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Three-Phonon Scattering Strengths and Ziman Limit of Resistivity Due to Three-Phonon Scattering Processes in Ge

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The contributions of different three-phonon scattering processes to the thermal resistivity of isotopically pure Ge are presented in the Ziman limit. The modified expressions of Hamilton and Parrott for three-phonon scattering strengths are considered.

I. INTRODUCTION

Using a variational treatment, Hamilton and Parrott[1] have recently estimated the contribution of different scattering processes to the thermal resistivity of Ge. They have considered an anharmonic isotropic continuum model. The grafting of umklapp (U) processes in the Debye way of specific heats has made their model much more rigorous, but unfortunately their expression for three-phonon scattering strengths are slightly in error.[2] As a result their calculated values are incorrectly estimated. It was, therefore, considered desirable to check at least the Ziman limit on the phonon resistivity due to three-phonon scattering processes. It is encouraging to note that one obtains reasonable values of the Ziman limit with the modified expressions of three-phonon scattering strengths. In the Appendix we also present the results of calculation of three-phonon scattering strengths for a simple cubic crystal. In spite of the several drastic assumptions made in the derivation for a simple cubic crystal, it is interesting to note that the results reduce to correct expressions of three-phonon scattering strengths for an isotropic case.

II. THEORY

The upper bound of the Ziman limit on the thermal resistivity $W_0^>$, due to a three-phonon process of the type $s+s' \rightleftharpoons s''$, is given by[3]

$$(W_0^>)_{ss's''} = \frac{N_0\Omega}{k_0} \frac{\langle \Phi, P\Phi\rangle_{ss's''}}{[\sum_s \langle \Phi, X\rangle_s]^2} . \quad (1)$$

The phonon-phonon collision operator $P_{ss'}^{\vec{q}\vec{q}'}$ due to a three-phonon process of the type $s+s' \rightleftharpoons s''$ is given by the relation[1,4]

$$P_{ss'}^{\vec{q}\vec{q}'} = \sum_{\vec{q}'',s''} [\tfrac{1}{2}\delta_{ss'}\delta_{\vec{q}\vec{q}'}(\bar{P}_{\vec{q}s,\vec{q}''s''}^{\vec{q}''s'''} + \bar{P}_{\vec{q}''s'',\vec{q}'''s'''}^{\vec{q}s} + \bar{P}_{\vec{q}''s'',\vec{q}s}^{\vec{q}'''s'''}) - \delta_{s's''}\delta_{\vec{q}'\vec{q}''}(\bar{P}_{\vec{q}s,\vec{q}''s''}^{\vec{q}'''s'''} + \bar{P}_{\vec{q}''s'',\vec{q}'''s'''}^{\vec{q}s} - \bar{P}_{\vec{q}''s'',\vec{q}s}^{\vec{q}'''s'''})] . \quad (2)$$

The equilibrium transition probability $\bar{P}_{\vec{q}s,\vec{q}'s'}^{\vec{q}''s''}$ is given by[4]

$$\bar{P}_{\vec{q}s,\vec{q}'s'}^{\vec{q}''s''} = \frac{\pi\hbar}{4\rho^3 N_0\Omega} \frac{\vec{q}^2\vec{q}'^2\vec{q}''^2}{\omega\omega'\omega''} |A_{ss's''}^{\vec{q}\vec{q}'\vec{q}''}|^2 N N'(\bar{N}''+1) \times \delta(\omega''-\omega-\omega')\delta_{\vec{q}+\vec{q}',\vec{q}''+\vec{G}} , \quad (3)$$

where N_0 is the number of unit cells, Ω is the volume of a unit cell, $\vec{G}$ is a reciprocal-lattice vector, $|A_{ss's''}^{\vec{q}\vec{q}'\vec{q}''}|^2$ is the three-phonon scattering strength for the process $s+s' \rightleftharpoons s''$, and $\bar{N}$ is the equilibrium distribution function related to the deviation function ϕ by the expressions[3] $N=\bar{N}+\psi\bar{N}(\bar{N}+1)$ and $\phi=\psi+\delta\psi$, where N is the distribution function. The denominator in (1) can be related to the Boltzmann equation in the form

$$\vec{X} = P\vec{\psi} , \quad (4)$$

with its left-hand side as

$$X_s^{\vec{q}} = -\frac{\vec{v}_s(\vec{q})\cdot\vec{\nabla}T}{T} \frac{\hbar\omega}{k_0 T} \bar{N}(\bar{N}+1) , \quad (5)$$

where $\vec{\nabla}T$ is the temperature gradient and k_0 is the Boltzmann constant.

The correct expressions for the three-phonon scattering strengths are as follows:

$$|A^{\vec{q}\vec{q}'\vec{q}''}_{TTT}|^2 = 0\,, \tag{6a}$$

$$|A^{\vec{q}\vec{q}'\vec{q}''}_{LLL}|^2 = [3\lambda + 6\mu + 2(A + 3B + C)]^2\,, \tag{6b}$$

$$|A^{\vec{q}\vec{q}'\vec{q}''}_{TLL}|^2 = \tfrac{1}{2}(\lambda + 3\mu + A + 2B)^2 \sin^2(\theta' + \theta'') \times \cos^2(\theta'' - \theta')\,, \tag{6c}$$

$$|A^{\vec{q}\vec{q}'\vec{q}''}_{TTL}|^2 = \tfrac{1}{16}\{[\lambda + \mu + \cos 2\theta'(\lambda + 3\mu + A + 2B)]^2 + [2\cos\theta'(\lambda + B) + \cos\theta'' \cos(\theta'' - \theta')(4\mu + A)]^2\}\,. \tag{6d}$$

III. RESULTS

In the evaluation of $(W_0^{>})_{ss's''}$ for different processes, the integrals have been evaluated using the Gaussian quadrature method. Use of five pivots has been made. The limits of integrations are the same as those used by Hamilton and Parrott. The trial function is chosen as $\phi = \vec{q}\cdot\hat{u}$, where $\hat{u} = -\vec{\nabla}T/T$. The Debye radius is calculated from the formula $q_D = (6\pi^2/\Omega)^{1/3}$, where $\Omega = (\frac{1}{2}a)^3$ and a is the lattice parameter of Ge. The use of modified expressions for $|A^{\vec{q}\vec{q}'\vec{q}''}_{ss's''}|^2$ ensures a correct picture of the results.

Table I shows the comparison of our calculated values of the Ziman limit of thermal resistivity due to three-phonon processes with the experimental values of thermal resistivity as determined by Slack and Glassbrenner.[5] The theoretical values of the thermal resistivity due to the three-phonon processes alone, as determined by Hamilton and Parrott, are also given in Table I. In their calculations different terms of the expansion of the trial function up to $R = 2$ are considered. It may be seen from Table I that, except at 100 °K, our values of the Ziman limit of thermal resistivity are in general higher than the experimental values, which should be the case. The low value of three-phonon resistivity at 100 °K shows that the mass-defect scattering should also be taken into account. In our calculation the deviation between the calculated and the experimental values of phonon resistivity decreases with the increase in temperature. This is expected because in the high-temperature region, three-phonon processes are expected to make a major contribution to the thermal resistivity. This, however, is not true for the calculations of Hamilton and Parrott as may be seen from Fig. 1.

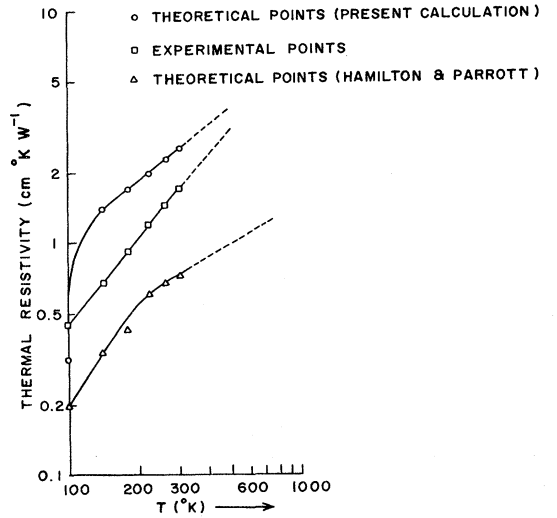

FIG. 1. Comparison of the theoretical values of the thermal resistivity due to three-phonon processes with the experimental values in Ge in the temperature range 100–300 °K.

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APPENDIX: THREE-PHONON SCATTERING STRENGTHS

The results of calculations of three-phonon scattering strengths for simple cubic crystals are as follows:

$$|A^{\vec{q}\vec{q}\vec{q}''}_{TTT}|^2 = 0\,, \tag{7a}$$

$$|A^{\vec{q}\vec{q}'\vec{q}''}_{LLL}|^2 = 9(c_{11} + c_{111})^2\,, \tag{7b}$$

$$|A^{\vec{q}\vec{q}'\vec{q}''}_{TLL}|^2 = \tfrac{1}{2}\{\sin(\theta' + \theta'')[c_{11}\cos\theta'\cos\theta'' + (2c_{44} + c_{12})\sin\theta'\sin\theta'' + (c_{166} + c_{44})\cos(\theta'' - \theta')]\}^2\,, \tag{7c}$$

$$|A^{\vec{q}\vec{q}'\vec{q}''}_{TTL}|^2 = \tfrac{1}{16}\{[c_{166}\cos 2\theta' + 2c_{11}\cos\theta'\cos\theta''\cos(\theta' + \theta'') - 2c_{44}\sin^2\theta' + (4c_{44} + 2c_{12})\cos\theta'\sin\theta''\sin(\theta' + \theta'')]^2 + \tfrac{1}{4}[(c_{166} + 2c_{11} - 2c_{12} - c_{144})\cos\theta'\cos 2\theta'' + (4c_{44} + c_{456})\sin\theta'\sin 2\theta'' + (c_{166} + 2c_{11} + 2c_{12} + c_{144})\cos\theta']^2\}\,. \tag{7d}$$

In order to evaluate $|A^{\vec{q}\vec{q}'\vec{q}''}_{ss's''}|^2$, one has to note that it depends on the angles between the vectors $\vec{q}$, $\vec{q}'$, and $\vec{q}''$ and also on the limitations which the crystal imposes on the sound-wave propogation. The first condition is handled by taking the three phonons to be coplanar. The second requirement is a tough one. We know that in the case of a cubic crystal there is no meaning at all to a pure transverse or a pure longitudinal phonon except in the [100], [110], and [111] directions. In order to

TABLE I. Ziman limit of the resistivity due to different three-phonon scattering processes (in units of cm °K W^{-1}).

Temp. (°K)	$(W_0^>)_{LLL}$	$(W_0^>)_{NTLL}$	$(W_0^>)_{NTTL}$	$(W_0^>)_{UTLL}$	$(W_0^>)_{UTTL}$	$W_0^>$	$W_{expt.}$[a]	W_{HP}[b]
100	0.0679	0.0480	0.1326	0.0099	0.0624	0.3208	0.456	0.198[c]
140	0.2552	0.2041	0.5761	0.0622	0.3330	1.4306	0.684	0.345
180	0.2729	0.2242	0.7625	0.0834	0.4141	1.7571	0.930	0.435
220	0.3017	0.2513	0.8925	0.1038	0.4932	2.0425	1.220	0.595
260	0.3353	0.2817	1.0270	0.1235	0.5727	2.3402	1.471	0.637
300	0.3718	0.3142	1.1630	0.1432	0.6531	2.6453	1.724	0.725[c]

[a]Reference 4.
[b]Reference 1.
[c]See Table II of Ref. 1; other values are read from Fig. 2 of Ref. 1.

make the problem tractable, we have made a drastic assumption that pure longitudinal and pure transverse waves do exist in a cubic crystal for all directions of the wave vector ($\vec{q}$, or $\vec{q}'$ or $\vec{q}''$), and that the direction of the transverse polarization vector is arbitrary. Furthermore, we know that in the case of cubic symmetry, except for waves propagating in the [100] and [111] directions, it is no longer possible to regard the two transverse waves as degenerate. But for the convenience of calculations, we assume that the two transverse waves are degenerate. These assumptions allow us to calculate the three-phonon scattering strengths for the processes $L+L \rightleftharpoons L$, $T+L \rightleftharpoons L$, and $T+T \rightleftharpoons L$ for a simple cubic crystal.

For the isotropic case, we use the following conditions:

$$c_{111} = \tfrac{1}{6}(c_{123} + c_{456} + 3c_{144}) , \tag{8a}$$

$$c_{112} = \tfrac{1}{2}(c_{123} + c_{144}) , \tag{8b}$$

$$c_{166} = c_{144} + \tfrac{1}{2} c_{456} , \tag{8c}$$

$$c_{44} = \tfrac{1}{2}(c_{11} - c_{12}) . \tag{8d}$$

Furthermore, expressing

$$c_{123} = 2C , \tag{9a}$$

$$c_{144} = 2B , \tag{9b}$$

$$c_{456} = 2A , \tag{9c}$$

$$c_{11} = \lambda + 2\mu , \tag{9d}$$

$$c_{12} = \lambda , \tag{9e}$$

we obtain Eqs. (6a)–(6d) for three-phonon scattering strengths, which have been obtained for the isotropic case.

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