Phonon conductivity of plastically deformed crystals: Role of stacking faults and dislocations

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We suggest that additional depression of the phonon conductivity κ at its peak value due to plastic deformation of close-packed structures might well be caused by phonon scattering by stacking faults and the dislocation cores. These two scattering processes would give rise to respectively ω^2 and ω^3 dependent relaxation rates in the full relaxation rate τ_F^{-1} resulted by various phonon-scattering processes. We achieve an excellent theoretical fit for the experimental results of phonon conductivity κ in plastically deformed LiF and Ge. The number of stacking faults/cm N_s and the radius of the core size of dislocations are strikingly within reasonable limits. Further, the computed value of N_s obtained in deformed Ge is an order less than in case of LiF for nearly the same dislocation density. This result is also theoretically expected as Ge is less close packed than LiF.

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I. INTRODUCTION

It is observed that the depression of phonon conductivity κ due to plastic deformation of crystals occurs not only at very low temperatures but to some extent extends up to temperatures beyond its peak value.[1–3] The problem is supposed to be challenging because the theory of most probable additional phonon-dislocation interaction in such systems is unable to account for this depression in κ.[1–4]

Phonons may be scattered by dislocations[5,6] due to their (i) static strain field, (ii) dynamic nature, and (iii) cores. As regards scattering of phonons due to static strain field of dislocations the theory of Klemens[6,7] and others[8] usually falls at least an order smaller than demanded by the experimental data. Ohashi's[9] theory yielded better results but it was sharply criticized by Granato.[10] Granato[10] and his co-workers[4] developed the theory for the phonon scattering by dynamic pinned as well as unpinned dislocations while Ninomiya,[11] adopting a different approach, solved the same problem for the case of unpinned dislocations only. Based on these theories of processes (i) and (ii), many workers analyzed the phonon-conductivity data of plastically deformed samples of Ge,[2] Cu-Al alloys,[12] and LiF.[1,4] From this literature it may be concluded that neither (i) nor (ii) can singly explain the above-mentioned depression of κ quantitatively even at low temperatures (<8 K). Roth and Anderson[3] and others[13–15] showed that the combination of the processes (i) and (ii) gives rise to a much better explanation of the experimental data also. It was almost settled that although the dynamic nature of this scattering process dominates over the static one, its strength still falls short by a factor of 2 at least in the case of LiF.[1,4]

Amidst the controversy to settle down the nature of phonon scattering by dislocations, which is most effective at low temperatures (<8 K), the process (iii), i.e., phonon scattering by dislocation cores, was totally ignored. According to Berman[5] and Klemens[6] its strength becomes significant when the phonon frequency is of the order of Debye frequency and hence may not be ignored at higher temperatures ($T>8$ K). In fact few workers[1,2,14] have extended the phonon conductivity data of deformed samples beyond its peak value and few attempts[2,14,15] have been made to explain it. Whereas Sato and Sumino[2] imagined the presence of a large number of vacancy clusters to explain this depression, Kumar *et al.*[15] used the interference between phonon-point defect and phonon-dislocation scattering for the same. In the following sections we disagree with these authors and show that the hitherto neglected phonon scattering by stacking faults as well as dislocation cores might well be the main cause behind the depression of phonon conductivity at its peak value. A discussion over another ignored process phonon scattering by grain boundaries is also presented in Sec. III.

II. THEORY

To explain κ of deformed Ge, Sato and Sumino[2] imagined presence of large number of vacancy clusters introduced during compression, which is not logical. They themselves agree that presence of the required high density of vacancies in deformed Ge is very doubtful because the samples were first annealed at $T=600$ °C before they were compressed. Even by taking a large number of vacancy clusters they could not explain their experimental data of plastically deformed Ge above 10 K; their theory overestimates κ much near the peak (~ 20 K), i.e., still higher concentration of vacancies will have to be assumed in their effort. Kaburaki *et al.*[14] added a relaxation rate $\propto\omega^2$ without clearly describing the cause of its origin and obtained quite a good fit with experimental data. Kumar *et al.*[15] associated ω^2 term with the interference between the point defect and the phonon dislocation (static strain) scattering. Kumar *et al.* made a drastic assumption that all the isotopic point defects are completely overlapping with dislocations. This assumption would automatically give rise to a much enhanced value of the interference term. We have shown[16] through a simpler case of elastic phonon-mass defect and phonon-electron scattering that if the two impurity centers are not overlapping, the interference term would be vanishingly small. Therefore we tend to disagree with these ideas and associate this ω^2 dependent term with the

phonon scattering by stacking faults ignored by all previous workers. We propose that the total relaxation rate τ_{def}^{-1} due to additional scattering processes in plastically deformed crystals should be given as

$$\tau_{\mathrm{def}}^{-1} = \tau_{\mathrm{sf}}^{-1} + \tau_{\mathrm{dc}}^{-1} + \tau_{\mathrm{ds}}^{-1} + \tau_{\mathrm{dd}}^{-1}, \tag{1}$$

where dd, ds, dc, and sf, respectively, stand for the scattering due to the dynamic nature, static strain field, dislocations cores, and stacking faults. In close-packed structures like LiF and Ge the splitting of mobile dislocations into partial dislocations and their reunion continuously takes place. Thus we expect a substantial number of partial dislocations, leaving a connecting sheet across which the sequence of close-packed structure is disturbed which is called stacking faults.[6] To investigate the role of phonon scattering by stacking faults in phonon conductivity of plastically deformed crystals we make use of the corresponding relaxation rate τ_{sf}^{-1} introduced by Klemens.[6] Considering only the specular reflection of phonons at stacking faults Klemens[6] found that

$$\tau_{\mathrm{sf}}^{-1} = 0.7\frac{a^2}{v}\gamma^2\omega^2 N_{\mathrm{s}} = D_{\mathrm{sf}}\omega^2, \tag{2}$$

where the stacking faults strength $D_{\mathrm{sf}}=0.7\frac{a^2}{v}\gamma^2 N_{\mathrm{s}}$, γ being the Grunessien's constant, N_{s} is the number of stacking faults per cm, a is the lattice parameter, ω is the angular frequency, and v is the phonon velocity. Earlier this scattering was ignored probably because generally all workers concentrated on the low-temperature region where ω^2 dependence of the relaxation rate would not give rise to T^2 to $T^{3.5}$ dependence of the phonon conductivity. The choice of the last two terms in Eq. (1) has already been justified by Roth and Anderson,[3] Klemens,[6] and Kumar *et al.*[15] The expression of τ_{ds}^{-1} due to Klemens[6] has found support on the basis of experimental work by Anderson[17] and its almost unanimously accepted form is given by

$$\tau_{\mathrm{ds}}^{-1} = N_{\mathrm{d}}b^2\gamma^2\omega\left(\frac{2^{3/2}}{3^{7/2}}\right) = D_{\mathrm{ds}}\omega, \tag{3}$$

where the static dislocation strength $D_{\mathrm{ds}}=N_{\mathrm{d}}b^2\gamma^2\left(\frac{2^{3/2}}{3^{7/2}}\right)$, N_{d} is the number of dislocations per unit area, and b is the Burger vector. If the dislocations are arranged at random, τ_{ds}^{-1} is reduced by a factor $\sim$0.55. In the case of τ_{dd}^{-1} instead of going for the complicated expression introduced by Ninomiya[11] and later somewhat simplified by Suzuki and Suzuki[1] and Klemens,[7] we opt for a simpler expression,

$$\tau_{\mathrm{dd}}^{-1} = \frac{D_{\mathrm{dd}}}{\omega}, \tag{4}$$

which is an approximate expression for phonon scattering by unpinned dislocation,[11] here D_{dd} is the strength of dynamic dislocation. The above choice can be justified on the basis of the work of Kneezel and Granato.[4] They have shown that even in the case of phonon scattering by pinned dislocations, the fundamental frequency ω_1 of these vibrating pinned dislocations is so small that the resonance effect in κ vs T data is visible at $T\sim 0.07-0.1$ K in LiF. Since our data start from $T=2$ K, the dominant heat carriers have frequency $\gg\omega_1$ at which the expression of τ_{dd}^{-1} can easily be approximated to Eq. (4) in both cases of phonon scattering by fluttering unpinned or vibrating pinned dislocations.[1,4]

The second term in Eq. (1) is due to the phonon scattering by dislocation cores. The expression of τ_{dc}^{-1} can be written[5,6,18] as

$$\tau_{\mathrm{dc}}^{-1} = N_{\mathrm{d}}\frac{r^4\omega^3}{v_{\mathrm{p}}^2} = D_{\mathrm{dc}}\omega^3, \tag{5}$$

where dislocation core strength $D_{\mathrm{dc}}=N_{\mathrm{d}}\frac{r^4}{v_p^2}$, r is the radius of dislocation core, and v_{p} is the phonon phase velocity. It is ω^3 dependent and therefore neglected by other workers due to the same reason for which scattering by stacking faults was dropped. Klemens[6] and Berman[5] have shown that around $\omega\sim 10^{13}$/sec, the magnitude of this process would be of the order of τ_{ds}^{-1}. So it would be desirable to investigate its role in the depression of the phonon conductivity at $T>20$ K where the phonons with $\omega\sim 10^{13}$/sec may not be ignored in the total heat transport.

III. RESULTS AND DISCUSSION

The calculation of the phonon conductivity of plastically deformed samples of LiF and Ge are performed on the basis of our earlier model[19] in which the correction term has also been included. The relevant formula is summarized in the Appendix. During the computation process it was noticed that together with $1/\omega$, ω, ω^2, and ω^4 dependent terms in τ_{F}^{-1} it was essential to include the extra ω^3 dependent term to produce the experimental data from $T=2$ to 50 K. The origin of $1/\omega$, ω, and ω^4 dependent terms are already discussed after Eq. (1). A survey of the literature revealed that the ω^3 dependent term may qualitatively be associated with the Rayleigh type of phonon scattering by dislocation cores. Also, following our discussion in theory Sec. II we prefer to associate the ω^2 dependent term with phonon scattering by stacking faults. Both of these interpretations will be justified later in this section.

TABLE I. Calculated values of strength parameters of different dislocation scattering used in the calculation of Figs. 1 and 2 for LiF. The estimated value of D_{ds} by Klemens (Ref. 6) formula is given in the brackets.

Sample B Dislocation density $=1.3\times10^8$ cm^{-2}	Sample C Dislocation density $=3.5\times10^8$ cm^{-2}
D_{dd} (dynamic) $=1.22\times10^8$ sec^{-1} K	D_{dd} (dynamic) $=1.65\times10^8$ sec^{-1} K
D_{ds} (static) $=4.5\times10^4$ sec^{-1} K^{-1} (8.46×10^3 sec^{-1} K^{-1}) (Ref. 6)	D_{ds} (static) $=6.75\times10^5$ sec^{-1} K^{-1} (2.27×10^4 sec^{-1} K^{-1})(Ref. 6)
D_{sf} (stacking faults) $=8.95\times10^3$ sec^{-1} K^{-2}	D_{sf} (stacking faults) $=3.2\times10^4$ sec^{-1} K^{-2}
D_{dc} (core) $=1.27\times10^1$ sec^{-1} K^{-3}	D_{dc} (core) $=1.40\times10^1$ sec^{-1} K^{-3}

TABLE II. Calculated values of strength parameters of different dislocation scattering used in the calculation of Figs. 3 and 4 for Ge. The estimated value of D_{ds} by Klemens (Ref. 6) formula is given in the brackets.

Sample B Dislocation density $=2.8\times10^{8}$ cm^{-2}	Sample C Dislocation density $=2.2\times10^{9}$ cm^{-2}
D_{dd} (dynamic) $= 5.8\times10^{6}$ sec^{-1} K	D_{dd} (dynamic) $= 2.5\times10^{7}$ sec^{-1} K
D_{ds} (static)$=1.0\times10^{4}$ sec^{-1} K $(1.55\times10^{4}$ sec^{-1} K) (Ref. 6)	D_{ds} (static) $=9.9\times10^{4}$ sec^{-1} K $(1.22\times10^{5}$ sec^{-1} K) (Ref. 6)
D_{sf} (stacking faults) $=6.2\times10^{3}$ sec^{-1} K^{-2}	D_{sf} (stacking faults) $=1.6\times10^{4}$ sec^{-1} K^{-2}
D_{dc} (core)$=3.4\times10^{2}$ sec^{-1} K^{-3}	D_{dc} (core) $=1.02\times10^{3}$ sec^{-1} K^{-3}

The value of parameters used by us in Eq. (1) are given in Tables I and II. Figures 1–4 show that the parameters used give an excellent agreement between the theoretical and experimental values[1,2] of phonon conductivity of plastically deformed LiF as well as Ge up to $T=50$ K and $T=30$ K, respectively. In Table III the values of phonon-point defect scattering parameter $A_1(=Av_s^3)$ in relaxation rate

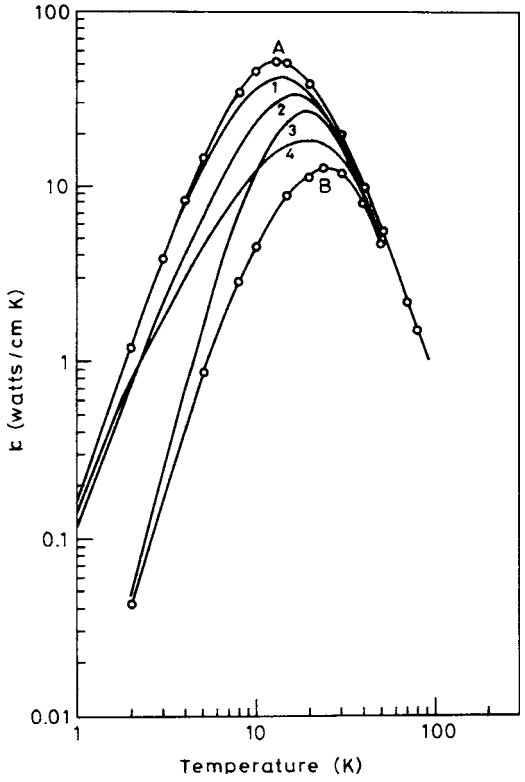

FIG. 1. Solid lines show the calculated value of κ and symbol (o) are the experimental points (Ref. 1) for LiF. A is the undeformed and B is the deformed sample. Numbers 1, 2, 3, and 4 represent, respectively, the calculated value of κ when only dislocation core, static dislocation, dynamic dislocation and stacking faults are present individually.

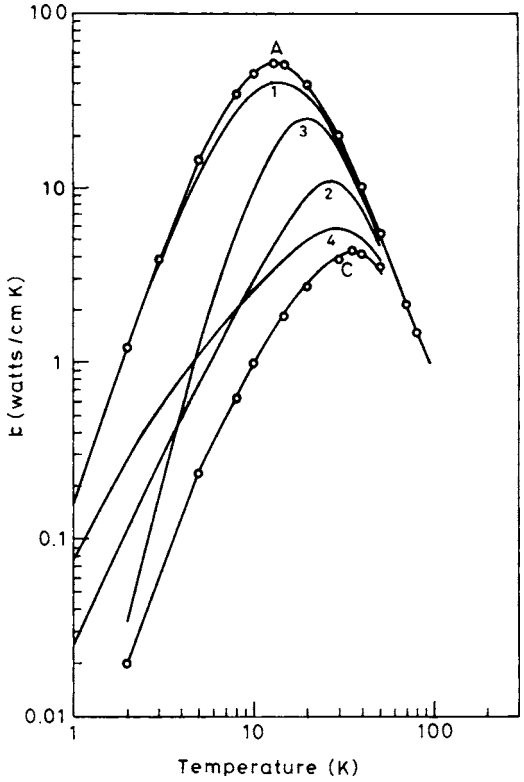

FIG. 2. Solid lines show the calculated value of κ and symbol (o) are the experimental points (Ref. 1) for LiF. A is the undeformed and C is the deformed sample. Numbers 1, 2, 3, and 4 represent, respectively, the calculated value of κ when only dislocation core, static dislocation, dynamic dislocation, and stacking faults are present individually.

$\tau_{pt}^{-1}(=Ax^4T^4)$ (Ref. 19) used in the present work for the samples of LiF (^{7}Li%95.4) and Ge are compared with its values in Refs. 2 and 20. Its value for LiF is nearly twice the value used by Berman and Brock.[20] This difference is chiefly due to the reason that in our work we have disregarded the presumption of the existence of large dislocation density in LiF samples as done in Ref. 20. In the case of Ge our value of A_1 is slightly lower than the value used by Sato and Sumino.[2] This small difference is caused due to the inclusion of phonon dispersion effects on A_1 which has been discussed in our earlier work[19] in detail.

Since any reliable assessment of D_{dd}, D_{ds}, D_{sf}, or D_{dc} may be made only if phonon-point scattering strength is known we ignore the analysis of the phonon conductivity of annealed samples in Ref. 1 in the present work. In annealed samples the number of point defects are expected to be different from the undeformed samples. Thus while in the case of unannealed deformed samples phonon-point defect scattering strength may be chosen to be the same as in undeformed samples and therefore assumed to be known, the same cannot be applied to the case of annealed samples. Table II shows that our adjusted value of D_{ds} for Ge is very close to its theoretically estimated value by Klemens[6] when the dislocations are randomly inclined. This result is very encouraging because together with this the value of the dislocation-core size obtained from the ω^3 dependent

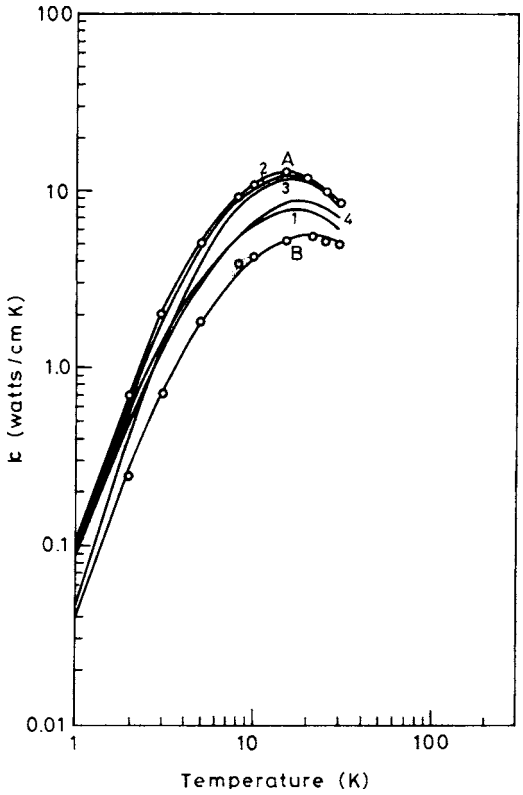

FIG. 3. Solid lines show the calculated value of κ and symbol (o) are the experimental points (Ref. 2) for Ge. A is the undeformed and B is the deformed sample. Numbers 1, 2, 3, and 4 represent, respectively, the calculated value of κ when only dislocation core, static dislocation, dynamic dislocation, and stacking faults are present individually.

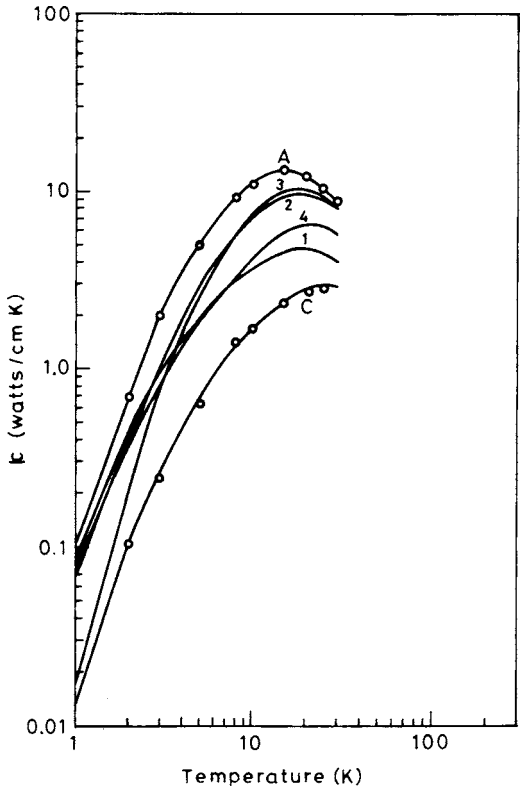

FIG. 4. Solid lines show the calculated value of κ and symbol (o) are the experimental points (Ref. 2) for Ge. A is the undeformed and C is the deformed sample. Numbers 1, 2, 3, and 4 represent, respectively, the calculated value of κ when only dislocation core, static dislocation, dynamic dislocation, and stacking faults are present individually.

term in τ_{dc}^{-1} is also very near the Burger vector in Ge (see Table IV). Moreover, if the ω^2 dependent term is associated with the phonon scattering by stacking faults, the value of $N_s(\sim 10/\text{cm})$ evaluated from the adjusted value of D_{sf} [Eq. (2)] also seems to be well within the reasonable limits.

In the case of LiF, however, while the values of the core size and the number of stacking faults (see Table IV) are still within the acceptable limits, the value of D_{ds} is almost an order higher than its theoretical value obtained from Klemens'[6] formula (see Table I). The values of D_{dd} are still expected to be nearly two times larger than their theoretical estimate[1,4,11] though they should be better than required in Refs. 1 and 4 due to the inclusion of other scattering processes in our work. There seem to be two possibilities for the discrepancies in D_{dd} as well as D_{ds}. Either (i) the number of pinned dislocations in the interior of deformed LiF is an order larger than their direct observations in Refs. 1 and 4 or (ii) the phonon scattering by grain boundaries is also effective.

According to Klemens, preposition (ii) would give rise to ω independent terms in τ_F^{-1} if the dislocations are sessile but yield a ω^2 dependent term if either (a) dislocations form an array or (b) other kinds of impurities move into the dislocation lines which is true only for the annealed sample in the present context. Roth and Anderson[21] as well as Klemens,[7] however, show that the ω^2 dependent term due to process (a) is negligible compared to the ω independent terms unless ω is of the order of Debye frequency. Consequently that the process (b) would be negligible in front of (a) at low temperatures ($\approx$2 K) is insignificant as compared to phonon-point defect scattering at higher temperatures ($\geq$5 K). Roth and Anderson[21] further show that although process (a) (ω independent) is compatible with experimental data of alkali halides qualitatively but its magnitude is too small to give any confirmed evidence for the contribution of this kind of phonon scattering towards κ. Hence we prefer to ignore phonon scattering by grain boundaries in our calculations.

The preposition (i) mentioned earlier seems to be more reasonable because due to jogging of line dislocations the

TABLE III. Calculated values of point defect parameters $A_1(=Av_s^3)$ written in $x(=\hbar\omega/kT)$ space for samples LiF (^{7}Li% 95.4) and normal Ge and their comparative values with Refs. 20 and 2, respectively. Here v_s is the average phonon velocity, L_c is the Casimir length.

	A_1 (calculated)	A_1 of Refs. 2 and 20	L_c
LiF	5.17×10^{16} cm^3 sec^{-4}K^{-4}	2.51×10^{16} cm^3 sec^{-4} K^{-4} (Ref. 20)	0.96 cm
Ge	4.75×10^{17} cm^3 sec^{-4} K^{-4}	5.22×10^{17} cm^3 sec^{-4} K^{-4} (Ref. 2)	0.365 cm

TABLE IV. Values of obtained number of stacking faults/cm N_s, core size, Burger vector for different samples of LiF and Ge.

	Samples (dislocation density in cm^{-2})	N_s in cm^{-1}	Core size in Å	Average core size	Burger vector $\sqrt{2}a$ (lattice parameter)
LiF	B(1.3×10^8)	8.85×10^1	5.70	5.13 Å	5.69 Å
	C(3.5×10^8)	3.16×10^2	4.56		
Ge	B(2.8×10^8)	3.12×10^1	9.52	8.50 Å	8.0 Å
	C(2.2×10^9)	8.0×10^1	7.48		

density of pinned dislocations in the interior of the crystal may be much larger than one expected. Moreover, with increasing stress, their density should increase at a faster rate but that of the unpinned dislocations at a slower rate than the number of dislocations directly observed through the surface. From Tables I and II it can be judged that in the present work the estimated changes in D_{dd} and D_{ds} with stress are consistent with this behavior. Incidentally, D_{dd} should be strongly governed by the number of unpinned and D_{ds} by pinned dislocations.[3]

Since in Refs. 1 and 2 there are no experimental data of number of stacking faults in their samples, we are not in a position to give any experimental evidence for the estimated number of stacking faults in the present theoretical analysis reported in Table IV. But this seems reasonable for the given dislocation densities. The obtained smaller value of N_s in Ge as compared to LiF for almost the same dislocation density is also according to the expectations because of the smaller number of partial dislocations in Ge due to its less close-packed structure (lower coordination number) than LiF. As can be seen from Figs. 1 and 2 for LiF that the depression of conductivity is maximum due to the stacking fault and is minimum due to the dislocation core near the peak. But Figs. 3 and 4 for Ge show that the depression due to scattering by dislocation cores dominates over stacking faults near the peak. This might be due to the larger value of the Burger vector in Ge than LiF.

From the above discussion it may be concluded that the ω^2 dependent term needed to explain the phonon conductivity data of plastically deformed LiF and Ge should be associated with phonon scattering by stacking faults and not by assumed point-defect vacancy clusters[2] or grain boundaries[21,22] or interference effects between various scattering processes.[15,22] It is also concluded that scattering by dislocation cores should not be altogether ignored as it does affect the heat flux due to high-frequency phonons, and hence helps explain the phonon conductivity depression at its peak.

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APPENDIX: OUR TREATMENT OF THERMAL CONDUCTIVITY INTEGRAL κ (REF. 19)

For convenience we reproduce here briefly the algebraic formula used in our numerical work. Consider phonons of wave vector $\mathbf{q}$, polarization $\lambda(\lambda=L$ for longitudinal, $\lambda=T$ for transverse), angular frequency $\omega_{q\lambda}$, group velocity $v_{g\lambda}$, and phase velocity $v_{p\lambda}$. Our aim is to account for the effect of dispersion in point defect relaxation rate $\tau_{pt}^{-1}(\omega_{q\lambda})$, boundary relaxation rate τ_b^{-1}, and also in total phonon conductivity integral κ ($\kappa=\kappa_D+\kappa_c$, κ_D being the Debye term and κ_c the correction term). For this purpose we divide κ into smaller domains labeled as (nm) in which the phonon group velocities are nearly constants. Then we can write

$$\kappa = \sum_{\lambda}\sum_{n}[\kappa_{D\lambda}(nm) + \kappa_{c\lambda}(nm)], \tag{A1}$$

$$\kappa_{D\lambda}(nm) = GT^3 b_\lambda \int_{x_n}^{x_m} v_{g\lambda}(x) v_{p\lambda}^{-2}(x)\tau_{F\lambda}(x,nm)J(x)dx, \tag{A2}$$

$$\kappa_{c\lambda}(nm) = GT^3 b_\lambda \beta' \int_{x_n}^{x_m} v_{p\lambda}^{-3}(x)\tau_{N\lambda}^{-1}(x,nm)\tau_{F\lambda}(x,nm)J(x)dx. \tag{A3}$$

Here b_λ is the weight factor for different polarizations λ, β' is the correction factor, and

$$m = n+1,\ G = k^4/2\pi^2\hbar^3,\ x = \hbar\omega/kT,\ J(x) = x^4e^x/(e^x-1)^2. \tag{A4}$$

The full relaxation rate $\tau_{F\lambda}^{-1}$ is given by

$$\tau_{F\lambda}^{-1} = \tau_{b\lambda}^{-1} + \tau_{pt\lambda}^{-1} + \tau_{N\lambda}^{-1} + \tau_{U\lambda}^{-1} + \tau_{def}^{-1}, \tag{A5}$$

where the successive terms on the right-hand side are the contributions due to boundary, point defect, normal processes, unklapp processes, and plastic deformation, respectively. In order to explain the experimental data for undeformed crystals (sample A of Figs. 1–4 for LiF and Ge) we set $\tau_{def}^{-1}=0$. For full details see Ref. 19.

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