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*NONLINEAR ANALYSIS OF HEAT TRANSFER IN AN INSULATOR

APPLICATION TO NaBr

H. H. BOGHOSSIAN and K. S. DUBEY

Department of Physics, College of Science, University of Basrah, Basrah, Iraq

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Using nonlinear heat transfer theory and following Kazakov and Nagaev, a simple expression for the lattice thermal conductivity has been derived for an insulator having resonance-producing impurities. The expression obtained has been used to analyse the lattice thermal conductivity of pure and doped samples of NaBr in the entire temperature range 0.05–5 K. Good agreement has been found between the calculated and experimental values of phonon conductivity at low temperatures.

The lattice thermal conductivity of an insulator with finite dimensions at low temperatures has acquired considerable importance recently. At these temperatures the important scatterers of phonons are those due to boundary walls and impurities of the crystal, whereas anharmonicity in the crystal has been neglected totally because of its very small contribution to the phonon conductivity at low temperatures. It was Erdős [1] who first, took the problem to find an analytical expression for the lattice thermal conductivity. Later, Kazakov and Nagaev [2] (KN) calculated nonlinear heat transfer in a lattice containing impurites, using boundary conditions consistent with the experimental situations. It has normally been assumed that there is an arbitrary small deviation of the system from its thermodynamical equilibrium, which leads to the absence of local temperature and the linearization of the kinetic equation with respect to the temperature gradient.

Following KN and considering the nonlinear heat transfer theory, an analytical expression has been found for the lattice thermal conductivity by Dubey [3–5] for a crystal having dislocations. The calculations of KN as well as Dubey clearly show that one can study the effect of impurity scattering on the lattice thermal resistivity more rigorously than the Callaway [6] phenomenological approach. The Callaway approach is very complicated due to the fact that for a particular temperature there are different scattering processes acting simultaneously.

In the present work, our aim is to derive a simple expression for the lattice thermal conductivity of a sample having resonance impurities, using the nonlinear heat transfer theory of KN, and to test its applicability in explaining the experimental data of phonon conductivity. With the use of the obtained expression, the

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lattice thermal conductivities of pure and doped samples of NaBr have been calculated in the entire temperature range 0.05 – 5 K as an example. A comparative study is also made between results obtained in the frame of the present expression and results obtained on the basis of the Callaway integral.

Theory

Assuming the validity of additivity of the scattering relaxation rates, one can write the combined scattering relaxation rate $\tau_c^{-1}(\omega)$ for a sample having Rayleigh type impurities (isotopic) as

$$\tau_c^{-1}(\omega) = \tau_B^{-1} + \tau_{pt}^{-1}(\omega)$$

where τ_B^{-1} is the boundary scattering relaxation rate defined by Casimir [7], given by $\tau_B^{-1} = v/L$; L is the Casimir length of the crystal, v is the average phonon velocity and $\tau_{pt}^{-1}(\omega)$ is the point defect scattering relaxation rate, given by Klemens [8] as $\tau_{pt}^{-1}(\omega) = A\omega^4$; A is the point defect scattering strength. Following KN as well as Dubey and using nonlinear heat transfer theory, one can obtain an expression for the lattice thermal conductivity K:

$$K = K_0[1 - (LA/2v)(k_B T/\hbar)^4 I_2/I_1] \tag{2}$$

where

$$K_0 = (3k_B/2\pi^2 v)(k_B T/\hbar)^3 (L/v) I_1 \tag{3}$$

$$I_1 = \int_0^\infty x^3(e^x - 1)^{-1}\mathrm{d}x = 6.5 \tag{4}$$

$$I_2 = \int_0^\infty x^7(e^x - 1)^{-1}\mathrm{d}x = 5.06 \times 10^3 \tag{5}$$

k_B is the Boltzmann constant and $\hbar$ is the Planck constant divided by 2π.

Pohl and his coworkers [9, 10] gave a phenomenological expression for the relaxation rate of the scattering of phonons by tunneling levels and it was successfully applied by several workers to explain phonon conductivity data of such systems. Later, the resonance phonon scattering relaxation rate $\tau_r^{-1}(\omega)$ was further studied by Klein and his coworkers [11, 12] including the damping factor, and a very complicated expression was reported by them. Singh and Verma [13] commented upon the Klein expression and reported that the expression given by Pohl gives better results than the complicated Klein expression. Keeping in view the above reports and following Singh and Verma, we have used Pohl's expression for the resonance scattering relaxation rate in the present work. This can be expressed as

$$\tau_r^{-1}(\omega) = \frac{R_1(\omega/\omega_{r1})^2}{(1 - \omega^2/\omega_{r1}^2)^2} + \frac{R_2(\omega/\omega_{r2})^2}{(1 - \omega^2/\omega_{r2}^2)^2} \tag{6}$$

where all other constants R_1 and R_2, which are treated as adjustable parameters due to the lack of necessary data related to the resonance scattering relaxation rate for NaBr; ω_{r1} and ω_{r2} are the resonance frequencies. Thus, the combined scattering relaxation rate $\tau_c^{-1}(\omega)$ for the doped sample can be expressed as

$$\tau_c^{-1}(\omega) = \tau_B^{-1} + \tau_{pt}^{-1}(\omega) + \tau_r^{-1}(\omega). \tag{7}$$

In the frame of the nonlinear heat transfer theory, Eq. (7) gives an expression for the lattice thermal conductivity of the doped sample as

$$K_{imp} = K_0[1 - (LA/2v)(k_B T/\hbar)^4 I_2/I_1 - (LR_1/2v)(T/T_{R1})^2 I_3/I_1 - (LR_2/2v)(T/T_2)^2 I_4/I_1] \tag{8}$$

where $$I_3 = \int_0^{T_{R1}/T} x^5(1 - x^2T^2/(T_{R1}^2)^{-2}(e^x - 1)^{-1}\,dx \tag{9}$$

$$I_4 = \int_{T_{R1}}^{T_{R2}/T} x^5(1 - x^2T^2/(T_{R2}^2)^{-2}(e^x - 1)^{-1}\,dx \tag{10}$$

and $T_{Ri} = n\omega_{ri}/k_B$; $i = 1$ and 2.

In obtaining Eq. (8), it is assumed that phonons with frequencies $0 < \omega < \omega_{r1}$ and $\omega_{r1} < \omega < \omega_{r2}$ are interacting from tunneling levels with scattering strengths R_1 and R_2, whereas phonons with frequencies $\omega_{r2} < \omega < \omega_D$ do not interact from tunneling levels and these phonons are not affected by such impurities.

Phonon conductivity of NaBr

Rollefson [14] measured the phonon conductivities of NaF-doped and pure samples of NaBr and found that the phonon conductivities of two heavily doped samples (having different concentrations of F^- ions) were the same. He concluded that the reduction in the phonon conductivity of a doped sample of NaBr was not due to the presence of F^- ions. To examine the presence of any other impurities, he made ultra-violet absorption measurements on the sample and observed

Table 1

Values of constants and parameters used in the calculations of the phonon conductivity of NaBr in the temperature range 0.05–5 K.

v	$= 1.09 \times 10^5$ cm/sec
L	$= 0.0775$ cm
A	$= 9.5 \times 10^{-44}$ sec^3
Θ_D	$= 225$ K
T_{R1}	$= 0.54$ K
T_{R2}	$= 1.8$ K
R_1	$= 8.5 \times 10^4$ sec^{-1}
R_2	$= 8.5 \times 10^3$ sec^{-1}

the presence of OH^- ions having a concentration of the order of 10^{16} cm^{-3} (see Table 2 of ref. [14]). He suggested that the reduction in the phonon conductivity of a doped sample may be due to OH^- ions introduced during the crystal growth process as an unwanted impurity. In view of these observations and on analyzing Fig. 9 of ref. [14], Dubey [15] explained the measurements of Rollefson, introducing resonance scattering as an extra scattering mechanism in a sample doped due to OH^- ions. To test the applicability of the obtained expression in explaining the phonon conductivity data of an insulator, the measurements of Rollefson for pure and doped samples of NaBr are taken as an example.

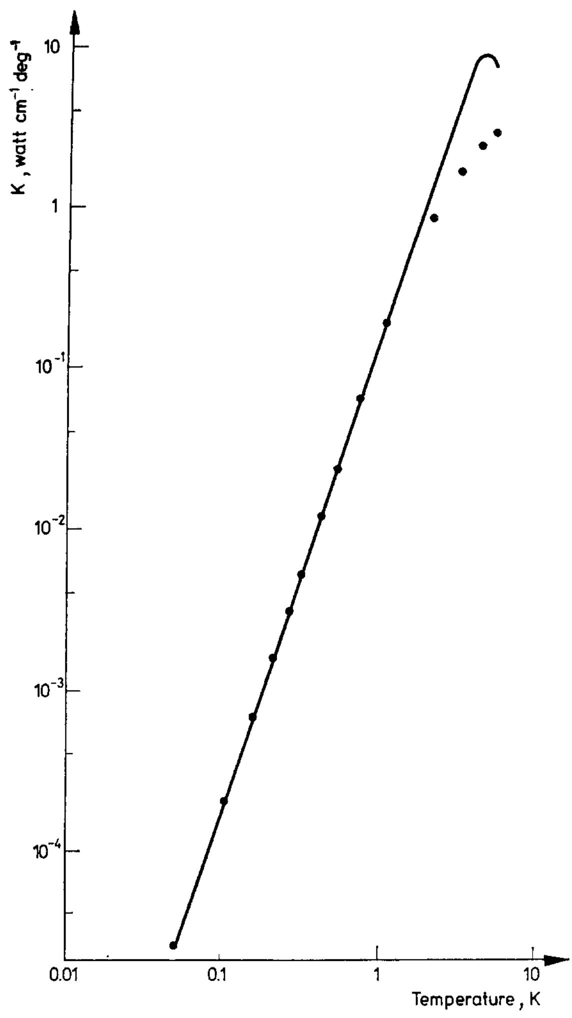

Fig. 1. Phonon conductivity of pure sample of NaBr in the temperature range 0.5–5 K. Solid line shows calculated values obtained in the frame of Eq. (2), and circles are the experimental points

Via calculation of the Casimir length L of the crystal and the point defect scattering strength A at 0.05 and 2 K, respectively, the phonon conductivity of a pure sample of NaBr has been calculated in the entire temperature range 0.05 – 5 K using Eq. (2), and the obtained value of the phonon conductivity is shown in Fig. 1. From a knowledge of the constants L and A, the resonance scattering strengths R_1 and R_2 are adjusted at 0.1 and 1K, respectively, whereas L and A remain constant because such impurities do not much affect these constants. The constants related to the resonance frequencies are taken from the earlier report of Dubey [15]. Thus, all the constants reported in Table 1 being known, the lattice thermal

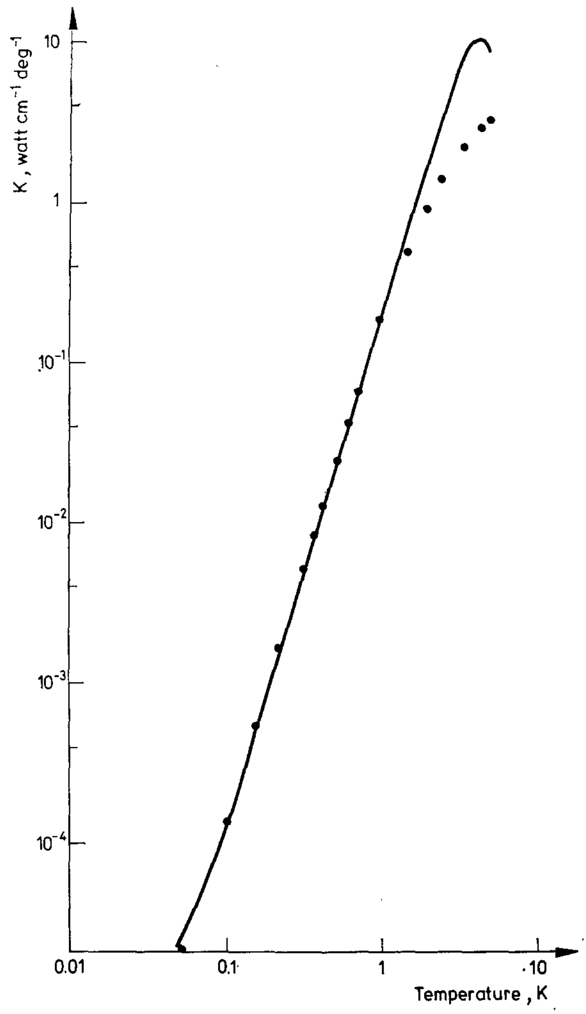

Fig. 2. Phonon conductivity of doped sample of NaBr in the temperature range 0.5 – 5 K. Solid line shows calculated values obtained in the frame of Eq. (8), and circles are the experimental points

conductivity of the doped sample of NaBr has been calculated in the entire temperature range 0.05 – 5K with the help of Eq. (8) and the results obtained are shown in Fig. 2. To make a comparative study of the present results obtained in the frame of Eq. (8) with the results obtained in the frame of Callaway integral, the earlier results of Dubey are also shown in Fig. 3, together with results calculated by using Eq. (8).

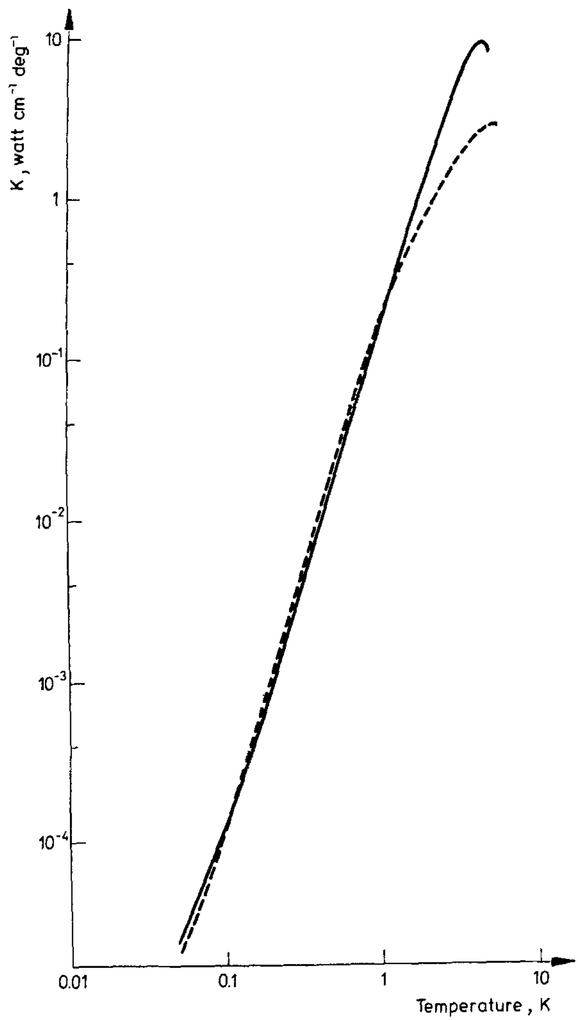

Fig. 3. A comparison between the present results and the earlier results of Dubey based on the phenomenological theory of Callaway for a doped sample of NaBr in the temperature range 0.05—5 K. Solid line represents results obtained in the present work using Eq. (8). Dotted line shows earlier results of Dubey based on Eq. (6) of ref. [15]

Results and discussion

With the use of nonlinear heat transfer theory and following KN as well as Dubey, an expression for the lattice thermal conductivity of a sample having resonance-producing impurities has been derived and is reported in Eq. (8). From Eq. (8), it is clear that the obtained expression is much simpler than the phenomenological and complicated expression given by Callaway. To see its applicability, phonon conductivities of pure and doped samples of NaBr have been calculated using the obtained expressions, and the results are shown in Figs 1 and 2.

From Figs 1 and 2, it can be concluded that the agreement between the calculated and experimental values of the phonon conductivity of both samples of NaBr, pure and doped, is satisfactory at low temperatures. However, there remain some discrepancies at high temperatures, which shows that the calculated values of the lattice thermal resistivity are less than the experimental values. This is due to the contribution of the three phonon scattering processes towards the total phonon resistivity, which has been totally ignored in the present calculation because of the validity of the KN theory, which can be applied only to those temperatures where the boundary scattering relaxation rate dominates over other scattering relaxation rates. Thus, the discrepancies shown in Figs 1 and 2 can be balanced by including the contribution due to the three phonon scattering processes.

From Fig. 3, it can be seen that at low temperatures the results obtained in the frame of the obtained expression are very close to values obtained by Dubey based on the Callaway theory. At high temperatures, however, the results do not match, which is due to the contribution of the three phonon scattering processes. Hence, we conclude that the obtained expression gives good results at low temperatures, where boundary scattering relaxation rate dominates over other relaxation rates. At the same time, it is more simpler than the Callaway expression.

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Résumé — En se servant de la théorie du transfert de chaleur non-linéaire et suivant Kazakov et Nagaev, on a trouvé une expression simple pour la conductibilité thermique du réseau dans le cas d'un isolant à impuretés produisant la résonance. L'expression obtenue a été utilisée pour analyser la conductibilité thermique du réseau d'échantillons de NaBr purs ou contenant des additifs, dans tout l'intervalle de température allant de 0.05 à 5 K. Un bon accord a été observé entre les valeurs expérimentales et calculées de la conductibilité des phonons aux basses températures.

Zusammenfassung — An Hand der nicht linearen Wärmeübertragungstheorie, sowie nach Kazakow und Nagaew wurde ein einfacher Ausdruck für die thermische Gitterleitfähigkeit für ein Isolator mit resonanzbildenen Verunreinigungen abgeleitet. Der erhaltene Ausdruck wurde zur Analyse der Gitter-Wärmeleitfähigkeit reiner und Zusatzstoffe enthaltender NaBr-Proben im ganzen Temperaturbereich von 0.05 bis 5 K eingesetzt. Eine gute Übereinstimmung der berechneten und Versuchswerte der Phononleitfähigkeit wurde bei niedrigen Temperaturen erhalten.

Резюме — Используя теорию нелинейного теплового переноса и согласно Казакову и Нагаеву, было выведено простое выражение для решеточной удельной теплопроводности изоляторов, имеющих резонансно-наведенные примеси. Полученное выражение было использовано для анализа решеточной удельной теплопроводности чистых и легированных образцов бромистого натрия во всей температурной области 0.05—5 K. Найдено хорошее согласие между экспериментальными и вычисленными значениями фононовой проводимости при низких температурах.