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ANALYSIS OF LATTICE THERMAL CONDUCTIVITIES OF RARE-EARTH SULPHIDES AT HIGH TEMPERATURES

APPLICATION TO GdS AND LaS

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The lattice thermal conductivities of rare-earth sulphides have been analyzed at high temperatures in the frame of the two-mode conduction of phonons for the first time by studying the total lattice thermal conductivities of GdS and LaS in the entire temperature range 100–1000 K. The temperature exponents for the three-phonon scattering relaxation rates are reported for the transverse as well as for the longitudinal phonons. The separate percentage contributions due to the transverse and longitudinal phonons towards the total lattice thermal conductivities of the above samples have similarly been studied. The role of the four-phonon processes too has been included in the present investigation.

The lattice thermal conductivities of numerous samples have been studied by a number of workers [1–10], experimentally as well as theoretically, at high and at low temperature, and it is now well established that high-temperature lattice thermal conductivity data can not be explained by one conductivity integral as proposed by Callaway [11]. It was Holland [12] who first introduced the two-mode conduction of phonons to explain the lattice thermal conductivities of Ge and Si at high temperatures. Later, following Guthrie [13, 14], the author and his co-workers [15–18] proposed a modification to the Holland model, which is known as the Sharma–Dubey–Verma (SDV) model [15–18]. In the SDV model [15–18], the phonon-phonon scattering events have been classified into two classes: class I events in which a carrier phonon is annihilated by combination, and class II events in which annihilation takes place by splitting. From their studies, it is clear that the SDV model gives a very good response in explaining experimental lattice thermal conductivity data at high temperatures. At the same time, the temperature-dependence of the phonon-phonon scattering relaxation rate used in the SDV model is also free from the Guthrie comments [13, 14].

Khusnutdinova *et al.* [19] tried to explain the high-temperature lattice thermal conductivity data on GdS and LaS by using an analytical expression obtained in the frame of the Callaway [11] expression based on the high-temperature approximations. They used an expression $\tau_{3ph}^{-1}\alpha_w^2 T$ for the three-phonon scattering relaxation rate, which is valid for longitudinal phonons only. At the same time, all of their calculations are based on the Callaway expression, which gives a good response to the experimental lattice thermal conductivity data at low temperatures only. Thus, it is interesting to analyse the lattice thermal conductivities of the above samples in the frame of the two-mode conduction of phonons.

The aim of the present work is to analyse the lattice thermal conductivities of rare-earth sulphides in the frame of the two-mode conduction of phonons at high temperatures by studying the total lattice thermal conductivities of GdS and LaS by estimating the separate contributions due to the transverse and longitudinal phonons for the first time. The separate percentage contributions due to the transverse and longitudinal phonons towards the total lattice thermal conductivities of the above samples have likewise been studied. The temperature exponents for transverse as well as for longitudinal phonons are also reported for both of the samples for the first time. The entire study has been performed in the frame of the SDV model of lattice thermal conductivity in the entire temperature range 100 – 1000 K. The role of the four-phonon scattering relaxation rate too has been included in the present analysis.

Phonon conductivity integral and temperature exponent $m(T)$

While commenting on the nature of the dependence of the lattice thermal conductivity on the frequency ω and temperature T, Guthrie [13] gave an expression for the three-phonon scattering relaxation rate $\tau_{3\,\mathrm{ph}}^{-1}$ in the form

$$\tau_{3\,\mathrm{ph}}^{-1} \propto g(\omega) f(T) \tag{1}$$

where $f(T) = T^{\mathrm{m}}$ and $m = m(T)$, $g(\omega) = \omega$ for transverse phonons and $g(\omega) = \omega^2$ for longitudinal phonons, similar to the findings of Herring [20]. Incorporating the Guthrie ideas and following Klemens [21], the author and his co-workers [15 – 18] gave an expression for $\tau_{3\,\mathrm{ph}}^{-1}$ as

$$\tau_{3\,\mathrm{ph}}^{-1} \propto g(\omega) T^{\mathrm{m(T)}} e^{-\Theta/\alpha \mathrm{T}} \tag{2}$$

where Θ is the Debye temperature and α is a constant which has the same meaning as given by Klemens [21]. According to the SDV model, the three-phonon scattering relaxation rate $\tau_{3\,\mathrm{ph,T}}^{-1}$ for transverse phonons is given by

$$\tau_{3\,\mathrm{ph}}^{-1} = B_{\mathrm{T,I}} \omega T^{\mathrm{m_{T,I}(T)}} \mathrm{e}^{-\Theta/\alpha \mathrm{T}} \tag{3}$$

since only class I events are possible for transverse phonons. For longitudinal phonons, one obtains

$$\tau_{3\,\mathrm{ph}}^{-1} = [B_{\mathrm{L,I}} T^{\mathrm{m_{L,I}(T)}} + B_{\mathrm{L,II}} T^{\mathrm{m_{L,II}(T)}}] \omega^2 \mathrm{e}^{-\Theta/\alpha \mathrm{T}}. \tag{4}$$

The value of the temperature exponent $m(T)$ is given by

$$m_{\mathrm{I}}(T) = X_{\mathrm{max}} (\mathrm{e}^{\mathrm{X_{max}}} - 1)^{-1} + 0.5 X_{\mathrm{max}} + \frac{\ln (1 + \Theta/\alpha T)}{\ln T} \tag{5}$$

$$m_{\mathrm{II}}(T) = 0.5 X_{\mathrm{max}} \mathrm{e}^{0.5\,\mathrm{X_{max}}} (\mathrm{e}^{\mathrm{X_{max}}} - 1)^{-1} + 0.5 + \frac{\ln (1 + \Theta/\alpha T)}{\ln T} \tag{6}$$

where $X_{max} = \dfrac{\hbar\omega_{max}}{K_B T}$, ω_{max} is the maximum frequency and the other term will be explained later.

In the above expressions suffixes T and L represent transverse and longitudinal phonons, respectively, and suffixes I and II refer to class I and class II events, respectively. The B's are the scattering strengths of the respective scattering processes.

At high temperatures, $e^{-\Theta/\alpha T}$ reduces to unity and the value of $m(T)$ tends to unity for both polarisation branches, with the result that $\tau_{3\,ph,T}^{-1} \propto \omega T$ for transverse phonons and $\tau_{3\,ph,T}^{-1} \propto \omega^2 T$ for longitudinal phonons, as found by Klemens [21]. At low temperatures, the exponent $m_{T,I}(T)$ tends to 4 and $m_{L,I}(T)$ tends to 3, which leads to a T^4-dependence for the transverse phonons and a T^3-dependence for the longitudinal phonons, similar to the earlier findings of Herring [20] for low-frequency phonons.

Using the above expression for the three-phonon scattering relaxation rate $\tau_{3\,ph}^{-1}$ applying the dispersion correction [4] and following the SDV model, the lattice thermal conductivity of a sample can be expressed as

$$K = K_T + K_L \tag{7}$$

where K_T and K_L are the lattice thermal conductivities due to transverse and longitudinal phonons, respectively, and can be expressed as

$$\begin{aligned} K_T = (C/V_{T1}) \int_0^{\Theta_1/T} \tau_{c,T}(1 + R_1x^2T^2)^2(1 + 3R_1x^2T^2)^{-1}F(x)dx + \\ + (C/V_{T2}) \int_{\Theta_1/T}^{\Theta_2/T} \tau_{c,T}(1 + R_2x^2T^2)^2(1 + 3R_2x^2T^2)^{-1}F(x)dx \end{aligned} \tag{8}$$

$$\begin{aligned} K_L = (C/2V_{L1}) \int_0^{\Theta_3/T} \tau_{c,L}(1 + R_3x^2T^2)^2(1 + 3R_3x^2T^2)^{-1}F(x)dx + \\ + (C/2V_{L2}) \int_{\Theta_3/T}^{\Theta_4/T} \tau_{c,L}(1 + R_4x^2T^2)^2(1 + 3R_4x^2T^2)^{-1}F(x)dx \end{aligned} \tag{9}$$

where $C = (K_B/3\pi^2)(K_BT/\hbar)^3$, $F(x) = x^4e^x(e^x - 1)^{-2}$, $x = \hbar\omega/K_BT$, $R_i = r_i(K_B/\hbar)^2$, $i = 1, 2, 3$ and 4, $\Theta_i = \dfrac{\hbar\omega_i}{K_B}$, $i = 1, 2, 3$ and 4, $\hbar$ is the Planck constant divided by 2π, K_B is the Boltzmann constant, the Θ's are the dispersion constants, the V's are the characteristic temperatures corresponding to the Brillouin zone boundary, the V's are the velocities (for details see refs. [4, 15–18]), $\tau_{c,T}$ and $\tau_{c,L}$ are the combined scattering relaxation times due to transverse phonons and longitudinal phonons, respectively, and can be expressed as

$$\tau_{c,T}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + \tau_{3\,ph,T}^{-1} + \tau_{4\,ph}^{-1} \tag{10}$$

$$\tau_{c,L}^{-1} = \tau_B^{-1} + \tau_{pt}^{-1} + \tau_{3\,ph,L}^{-1} + \tau_{4\,ph}^{-1} \tag{11}$$

where τ_B^{-1} and τ_{pt}^{-1} are the scattering relaxation rates due to boundary [22] and point-defect [21] scattering, respectively, and τ_{4ph}^{-1} is the four-phonon scattering [23, 24] relaxation rate.

It should be noted that the correction term [11] due to the three-phonon normal processes has been ignored in Eqs (8) and (9), due to its very small contributions [25–29]. The boundary scattering relaxation rate τ_B^{-1} has also been ignored in the actual calculation, due to its very small contribution at high temperatures.

Analytical expression

The conductivity integrals in Eqs (8) and (9) are very complicated and one has to perform the numerical integration of these integrals at each temperature, which is not an easy task. To have an approximate value of the lattice thermal conductivity at high temperatures, an analytical expression can be obtained by evaluating these conductivity integrals under the high-temperature approximations. At the same time, these analytical expressions can also be used in approximating the values of the scattering strengths.

At high temperatures, $x \ll 1$ and $x^2e^x(e^x - 1)^{-2}$ can be approximated to unity. At the same time, the exponential factor $e^{-\Theta/\alpha T}$ can also be approximated to unity, due to the large value of T. Neglecting τ_B^{-1} due to its negligibly small contribution at such temperatures, following the earlier work of the author [30, 31], and using the above approximations, the contribution K_Γ as given in Eq. (8) can be approximated as

$$K_T = (C/B_1V_{T1}) \int_0^{\Theta_1/T} x[1 - (H_1/B_1)x - (D/B_1)x^3]dx + \\ + (C/B_1V_{T2}) \int_{\Theta_1/T}^{\Theta_2/T} x[1 - (H_1/B_1)x - (D/B_1)x^3]dx \quad (12)$$

where

$$B_{T1}\omega T^{m_{T,1}(T)} = B_1x = b_1T^{m_1+1}x, \; m_{T,1}(T) = m_1$$

$$B_{HT}\omega^2T^2 = H_1x^2 = h_1T^4x^2$$

$$A\omega^4 = Dx^4 = dT^4x^4.$$

To avoid complications, the dispersion correction has not been included and the constants r_1 and r_2 are assumed to be zero. Evaluating the integrals in Eq. (12), one has

$$K_T = [C(1 - a)/(2b_1V_{T1})]T^{-m_1}\Theta_1^2[1 - (2/3)(h_1/b_1)T^{2-m_1}\Theta_1 - (2/5)(d/b_1)T^{-m_1}\Theta_1^3] + \\ + (C/2b_1V_{T2})T^{-m_1}\Theta_2^2[1 - (2/3)(h_1/b_1)T^{2-m_1}\Theta_2 - (2/5)(d/b_1)T^{-m_1}\Theta_2^3[\quad (13)$$

where $a = V_{T2}/V_{T1}$.

Similarly, the contribution K_L in Eq. (9) can be approximated as

$$K_L = [C(1 - a')/(2b_3V_{L1})]T^{-m_3}\Theta_3[1 - (h_2/b_3)T^{2-m_3} - (b_2/b_3)T^{m_2-m_3} - (d/3b_3)T^{-m_3}\Theta_3^3] + (C/2b_3V_{L2})T^{-m_3}\Theta_4[1 - (h_2/b_3)T^{2-m_3} - (b_2/b_3)T^{m_2-m_3} - (d/3b_3)T^{-m_3}\Theta_4^3] \quad (14)$$

where

$$B_{L,I}\omega^2T^{m_{L,I}(T)} = b_2T^{m_2+2}x^2;\ m_{L,I}(T) = m_2,$$

$$B_{L,II}\omega^2T^{m_{L,II}(T)} = b_3T^{m_3+2}x^2;\ m_{L,II}(T) = m_3,$$

$$a' = V_{L2}/V_{L1}.$$

For simplicity, the dispersion constants r_3 and r_4 are assumed to be zero, similar as for K_T.

Phonon conductivity of GdS

Khusnutdinova et al. [19] measured the thermal conductivity of GdS in the temperature range 80–720 K, where they also determined the Lorenz number and the electrical conductivity of the same sample of GdS, which can be used to estimate the electronic part of the thermal conductivity with the help of the Wiedman–Franz law. Following the earlier work of the author [8], the lattice thermal conductivity of GdS has been separated by subtracting the electronic part of the thermal conductivity from the total thermal conductivity measured by Khusnutdinova et al. [19].

Following the earlier work of the author [8, 9] and using the analytical expressions reported in the earlier section, the three-phonon scattering strengths $B_{T,I}$, $B_{L,I}$, and $B_{L,II}$ have been determined at 500 K, while the four-phonon scattering strengths $B_{H\Gamma}$ and B_{HL} are estimated at 700 K. The point-defect scattering strength A has been taken from the earlier report of Edani and Dubey [32]. The constants and parameters used in the present analysis of the lattice thermal conductivity of GdS are listed in Table 1. Using the constants reported in this Table, the temperature exponents $m(T)$ have been calculated for both transverse and longitudinal phonons in the entire temperature range 100–1000 K with the help of Eqs (5) and (6), and the results obtained are reported in Table 2.

Using the constants listed in Table 1 and the values of the temperature exponents $m(T)$ reported in Table 2, the total lattice thermal conductivity of GdS has been calculated in the entire temperature range 100–1000 K by estimating the separate contributions K_T and K_L due to transverse and longitudinal phonons, respectively, and the results obtained are illustrated in Fig. 1. To study the relative contributions due to transverse and longitudinal phonons, the separate percentage contributions due to each mode of the phonons are also calculated in the entire temperature range of study and the results obtained are reported in Table 3.

Table 1

The constants used in the calculations of the lattice thermal conductivity of GdS and LaS in the temperature range 100—1000 K

Constants	GdS	LaS
V_{T1}, m/sec	2.68×10^{3}	2.59×10^{3}
V_{T2}, m/sec	8.4×10^{2}	8.10×10^{2}
V_{L1}, m/sec	4.42×10^{3}	4.27×10^{3}
V_{L2}, m/sec	4.42×10^{3}	4.27×10^{3}
θ_1, K	47	44
θ_2, K	62	58
θ_3, K	150	142
θ_4, K	256	241
θ_4, K	266	251
	2	2
R_1, sec^2	4.5×10^{-27}	4.1×10^{-27}
R_2, sec^2	1.08×10^{-26}	9.98×10^{-27}
R_3, sec^2	0	0
R_4, sec^2	1.13×10^{-27}	1.03×10^{-27}
A, sec^3	2.0×10^{-42}	2.7×10^{-42}
$B_{T,I}$, deg^{-n}	4.25×10^{-6}	4.45×10^{-6}
$B_{L,I}$, sec deg^{-m}	1.0×10^{-20}	1.0×10^{-20}
$B_{L,II}$, sec deg^{-m}	1.0×10^{-17}	8.0×10^{-18}
H_T, sec deg^{-2}	3.5×10^{-24}	1.5×10^{-24}
H_L, sec deg^{-2}	3.5×10^{-24}	1.5×10^{-24}

Table 2

The value of the temperature exponent $m(T)$ obtained using Eqs (5) and (6) used to calculate the lattice thermal conductivity of GdS. $m_{T,I}(T)$ is the temperature exponent of the three phonon scattering relaxation rate due to transverse phonons for class I event, whereas $m_{L,I}(T)$ and $m_{L,II}(T)$ are the same due to longitudinal phonons for class I and class II event respectively.

T, K	$m_{T,I}(T)$	$m_{L,I}(T)$	$m_{L,II}(T)$
100	1.1366	1.7702	1.0
200	1.0590	1.2885	1.0
300	1.0365	1.1681	1.0
400	1.0261	1.1165	1.0
500	1.0201	1.0883	1.0
600	1.0163	1.0707	1.0
700	1.01361	1.0587	1.0
800	1.0117	1.0501	1.0
900	1.0102	1.0435	1.0
1000	1.0090	1.0385	1.0

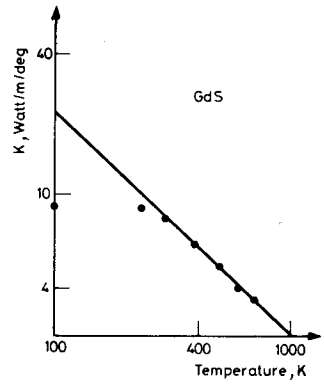

Fig. 1. The lattice thermal conductivity of GdS in the temperature range 100—1000 K. Solid line is the calculated value and circles are the experimental points

Phonon conductivity of LaS

The thermal conductivity of LaS was measured by Golubkov *et al.* [33] and its lattice thermal conductivity has been subtracted similarly as in the earlier section for GdS. The scattering strengths of the respective processes are estimated similarly as for GdS in the earlier section and the values of the constants and parameters used in the analysis of the lattice thermal conductivity of LaS are listed in Table 1.

Table 3

The relative contributions due to transverse and longitudinal phonons for GdS in the entire temperature range 100—1000 K. $\%\ K_T$ is the percentage contribution due to transverse phonons and $\%\ K_L$ is the same due to longitudinal phonons

T, K	$\%\ K_T$	$\%\ K_L$
100	84.77	15.23
200	89.85	10.15
300	91.20	8.80
400	91.79	8.21
500	92.12	7.88
600	92.32	7.68
700	92.46	7.54
800	92.56	7.44
900	92.63	7.37
1000	92.69	7.31

Table 4

The values of the temperature exponent $m(T)$ obtained using Eqs (5) and (6) used to calculate the lattice thermal conductivity of LaS. $m_{T,I}(T)$ is the temperature exponent of the three phonon scattering relaxation rate due to transverse phonon for class I events, whereas $m_{L,I}(T)$ and $m_{L,II}(T)$ are the same due to longitudinal phonons for class I and class II events respectively

T, K	$m_{T,I}(T)$	$m_{L,I}(T)$	$m_{L,II}(T)$
100	1.1272	1.7092	1.0
200	1.0551	1.2674	1.0
300	1.0341	1.1566	1.0
400	1.0243	1.1088	1.0
500	1.0188	1.0826	1.0
600	1.0152	1.0662	1.0
700	1.0127	1.0550	1.0
800	1.0109	1.0469	1.0
900	1.0095	1.0408	1.0
1000	1.0084	1.0361	1.0

Using the constants reported in Table 1, the temperature exponents $m_{T,I}(T)$, $m_{L,I}(T)$ and $m_{L,II}(T)$ have been calculated via Eqs (5) and (6) and the results obtained are reported in Table 4. Using these values of the temperature exponents, the total lattice thermal conductivity of LaS has been calculated in the entire

Table 5

The relative contributions due to transverse and longitudinal phonons for LaS in the temperature range 100 – 1000 K. % K_T is the percentage contributions due to transverse phonons and % K_L is the same due to longitudinal phonons

T, K	% K_T	% K_L
100	87.62	12.38
200	92.24	7.76
300	93.38	6.62
400	93.87	6.13
500	94.13	5.87
600	94.29	5.71
700	94.40	5.60
800	94.48	5.52
900	94.54	5.46
1000	94.59	5.41

temperature range 100–1000 K by estimating the separate contributions K_T and K_L due to transverse and longitudinal phonons, respectively, by means of numerical integration of the conductivity integrals in Eqs (8) and (9), and the results obtained are reported in Fig. 2. To study the relative contributions due to transverse and longitudinal phonons, the percentage contributions % K_T and % K_L are also calculated and the results obtained are listed in Table 5.

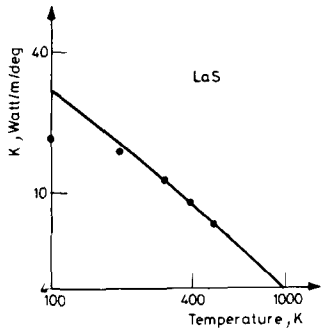

Fig. 2. The lattice thermal conductivity of LaS in the temperature range 100–1000 K. Solid line is the calculated value and circles are the experimental points

Results and discussion

From Figs 1 and 2, it can be seen that above 250 K the agreement between the calculated and the experimental values of the lattice thermal conductivity is good enough for both GdS and LaS. However, below 250 K the calculated values of the lattice thermal conductivity are larger than the experimental values for both of the samples, i.e. the calculated values of the lattice thermal resistivity are lower than the experimental values, which suggests the presence of some other scattering mechanisms too. With the help of the information reported on the samples by earlier workers, the possible scattering mechanisms are the scattering of phonons by electrons and the scattering of phonons by the paramagnetic ions. The possibility of electron-phonon scattering has been tested and it is found that the inclusion of the electron-phonon scattering processes can modify the results a little, but it can not explain the experimental data on the lattice thermal conductivities of GdS and LaS below 250 K. Thus, it can be concluded that to explain the experimental data on the lattice thermal conductivities of GdS and LaS below 250 K it is necessary to include the scattering of phonons by the paramagnetic ions, which is similar to the earlier prediction of Khusnutdinova *et al.* for GdS. Due to the lack of information about the constants relating to the scattering relaxation rate of phonon-paramagnetic ions, it has not been included in the present work.

From Table 3 and 5, it can be seen that the percentage contribution $\%\ K_T$ due to transverse phonons is much larger than the contribution due to longitudinal phonons, which predicts that at high temperatures the transverse phonons are mainly responsible for the transfer of heat, similar to the findings of the earlier workers. At the same time, with the help of these two Tables, it can also be concluded that the percentage contribution $\%\ K_T$ due to transverse phonons increases with increasing temperature in the range 100 – 1000 K, while the opposite is true for the longitudinal phonons.

The temperature exponents for the three-phonon scattering relaxation rates used in the present analysis can be studied *via* the data in Tables 2 and 4 GdS and LaS, respectively. From these two Tables, it can be seen that the temperature exponent $m(T)$ used in the present analysis is nearly unity for transverse as well as for longitudinal phonons, which is similar to the findings of the earlier workers. At the same time, it is necessary to state that the temperature exponents used in the present analysis of the lattice thermal conductivities of GdS and LaS are also free from the Guthrie comments.

It is interesting to note that during the numerical analysis of the conductivity integrals in Eq. (9) for the contribution K_L, it is found that the scattering relaxation rate due to class II events for longitudinal phonons is much larger than that due to class I events in the entire temperature range 100 – 1000 K, which is similar to the earlier findings of the author and his co-workers for other samples. It has also been observed that at high temperatures, the three-phonon scattering relaxation rate dominates over other scattering relaxation rates.

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RÉSUMÉ — On a analysé la conductivité thermique du réseau des sulfures des terres rares à températures élevées, dans le cadre de la conduction à deux modes de phonon en étudiant, pour la première fois, les conductivités thermiques totales du réseau de GdS et LaS dans tout l'intervalle des températures comprises entre 100 et 1000 K. On communique les exposants de température pour les vitesses de relaxation de dispersion à trois phonons, tant pour les phonons transversaux que pour les phonons longitudinaux. Les pourcentages de contribution séparés, dus aux phonons transversaux et longitudinaux, aux conductivités thermiques du réseau des échantillons considérés ont été déterminés de même. Le rôle des processus à quatre phonons est pris en compte dans la présente étude.

ZUSAMMENFASSUNG — Die Gitter-Wärmeleitfähigkeiten von Sulphiden der Seltenen Erden wurden bei hohen Temperaturen im Rahmen der Zwei-Arten Leitfähigkeit von Phononen zum ersten Mal durch das Studium der totalen Gitter-Wärmeleitfähigkeiten von GdS und LaS im Temperaturbereich von 100 bis 1000 K ermittelt. Die Temperaturexponenten für die Drei-Phononen-Streuungs-Relaxationsgeschwindigkeiten werden sowohl für die transversalen als auch für die longitudinalen Phonone angegeben. Die einzelnen prozentualen Beiträge der transversalen und longitudinalen Phonone zur totalen Gitter-Wärmeleitfähigkeit der obigen Proben wurden auf ähnliche Weise untersucht. Auch die Rolle der Vier-Phononen-Vorgänge wurde in die vorliegenden Untersuchungen mit aufgenommen.

Резюме — Проведен анализ решеточной термической проводимости для сульфидов щелочно-земельных металлов в рамах двух-фононовой проводимости первоначально изучением общей решеточной проводимости CdS и LaS в области температур 100—1000 К. Температурные экспоненты трех-фононовых релаксационных скоростей рассеяния приведены как для поперечных, так и продольных фононов. Ордельно изучен процентный вклад поперечных и продольных фононов в общую решеточную проводимость для вышеуказанных образцов. При изучении рассматривалась роль четырех фононовых процессов.