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# Thermal Conductivity and Heat Capacity of Solid AgCl Under Pressure

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Using the transient hot-wire method, measurements were made for solid AgCl of both the thermal conductivity, $\lambda$, and the heat capacity per unit volume, $\rho c_p$, where $\rho$ is the mass density. Measurements were made in the temperature range 100 to 400 K, and at pressures up to 2 GPa. $c_p(P, T)$ could be adequately described if the acoustic modes were represented by a Debye model and the optic modes by an Einstein model. Analysis of $\lambda(T)$ showed that only the acoustic modes needed to be taken into account up to 300 K, but that the optic modes were increasingly effective in carrying heat at higher temperatures. $\lambda(P)$ was adequately described by the Lawson formula, but not by the Leibfried-Schlömann formula, to which it is formally equivalent. Agreement with experiment could be achieved by two different modifications of the Leibfried-Schlömann formula, although neither has a firm theoretical basis.

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**KEY WORDS:** heat capacity; high pressure; silver chloride (AgCl); thermal conductivity.

## 1. INTRODUCTION

AgCl in the solid state is a substance of technical importance as a pressure-transmitting medium in high pressure research. It is also a relatively simple substance, and we shall explore in some detail how well its thermal properties can be understood in terms of available theoretical models.

## 2. EXPERIMENTAL DETAILS

We used the transient hot-wire method to measure simultaneously both the thermal conductivity, $\lambda$, and the heat capacity per unit volume, $\rho c_p$,

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where $\rho$ is the mass density. Details of the method have been given elsewhere [1]. It will sometimes be convenient [2] to present and discuss results in terms of the thermal resistivity $r$ $(=1/\lambda)$.

AgCl powder was compacted under a pressure of 0.3 GPa to form polycrystalline plates, which were loaded into our high pressure cell [3]. We used samples of both 99% and 99.999% purity, but this made no difference to the results. Ordinarily, samples were handled under darkroom conditions, but, in one case, we used plates which had been deliberately darkened by exposure to light. Our purpose was to test for a radiative contribution to the total measured value of $\lambda$ for undarkened samples; this was not found since the same results were observed for all samples.

Measurements were made in the temperature ($T$) range 100 to 400 K, and at pressures ($P$) up to 2 GPa. Over these ranges, AgCl has only a single phase [4] of NaCl-type of structure. The accuracy was estimated as ±5% in $\lambda$ and ±10% in $\rho c_p$, but the precision was much higher.

## 3. RESULTS AND COMPARISON WITH PREVIOUS WORK

### 3.1 Thermal Conductivity

Our results for $\lambda(P)$ are shown in Fig. 1, and for $r(T)$ in Fig. 2. Equivalent numerical information is presented in Tables I and II. From Fig. 2, it can be

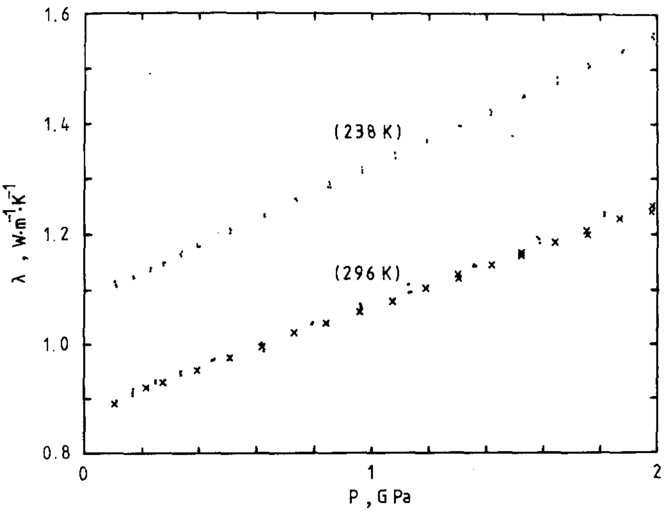

Fig. 1. Isothermal pressure dependence of thermal conductivity, $\lambda$, of AgCl. Temperature is given in parentheses. Data shown for 296 K correspond to two separate experiments.

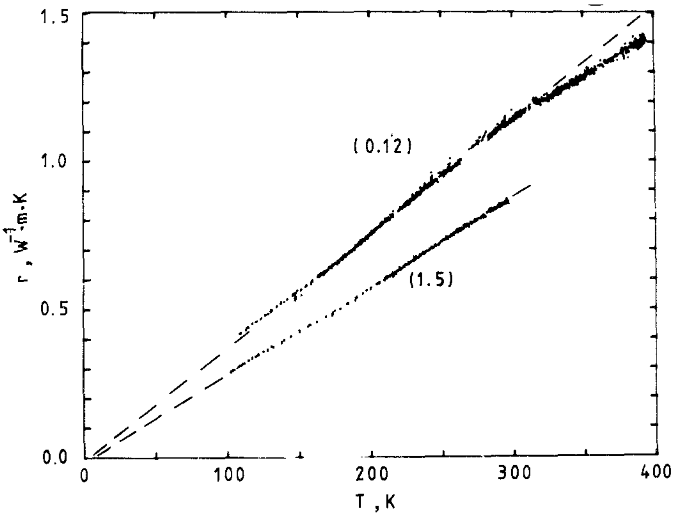

**Fig. 2.** Isobaric temperature dependence of thermal resistivity, *r*, of AgCl. Pressure in GPa is given in parentheses. Broken lines correspond to fitted equations (Table II) for $T < 300$ K.

seen that we find $r \propto T$, or equivalently $\lambda \propto T^{-1}$, at pressures of both 0.12 and 1.5 GPa, for $T \lesssim 300$ K. This temperature dependence is in agreement with theoretical prediction [5], taking only three-phonon interactions into account. Figure 2 also shows a deviation from this predicted behavior for $T > 300$ K at low pressure, which we shall argue can plausibly be attributed to a contribution to $\lambda$ from optic phonons.

Figure 3 shows comparison with previous work at zero pressure. Present work is in reasonable agreement with that of Andersson and Bäckström [6] for a polycrystalline sample using the transient hot-wire method at an early state of development, but agreement is generally poor with previous work using single-crystal samples [7–11]. For $T > 200$ K, the most likely source of the discrepancy between results for single and polycrystalline samples is radiative heat transport in the former [10]. We infer that a radiative

**Table I.** Isothermal Pressure Dependence of Thermal Conductivity $\lambda$ of AgCl Fitted to Equations of Form $\lambda = A + BP$, Where $\lambda$ is in $W \cdot m^{-1} \cdot K^{-1}$, and $P$ is in GPa

| *A* | *B* | *T* (K) | *P* (GPa) |
|---|---|---|---|
| 0.878 | 0.192 | 296 | 0–2.0 |
| 1.083 | 0.239 | 238 | 0–2.0 |

**Table II.** Isobaric Temperature Dependence of Thermal Resistivity $r$ of AgCl Fitted to Equations of Form $r = D + ET$, Where $r$ is in $W^{-1} \cdot m \cdot K$ and $T$ is in K

| $D$ | $E$ | $P$ (GPa) | $T$ (K) |
|---|---|---|---|
| −0.020 | $3.86 \times 10^{-3}$ | 0.12 | 108–293 |
| 0.284 | $2.85 \times 10^{-3}$ | 0.12 | 293–394 |
| −0.019 | $2.97 \times 10^{-3}$ | 1.5 | 102–294 |

component was absent for our samples, since darkening by exposure to light made no difference to the results.

For the pressure dependence of $\lambda$, the present work yields a value for the ratio $\lambda(2 \text{ GPa})/\lambda(0) = 1.44$ at 296 K, as may be deduced from Fig. 1 or Table I. Vereshchagin et al. [12] measured the pressure dependence of $\lambda$ for AgCl using a steady-state method at about 300 K, and we deduce from their measurements a value for this ratio of 1.4. Andersson and Bäckström [6] made measurements using the transient hot-wire method up to 1 GPa, from which a value $\lambda(1 \text{ GPa})/\lambda(0) = 1.25$ may be deduced. The corresponding value from the present work is 1.22. Results from the present work for the pressure dependence of $\lambda$ are thus in satisfactory agreement with both of these previous measurements. Poor agreement is found, however, with previous

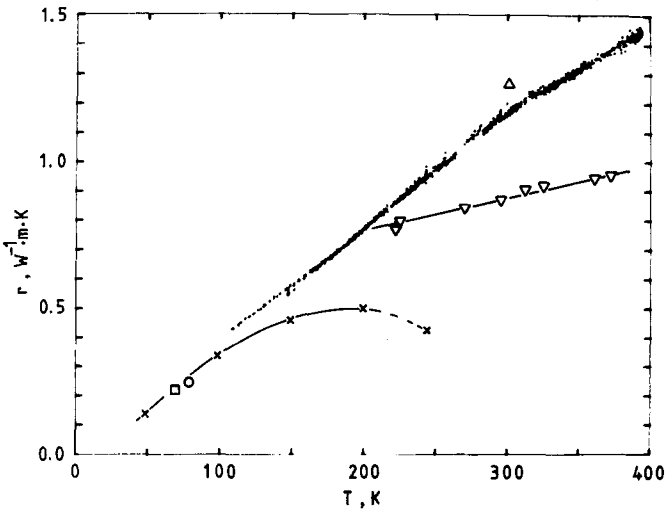

**Fig. 3.** Isobaric temperature dependence of thermal resistivity, $r$, of AgCl at atmospheric pressure showing comparison with previous work. Polycrystalline samples: present work (extrapolated) (●), [6] (△). Single-crystal samples: [7, 8] (▽), [9] (○), [10] (×), [11] (□).

results due to Rosander and Bäckström [13], obtained using a steady-state method, which yielded $\lambda(2\ \mathrm{GPa})/\lambda(0) = 1.21$. It seems likely [12] that this relatively low value was due to systematic error.

### 3.2 Heat Capacity

Our results for $\rho c_p$ are shown in Figs. 4 and 5. Figure 4 shows a comparison with previous work at zero pressure, using measurements of $c_p$ due to Eastman and Milner [14], and taking a constant value for $\rho = 5.57\ \mathrm{g \cdot cm^{-3}}$. Agreement is within experimental error. The relatively rapid variation of $\rho c_p$ with $T$ at the lowest temperatures at 1.5 GPa (Fig. 5) may be due in part to systematic error.

## 4. DISCUSSION

The dispersion relation is basic to any discussion of thermal properties. Since AgCl has a diatomic basis, there are three acoustic and three optic modes, the transverse modes being degenerate by symmetry in the [100] and [111] directions. Vijayaraghavan et al. [15] used neutron inelastic scattering to measure the dispersion relation for AgCl. They measured only one of the two nondegenerate transverse modes in the [110] direction. The slopes of all acoustic modes in the long-wavelength limit around the [100]-[110]-[111]

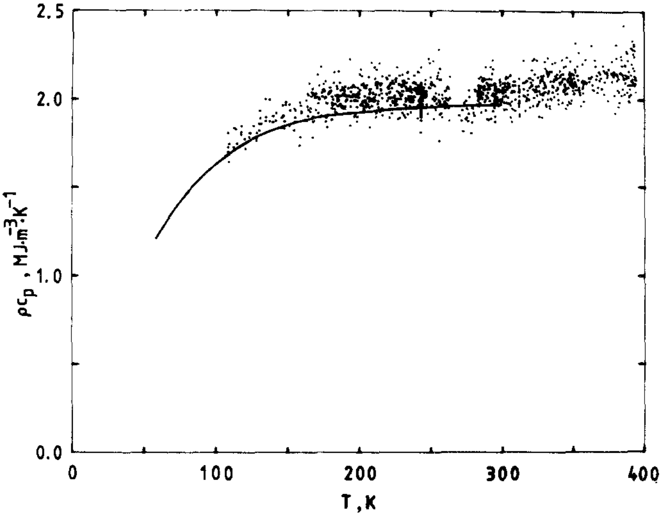

Fig. 4. Isobaric temperature dependence of heat capacity per unit volume, $\rho c_p$, of AgCl at 0.12 GPa. Previous work at atmospheric pressure [14] shown by solid line, taking $\rho = 5.57\ \mathrm{g \cdot cm^{-3}}$.

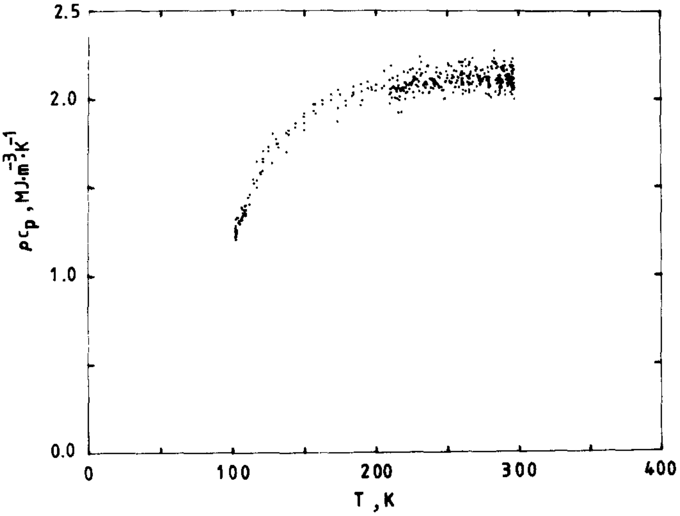

**Fig. 5.** Isobaric temperature dependence of heat capacity per unit volume, $\rho c_p$, of AgCl at 1.5 GPa.

triangle are available from the single-crystal sound velocity measurements of Loje and Schuele [16]. In principle, all acoustic and optic modes need to be taken into account for both $\lambda$ and $c_p$.

Experimental data for $c_p$ can be fitted with adequate accuracy to a very simple model. This reflects the well-known fact that the calculation of the heat capacity is rather insensitive to the detailed features of the dispersion relation, or the corresponding density of modes. This is exemplified for AgCl by the calculation made by Donecker [17]. He assumed that the acoustic modes could be represented by a Debye model, and the optic modes by an Einstein model, with characteristic temperatures $\theta_D$ and $\theta_E$, respectively. The total heat capacity was then the sum of these two contributions. Donecker's additional assumption of an Einstein oscillator having a characteristic temperature of 50 K makes no significant difference to the results for $T > 100$ K, so we ignore it. Donecker showed that the choice of parameters $\theta_D = 120$ K and $\theta_E = 200$ K gave good agreement with the experimental results of Eastman and Milner [14] and hence with the present results at low pressure (Fig. 4).

In order to use Donecker's model to predict the pressure dependence of $c_p$, we need estimates for the pressure dependences of $\theta_D$ and $\theta_E$. Voronov and Grigor'ev [18] measured longitudinal and transverse sound velocities under pressure for polycrystalline specimens of AgCl. Using a formula for an isotropic elastic continuum, they calculated $\theta_D(P)$ and found it to be almost independent of pressure up to 2 GPa. The implication that such a result has for $\lambda$ will be discussed below. From the infrared transmission measurements of Lowndes and Rastogi [19], we estimate that $\theta_E$ increases by about 15% on pressurization to 1.5 GPa. At the latter pressure, we therefore take $\theta_D = 120$ K and $\theta_E = 230$ K. The result, compared with zero pressure, is a shift of the calculated curve for $c_p(T)$ by about 15 K to higher temperatures. As can be seen by comparing Figs. 4 and 5, this is about the shift we observed in the measured values of $\rho c_p$.

Analysis of the thermal conductivity is more difficult. It is usual to start with the Debye formula

$$\lambda = \frac{1}{3}\sum_i c_i v_i l_i \tag{1}$$

where, for the $i$th mode, $c_i$ is the heat capacity per unit volume, $v_i$ the phonon velocity, $l_i$ the phonon mean free path, and $\Sigma_i$ indicates summation over the modes. The determination of $\lambda$ using a reasonably realistic dispersion relation involves complex calculations and has been attempted in only a few cases [20, 21], but a simple conclusion that can be drawn immediately from Eq. (1) is that phonons having a small velocity will be ineffective in carrying heat. Such phonons are those having a flat dispersion curve, as is often the case for the optic modes of a crystal of greater than monatomic basis. However, if we assume that the thermal resistivity arises entirely from phonon-phonon umklapp processes, then in principle, low velocity phonons cannot be ignored, since they may scatter other phonons. For the case where the low velocity phonons belong to an optic mode of a crystal of diatomic basis, Slack [22] has derived a formula to take into account the resultant optic-acoustic phonon interaction.

In general, optic phonon velocities are not necessarily small. For AgCl, the results of Vijayaraghavan et al. [15] show that the transverse optic and transverse acoustic velocities are similar in parts of the Brillouin zone. At temperatures sufficiently high that significant numbers of optic phonons are excited, our previous discussion implies that these can be expected to affect $\lambda$ in two competing ways. The optic phonons will tend to increase $\lambda$ by carrying heat, and to decrease $\lambda$ through the optic-acoustic interaction. For crystals of diatomic basis, it was argued by Devyatkova and Smirnov [23] that the competition between these two effects would be regulated primarily by the mass ratio $\sigma$ and displayed through the temperature dependence $r(T)$. Their arguments will now be restated briefly. Analysis of a linear diatomic model with nearest neighbor interactions shows that as $\sigma \rightarrow 1$, optic phonon velocities increase, and the separation in energy of the optic and acoustic modes decreases. The former would be expected to lead to greater heat transport by optic phonons, while the latter should enhance the optic-acoustic interaction. In order to decide which of these two influences on the thermal conductivity was the more important for a given material, Devyatkova and Smirnov proceeded empirically. Experimental data for a number of crystals of diatomic basis shows that $r(T)$ generally exhibits three distinct linear regions. For $T \lesssim \theta_D$, $r = bT$, where $b$ is a constant, the general form of which will be discussed below. This is the temperature dependence which is expected when the thermal resistivity arises from three-phonon umklapp processes in a solid of monatomic basis [5], strictly at constant volume. At such low temperatures, optic modes are considered to have negligible influence on the thermal conduction process. At the highest temperatures, $r = b'T$, where $b' \neq b$. Optic modes may be considered to be fully effective in relation to thermal conduction. In this connection, it seems reasonable that at sufficiently high temperatures, energy gaps in the dispersion relation should be relatively unimportant on the scale of thermal energies. There is thus a range of temperature intermediate between the regions characterized by differing values of the constant of proportionality ($b$ or $b'$), and this intermediate region may be taken to correspond to the change from negligible to full influence of the optic modes on thermal conduction. When $r$ increased less rapidly than $bT$ in the intermediate region, Devyatkova and Smirnov inferred that the effect of increasing influence of optic phonons was exhibited predominantly through the heat which they transported rather than through optic-acoustic interactions, and conversely. The former behavior was correlated empirically with $\sigma > 2$.

Our results for AgCl at 0.12 GPa (Fig. 2) are reasonably consistent with the analysis of Devyatkova and Smirnov [23]. The boundary between the low and intermediate temperature regions described above may be taken to occur at about 300 K. This is substantially above the Debye temperature, which is different than for the substances considered by Devyatkova and Smirnov. We infer that for $T \gtrsim 300$ K, the optic modes exert an increasing influence on thermal conduction in such a way that heat transport by optic phonons is more important than optic-acoustic interactions. This inference is consistent with the analysis of Devyatkova and Smirnov since $\sigma = 3.1$ for AgCl. The highest temperature region described above ($r = b'T$) was not observed in the temperature range of our experiments.

According to this analysis, and from Fig. 2, we infer that optic phonons have an insignificant influence, or at least an insignificant net influence, on thermal conduction at 300 K. By contrast, the total heat capacity at this temperature is made up of approximately equal contributions from the acoustic and optic modes. This fact may be demonstrated either by detailed inspection of Donecker's [17] model, or more directly by simply noting that $c_p$ at 300 K is nearly $6R$ per mole, where $R$ is the gas constant. Such a difference in the influence of the optic modes on the heat capacity on the one hand, and on thermal conduction on the other, must presumably be connected, probably in a complex way, with the velocities and mean free paths of the optic phonons.

We now consider the pressure dependence of $\lambda$ for $T \cong 300$ K. For reasons just described, we assume that only the acoustic modes need to be taken into account, as is done (strictly at constant volume) by the Leibfried-Schlömann (LS) formula [5], which is

$$\lambda = B\frac{Ma\theta_D^3}{\gamma^2 T} \tag{2}$$

In Eq. (2), $M$ is the atomic mass, $a$ is the lattice parameter, $\theta_D$ is the Debye temperature, $\gamma$ is the Grüneisen parameter, and $B$ is a constant. Although the LS formula was derived for a crystal of monatomic basis, it seems reasonable to suppose that it can be applied to the acoustic modes of a crystal of diatomic basis if $M$ is taken to be some reduced or averaged mass.

Suppose we use Eq. (2) to consider $\lambda(P)$, and employ the data of Voronov and Grigor'ev [18]. As described above, these workers deduced from their measurements of sound velocity on polycrystalline samples that $\theta_D$ was almost independent of pressure up to 2 GPa. Equation (2) then implies that the pressure dependence of $\lambda$ is due almost entirely to the pressure dependence of $\gamma$. In the range 0–2 GPa, $\lambda$ increases by about 40%, so $\gamma$ would need to decrease by about 20% to satisfy Eq. (2). Such a large pressure dependence of $\gamma$ is improbable [12, 24], but there are no independent data.

In a previous discussion of the pressure dependence of $\lambda$ for AgCl, Vereshchagin et al. [12] used the Lawson formula, which is

$$\lambda \sim (\gamma^2\kappa^{3/2}\rho^{5/6}T)^{-1} \tag{3}$$

where $\kappa$ is the compressibility. With the same data of Voronov and Grigor'ev [18] as was used in the preceding paragraph, only now for $\kappa$ rather than for $\theta_D$, Vereshchagin et al. were able to achieve good agreement with experiment for the pressure dependence of $\lambda$, with the assumption that $\gamma$ was independent of pressure. Such agreement may be fortuitous, but we can at least conclude that the LS and Lawson formulas are inconsistent in relating acoustic and thermal data for AgCl, as was noted previously by Rosander and Bäckström [13].

However, working at the same level of approximation as was used in the original derivation of the Lawson formula [25], we can also derive it from the LS formula. We write $\theta_D \sim va^{-1} \sim \kappa^{-1/2}\rho^{-1/2}a^{-1}$, where $v$ is some average sound velocity, and note that $a \sim \rho^{-1/3}$. Substitution in the LS formula then yields the Lawson formula. This formal derivation is not, in fact, valid for

AgCl, since the implied relation $\theta_D \sim \kappa^{-1/2}\rho^{-1/6}$ is not satisfied, according to data of Voronov and Grigor'ev [18].

This leads us to examine more closely the quantity $\theta_D$. As described above, Voronov and Grigor'ev [18] used an isotropic elastic continuum approximation, with the result that $\theta_D$ was nearly independent of pressure up to 2 GPa. To show that this result is not simply a consequence of the continuum approximation, we consider the data of Loje and Schuele [16], obtained using single-crystal specimens. These workers made measurements up to 0.1 GPa, and deduced initial derivatives $d\ln C/dP$, where $C$ represents an elastic constant. If we assume that these initial logarithmic derivatives are independent of pressure up to 1 GPa, we can calculate the elastic constants at the latter pressure. Using standard formulas relating elastic constants to $\theta_D$, as described by Alers [26], we can calculate this quantity at 0 and 1 GPa. The result is that we find almost no change of $\theta_D$ over this range of pressure, just as for the continuum approximation, compared with a corresponding change of $\lambda$ of about 20%. Use of a continuum approximation for $\theta_D$ is therefore not the reason why the LS formula fails to adequately describe the pressure dependence of $\lambda$.

Such a failure of the LS formula has been previously noted [24,27] in connection with the alkali halides. Although a modest pressure dependence of $\gamma$ was allowed for, agreement between the LS formula and experiment could only be achieved for the alkali halides [24] through an additional factor involving elastic constants, $(c_{11}(P)/c_{44}(P))^{0.6}$[28]. This factor had been deduced empirically, based on semiquantitative arguments concerning elastic anisotropy. For AgCl, this factor can be evaluated at 0 and 1 GPa, using the data of Loje and Schuele [16] as described above. The ratio for the two pressures then yields an estimate for $\lambda(1 \text{ GPa})/\lambda(0)$, taking the unmodified LS formula to give no change of $\lambda$ with pressure. The result is the value 1.17, compared with an experimental value of 1.22. This empirical modification of the LS formula therefore yields reasonable agreement with experiment for AgCl.

An alternative modification of the LS formula may also be devised. We note that it is of the essence of the Debye model, and of the LS formula for $\lambda$, that the vibrational spectrum of the acoustic modes is characterized by means of a single parameter $\theta_D$. Similarly, in the LS formula, the anharmonicity is characterized by means of a single parameter $\gamma$. The consequent averaging may obscure significant features of the pressure dependence, especially in relation to $\lambda$. We shall attempt to reduce this inherent averaging by assuming without proof that an expression of the form of the LS formula may be applied to each polarization branch of the acoustic spectrum, considered separately, and the result summed. This implies that for the $i$th mode, with velocity $v_i$, we define a corresponding characteristic temperature $\theta_i \sim v_i\rho^{1/3}$, and also make use of the mode Grüneisen parameter $\gamma_i$. Also implied is an expression of the form

$$\lambda \sim \rho^{2/3} \sum_i \frac{v_i^3}{\gamma_i^2} \tag{4}$$

where $\Sigma_i$ indicates summation over the modes. The elastic constants at 0 and 1 GPa, calculated from the data of Loje and Schuele [16] as described above, yield the corresponding mode sound velocities at these pressures in the [100], [110], and [111] directions. The corresponding mode Grüneisen parameters have been tabulated by Loje and Schuele, and we assume these to be independent of pressure. We can then calculate the ratio $\lambda(1\ \mathrm{GPa})/\lambda(0)$ using Eq. (4), and the results are the values 1.20, 1.18, and 1.18 in the [100], [110] and [111] directions, respectively. The corresponding experimental result is 1.22. Although such good agreement may be to some extent fortuitous, it seems reasonable to conclude that the LS formula needs significant modification before it can be used with confidence to predict the pressure dependence of $\lambda$. The modification we have proposed, Eq. (4), leads to a successful prediction of $\lambda(P)$ from elastic constants for AgCl but requires both theoretical and further experimental justification before it can be considered to be of more general validity.

## 5. CONCLUSIONS

Both acoustic and optic modes need to be taken into account in discussing the thermal properties of AgCl. The heat capacity can be adequately described if the acoustic modes are represented by a Debye model and the optic modes by an Einstein model. Analysis of the thermal conductivity is more complex, and less certain. From the temperature dependence of the thermal resistivity, it appears that only the acoustic modes are effective in thermal conduction up to about 300 K. At higher temperatures, the optic modes appear to show their effect predominantly in the transport of heat, rather than in optic-acoustics scattering, Using available sound velocity data, the pressure dependence of the thermal conductivity at 300 K is adequately described by the Lawson formula, with the assumption that the Grüneisen parameter is independent of pressure. On the same basis, the Leibfried-Schlömann formula fails to agree with experiment, unless either of two possible modifications is made. One modification is to take into account elastic anisotropy through an empirical function of the elastic constants, and the other is to assume that a formula of Leibfried-Schlömann type may be applied to each acoustic mode separately. Both modifications yield reasonable agreement with experiment, but neither has a firm theoretical basis.

## ACKNOWLEDGMENT

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