transport coefficients of a rare gas solid and liquid may, in first approximation, be considered as representative for those properties of a normal molecular solid and liquid near the phase transition. Finally, the thermal transport coefficients in both the solid and the liquid depend much more on volume than on temperature.

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