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Comments on Klein's Theory of the Thermal Conductivity of Doped Alkali Halides

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Klein's theory of the resonant phonon scattering from a two-level tunneling system has been applied to the KCl : CN⁻ system. Unlike the KCl : OH⁻ and NaCl : OH⁻ systems, agreement between theory and experiment is achieved with $n = 1$. It is concluded that the scattering due to the mass defect ($n = 2$) and the scattering due to force-constant changes ($n = 4$) do not play a significant role in causing the resonance dips in the K-vs-T curves in KCl : CN⁻ systems.

Recently Klein[1] has discussed the resonant phonon scattering from a two-level tunneling system. For a crystal with two atoms per unit cell, the result in the Lorentzian approximation is

$$\tau_r^{-1} = \frac{pdS(T)}{3\pi\rho(\omega_r)} \; \frac{\gamma(T)\gamma(0)(\omega/\omega_r)^{2n}}{(1-\omega^2/\omega_r^2)^2 + \gamma^2(T)(\omega/\omega_r)^{2n+2}} \; . \tag{1}$$

Here p is the fractional impurity concentration, d is the degeneracy of the resonant mode, $\rho(\omega)$ is the (unperturbed) phonon density of states normalized to unity, ω_r is the resonance angular frequency, $\gamma = \Gamma(\omega_r)/\omega_r^2$ is the dimensionless halfwidth at $\omega = \omega_r$, and $S(T)$ is the temperature-dependent strength of the resonance. $\Gamma(\omega)$ is given by $\Gamma(\omega) = \omega_r^2(\omega/\omega_r)^{n+1}\gamma$, where $n = 2$ for a substitutional mass defect and $n = 4$ for an odd-parity pure force-constant resonance. Rosenbaum, Chau, and Klein[2] explained the resonance dips in the K-vs-T curves of the KCl : OH⁻ and NaCl : OH⁻ systems by taking $n = 2$.

Application of the above theory to KCl : CN⁻ systems, however, leads to some interesting conclusions. For a system in which there occur two resonance dips which are quite far apart from each other, one can write the resonance-scattering relaxation rate as

$$\tau_r^{-1} = \frac{p}{3\pi}\left(\frac{d_1S_1(T)}{\rho(\omega_1)} \; \frac{\gamma_1(T)\gamma_1(0)(\omega/\omega_1)^{2n}}{(1-\omega^2/\omega_1^2)^2 + \gamma_1^2(T)(\omega/\omega_1)^{2n+2}} + \frac{d_2S_2(T)}{\rho(\omega_2)} \; \frac{\gamma_2(T)\gamma_2(0)(\omega/\omega_2)^{2n}}{(1-\omega^2/\omega_2^2)^2 + \gamma_2^2(T)(\omega/\omega_2)^{2n+2}}\right). \tag{2}$$

For the KCl : CN⁻ system[3] $\omega_1 = 3\times10^{11}$ sec⁻¹ and $\omega_2 = 3.54\times10^{12}$ sec⁻¹. Thus the above equation contains d_1, d_2, $S_1(T)$, $S_2(T)$, $\gamma_1(0)$, $\gamma_2(0)$, $\gamma_1(T)$, and $\gamma_2(T)$ as adjustable parameters. To a fair approximation one can take $S_1(T) = S_2(T) = 1$ and $d_1 = d_2 = 3$. Further, $\rho(\omega)$ for KCl is given by[2] $\rho(\omega) \simeq 0.75\times10^{-40}\omega^2$. After these simplifications, calculations were made with $n = 2$ and 4 and adjustable temperature-independent γ's. The calculated values produced curves that had the dips in the wrong places, and the quantitative agreement was also very poor. Even temperature-dependent γ's could not give better agreement. However, if one takes $n = 1$, excellent agreement is obtained between theory and experiment[3,4] for KCl : CN⁻ system. The resonance-scattering relaxation rate is given by

$$\tau_r^{-1} = p\left(\frac{8.9792\times10^{15}\gamma_1^2x^2T^2}{(1-0.1904x^2T^2)^2 + 3.6255\times10^{-2}\gamma_1^2x^4T^4} + \frac{4.6308\times10^{11}\gamma_2^2x^2T^2}{(1-1.3674\times10^{-3}x^2T^2)^2 + 1.8698\times10^{-6}\gamma_2^2x^4T^4}\right) . \tag{3}$$

The combined relaxation rate for pure KCl is the same as has been used by Walker and Pohl,[5] namely,

$$\tau_{\text{pure}}^{-1} = 4.9\times10^5 + 17.83x^4T^4 + 6.24\times10^4x^2T^3e^{-(40/T)}, \tag{4}$$

where T is in °K.

Since one can explain the experimental results in the KCl : CN⁻ system by taking $n=1$, the present

TABLE I. Values of the parameters used in the analysis of the phonon-conductivity data on the KCl : CN⁻ system.

Sample	Concentration of CN⁻ ions (cm⁻³)	Concentration of CN⁻ ions (ppm)	γ_1^2 (10^{-5})	γ_2^2 (10^{-2})
Curve B	9×10^{17}	50	5.62	5.81
Curve C	8.4×10^{18}	466	6.80	8.41
Curve D	4.9×10^{19}	2722	4.76	4.84

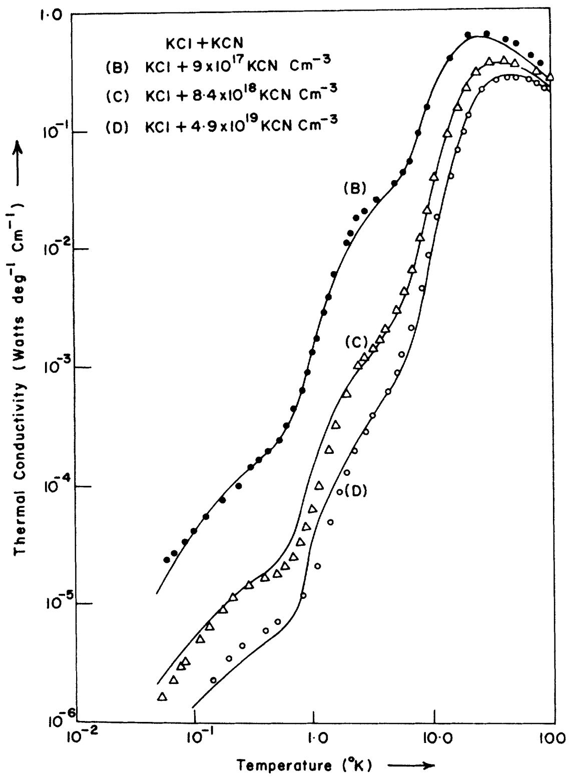

FIG. 1. Thermal conductivity of KCl : CN after Seward and Narayanmurthi (Ref. 3), extended to 0.05 °K by Harrison and Peressini (unpublished data). The solid curves are the theoretical curves calculated on the basis of Eq. (3).

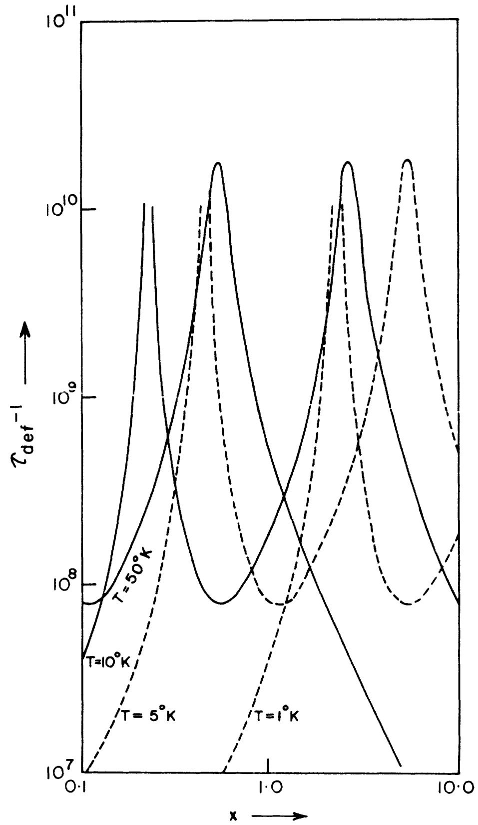

FIG. 2. Curves for τ_r^{-1} vs x at four different temperatures for KCl + (9×10^{17} KCN cm^{-3}) as given by Eq. (3) with $\gamma_1^2 = 5.62\times10^{-5}$ and $\gamma_2^2 = 5.81\times10^{-2}$. Note $\tau_{def}^{-1} = \tau_r^{-1}$.

calculations suggest that the scattering due to the difference in mass as well as that due to changes in the force constant is not significant in causing the resonance dips in the K-vs-T curves in KCl : CN⁻ systems. However, it does play an important role in the KCl : OH⁻ and NaCl : OH⁻ systems, as is evident from the calculations of Rosenbaum, Chau, and Klein.

In view of the fact that the present calculations make use of Klein's theoretical expressions, which include damping terms explicitly, and not of phenomenological expressions, the present comparison between theory and experiment is more meaningful. The experimental results of Seward and Narayanmurthi for KCl : CN⁻ systems have been extended from 0.3 °K to 0.05 °K in the low-temperature range by Harrison and Peressini.[6] This brings out the resonance nature of the dip occurring at 0.6 °K more prominently. The earlier calculations,[3,7] based on the phenomenological expression $\tau_r^{-1} \propto \omega^2/(\omega_r^2 - \omega^2)^2$, were limited to 0.3 °K only, the lowest temperature at which the phonon conductivity was measured by Seward and Narayanmurthi. The present calculations cover the temperature range up to 0.05 °K. The results of calculations are shown in Fig. 1. The values of the adjustable parameters γ_1^2 and for γ_2^2 for KCl : CN⁻ systems containing different concentrations of CN⁻ ions are given in Table I. Typical curves for τ_r^{-1} vs $x = \hbar\omega/k_B T$ are also shown in Fig. 2.

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[1] M. V. Klein, Phys. Rev. 186, 839 (1969).

[2] R. L. Rosenbaum, Cheuk-Kin Chau, and M. V. Klein, Phys. Rev. 186, 852 (1969).

[3] W. D. Seward and V. Narayanmurthi, Phys. Rev. 148, 463 (1966).

[4] V. Narayanmurthi and R. O. Pohl, Rev. Mod. Phys. 42, 201 (1970).

[5] C. T. Walker and R. O. Pohl, Phys. Rev. 131, 1433 (1963).

[6] Harrison and Peressini (unpublished data). See Fig. 8 on p. 207 of Ref. 4.

[7] A. Kumar, A. K. Srivastava, and G. S. Verma, Phys. Rev. 178, 1480 (1969).