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Author(s)	Tamura, S.; Sangu, A.; Maris, H. J.
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Anharmonic scattering of longitudinal acoustic phonons: Herring processes in tetragonal TeO₂ crystals

S. Tamura,¹ A. Sangu,¹ and H. J. Maris²

¹*Department of Applied Physics, Hokkaido University, Sapporo 060-8628, Japan*

²*Department of Physics, Brown University, Providence, Rhode Island 02912, USA*

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We calculate the anharmonic scattering rates of low-frequency longitudinal (L) acoustic phonons with thermally excited phonons via the Herring process $L+ST\rightarrow FT$ (with ST and FT the slow-transverse and fast-transverse phonons, respectively) in a tetragonal TeO₂ crystal. For temperatures well below the Debye temperature, we find a scattering rate $\Gamma=A\omega^2T^3$ as predicted by Herring. An analytical expression is derived for the variation of the coefficient with propagation direction. A typical value of A is of the order of 10^{-22} s K⁻³. For propagation along the fourfold axis, i.e., the [001] direction, however, $A=0$, indicating that in this direction L phonons are stable against the Herring processes. This is in contradiction to the recent experimental result reported by Damen *et al.* [Phys. Rev. B **59**, 349 (1999)]. We show that the vanishing of the scattering rate in the [001] direction can be understood in terms of the physics of the Herring process.

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I. INTRODUCTION

The study of the propagation and scattering of acoustic phonons in crystalline solids is of both fundamental and practical interest. In an important paper, Herring investigated the collision rate of low-frequency longitudinal (L) phonons satisfying $\omega \ll k_B T / \hbar$ (with T the temperature), and argued that these phonons play a special role in determining the heat conduction in crystals.¹ He showed that the interaction rate for these L phonons is dominated by collisions of the type $L+ST\rightarrow FT$, where ST and FT denote the slow-transverse and fast-transverse thermal phonons, respectively.² These processes occur for ST and FT phonons with wave vectors close to the degeneracy points of the constant-frequency surfaces (or slowness surfaces) in the Brillouin zone. Herring's analysis is limited to cubic anharmonicity and to frequencies of L phonons much less than the frequency of the thermally excited ST or FT phonons. Detailed studies of the scattering of L phonons in anisotropic solids are quite limited both experimentally and theoretically, though the expression for the scattering rate in cubic crystals has been given by Simons.³

Recently, with a new technique for the generation of monochromatic Fresnel-diffracted phonon beams in the GHz range,⁴ Damen *et al.* measured the attenuation rate of L phonons propagating along the [001] axis as a function of frequency and temperature in a single crystal of TeO₂ with tetragonal symmetry.⁵ The measured attenuation rate Γ varied according to $\tilde{A}\omega^a T^b$ with $a=1.84\pm 0.2$ and $b=2.81\pm 0.2$ below about 50 K, consistent with Herring's theory which gives $\Gamma \propto \omega^2 T^3$ for tetragonal crystals with the D_4 symmetry. The coefficient \tilde{A} was determined to have the value $\tilde{A}=(4.0\pm 0.5)\times 10^{-18}$ s^{a-1} K^{-b}.

The purpose of the present work is to derive an analytical expression for the scattering rate of the L phonons due to the Herring processes $L+ST\rightarrow FT$, and to see whether or not the above experimental results, including the magnitude of the coefficient \tilde{A} , are reproduced.

II. FORMULATION

For temperatures much less than the Debye temperature, phonon interactions can be considered using non-linear elasticity theory. To third order in the elastic strain η_{ij} , the elastic energy W stored in a strained crystal of initial volume V is

$$W = V \left(\frac{1}{2} c_{ijkl} \eta_{ij} \eta_{kl} + \frac{1}{6} c_{ijklmn} \eta_{ij} \eta_{kl} \eta_{mn} \right), \quad (1)$$

where

$$\eta_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} + \frac{\partial u_k}{\partial x_i} \frac{\partial u_k}{\partial x_j} \right) \quad (i, j = 1, 2, 3), \quad (2)$$

$\mathbf{u}(\mathbf{r}, t)$ is the displacement at position $\mathbf{r}=(x, y, z) = (x_1, x_2, x_3)$ at time t , and the summation convention over repeated indices is implied. For a TeO₂ crystal (belonging to the tetragonal crystal of class D_4 , or the point group 422) the fourth- and sixth-order elastic stiffness tensors c_{ijkl} and c_{ijklmn} have six and 12 independent components, respectively.

In the harmonic approximation, the equations governing elastic wave propagation in crystalline solids are

$$\rho \ddot{u}_i = c_{ijkl} \frac{\partial^2 u_k}{\partial x_j \partial x_l} \quad (i = 1, 2, 3), \quad (3)$$

where ρ is the mass density of the medium. By putting

$$\mathbf{u} = a \mathbf{e} \exp\{i(\mathbf{q} \cdot \mathbf{r} - \omega t)\}, \quad (4)$$

with a an amplitude, \mathbf{e} a unit polarization vector, $\mathbf{q} = (q_x, q_y, q_z) = (q_1, q_2, q_3)$ a wave vector and ω the angular frequency, Eq. (3) is converted to an eigenvalue equation

$$(\tilde{M}_{ik} - \rho \omega^2 \delta_{ik}) e_k = (c_{ijkl} q_j q_l - \rho \omega^2 \delta_{ik}) e_k = 0 \quad (i = 1, 2, 3). \quad (5)$$

The explicit expression for the matrix (\tilde{M}_{ik}) for TeO₂ is⁶

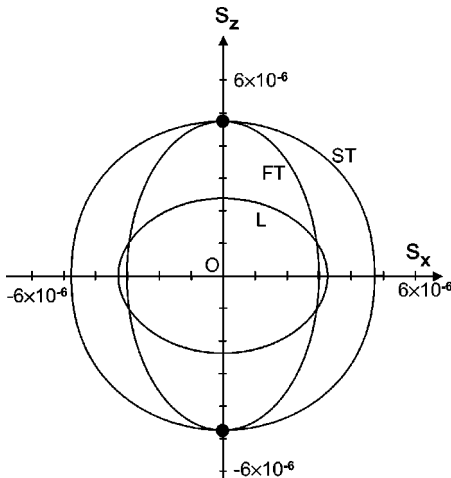
$$\tilde{M} = \begin{pmatrix} c_{11}q_x^2 + c_{66}q_y^2 + c_{44}q_z^2 & \tilde{c}_{12}q_xq_y & \tilde{c}_{13}q_zq_x \\ \tilde{c}_{12}q_xq_y & c_{66}q_x^2 + c_{11}q_y^2 + c_{44}q_z^2 & \tilde{c}_{13}q_yq_z \\ \tilde{c}_{13}q_zq_x & \tilde{c}_{13}q_yq_z & c_{44}(q_x^2 + q_y^2) + c_{33}q_z^2 \end{pmatrix}, \quad (6)$$

with c_{IJ} the Voigt elastic constants with two indices, $\tilde{c}_{12} = c_{12} + c_{66}$ and $\tilde{c}_{13} = c_{13} + c_{44}$. From the eigenvalues $\rho\omega_J^2$, ($J=1,2,3$), of the matrix \tilde{M} we obtain, for a given wave vector \mathbf{q} , the phase velocities $v_J = \omega_J/q$ (with $q = |\mathbf{q}|$) of three bulk modes, i.e., slow transverse (ST, $J=1$), fast transverse (FT, $J=2$), and longitudinal (L, $J=3$).

The Herring processes occur for thermally excited ST and FT phonons with wave vectors close to the directions in which the velocities of these modes are equal. At these points of degeneracy, the two slowness surfaces either touch or intersect. According to Herring's analysis, the degenerate points at which two slowness surfaces *touch* each other (rather than intersect) play the dominant role in determining the scattering rate of L phonons. Thus, in studying the Herring processes in a tetragonal crystal TeO₂, it is important to note that two transverse sheets touch only along the [001] and [00 $\bar{1}$] directions.⁶ The slowness surfaces of TeO₂ are shown in Fig. 1.

For \mathbf{q} parallel to [001] or [00 $\bar{1}$] the velocity of ST and FT phonons is $\rho v_T^2 = c_{44}$. At a polar angle θ slightly rotated away from the [001] (or [00 $\bar{1}$]) direction the degeneracy in the velocity of ST and FT is removed, and

$$\rho v_{ST}^2 = c_{44} + x_- \theta^2 = c_{44}(1 + 2\alpha\theta^2), \quad (7)$$



SLOWNESS (cm⁻¹s)

FIG. 1. Sections of the slowness surfaces of the slow transverse (ST), fast transverse (FT) and longitudinal (L) phonons in TeO₂ in the s_x - s_z plane [the slowness vector \mathbf{s} is defined by $\mathbf{s} = \hat{\mathbf{q}}/v(\mathbf{q})$]. Solid circles are the points where the ST and FT sheets touch each other.

$$\rho v_{FT}^2 = c_{44} + x_+ \theta^2 = c_{44}(1 + 2\beta\theta^2), \quad (8)$$

where $\alpha = x_-/2c_{44}$ and $\beta = x_+/2c_{44}$ with

$$x_{\pm} = \frac{1}{2} \left[\frac{\pm H^{1/2} - \tilde{c}_{13}^2}{c_{33} - c_{44}} + c_{11} + c_{66} - 2c_{44} \right]. \quad (9)$$

In Eq. (9) H is

$$H = H(\phi) = (\tilde{c}_{33}\tilde{c}_{11} - \tilde{c}_{13}^2)^2 \cos^2 2\phi + (\tilde{c}_{33}\tilde{c}_{12} - \tilde{c}_{13}^2)^2 \sin^2 2\phi, \quad (10)$$

where $\tilde{c}_{33} = c_{33} - c_{44}$ and $\tilde{c}_{11} = c_{11} - c_{66}$, and ϕ is the azimuthal angle of \mathbf{q} .

The expression for the scattering rate Γ_{λ} , or the reciprocal of the relaxation time τ_{λ} of a phonon specified by $\lambda = (\mathbf{q}, J)$ via three-phonon processes, is given by⁷

$$\Gamma_{\lambda} = \tau_{\lambda}^{-1} = \frac{\pi\hbar}{8\rho^3 V \omega_{\lambda} \omega_{\lambda'} \omega_{\lambda''}} \sum_{\lambda', \lambda''} \frac{|\Phi_{\lambda\lambda'\lambda''}|^2}{\omega_{\lambda'} \omega_{\lambda''}} \times 2(n_{\lambda'} - n_{\lambda''}) \delta_{\mathbf{q}+\mathbf{q}', \mathbf{q}''} \delta(\omega_{\lambda} + \omega_{\lambda'} - \omega_{\lambda''}), \quad (11)$$

where V is the system volume and $n_{\lambda} = n(\omega_{\lambda}) = [\exp(\hbar\omega_{\lambda}/k_B T) - 1]^{-1}$ is the occupation number of phonons at temperature T . The explicit expression for the three-phonon matrix element $\Phi_{\lambda\lambda'\lambda''}$ is lengthy and is given in the Appendix. Equation (11) is applied to the scattering of L phonons via Herring processes with $J=L$, $J'=ST$, and $J''=FT$. Thus, we write $\omega_{\lambda} = \omega_{\mathbf{q},L} = \omega$, $\omega_{\lambda'} = \omega_{\mathbf{q}',ST} = \omega'$ and $\omega_{\lambda''} = \omega_{\mathbf{q}'',FT} = \omega''$. The wave vectors of the initial L phonon and the ST phonon are expressed in polar coordinates as $\mathbf{q} = (q, \theta, \phi)$, and $\mathbf{q}' = (q', \theta', \phi')$. We then set

$$|\Phi_{\lambda\lambda'\lambda''}|^2 = (qq'q'')^2 |\tilde{\Phi}_{L+ST \rightarrow FT}(\theta', \phi')|^2, \quad (12)$$

where $\tilde{\Phi}_{L+ST \rightarrow FT}(\theta', \phi')$ is a function that does not depend on the magnitude of the wave vectors. We then have

$$\Gamma_L = \frac{\pi\hbar q^2}{4(2\pi)^3 \rho^3 \omega} \int d\phi' \int q'^2 dq' \times \int \sin\theta' d\theta' \frac{|\tilde{\Phi}_{L+ST \rightarrow FT}(\theta', \phi')|^2}{\omega' \omega''} \times (q'q'')^2 (n' - n'') \delta(\omega + \omega' - \omega''), \quad (13)$$

where $n' = n_{\lambda'}$ and $n'' = n_{\lambda''}$. We further write

$$\delta(\omega'' - \omega' - \omega) = \delta(v_{FT}q'' - v_{ST}q' - v_Lq) = \frac{1}{v_{FT}} \delta[g(\theta')], \quad (14)$$

where v_L is the sound velocity of the L mode⁸ depending on the direction of \mathbf{q} , and

$$g(\theta') = q'' - \gamma_{SF}q' - \gamma_{LF}q, \quad (15)$$

with $\gamma_{SF} = v_{ST}/v_{FT}$ and $\gamma_{LF} = v_L/v_{FT}$. Since we are considering $\omega \ll k_B T/\hbar$, the relation $q \ll q' \approx q''$ holds. Also

$$\gamma_{SF} = v_{ST}/v_{FT} \approx 1 + (\alpha - \beta)\theta'^2, \quad (16)$$

$$\gamma_{LF} = v_L/v_{FT} \approx \gamma(1 - \beta\theta'^2), \quad (17)$$

with $\gamma = v_L/v_T$. Now, we see that $g(\theta') = 0$ has two solutions $\theta' = \theta_n$, ($n=1,2$), with

$$\theta_1 = \left(\frac{\cos\theta + \gamma}{\beta - \alpha} \cdot \frac{q}{q'} \right)^{1/2} \quad \text{and} \quad \theta_2 = \pi - \left(\frac{-\cos\theta + \gamma}{\beta - \alpha} \cdot \frac{q}{q'} \right)^{1/2} \quad (18)$$

and

$$\beta - \alpha = \frac{x_+ - x_-}{2c_{44}} = \frac{H^{1/2}(\phi')}{2c_{44}\tilde{c}_{33}}. \quad (19)$$

Thus, we note that, for a smooth function f of θ' ,

$$\begin{aligned} & \int d\theta' \sin\theta' f(\theta') \delta[g(\theta')] \\ &= \sum_{n=1}^2 \sin\theta_n f(\theta_n) \left[\frac{dg(\theta)}{d\theta} \Big|_{\theta_n} \right]^{-1} \\ &= \frac{1}{2(\beta - \alpha)q'} \sum_{n=1}^2 f(\theta_n). \end{aligned} \quad (20)$$

Again using the condition $\omega \ll k_B T/\hbar$, we can approximate

$$n' - n'' = n(\omega') - n(\omega' + \omega) = -\frac{\partial n(\omega')}{\partial \omega'} \omega, \quad (21)$$

and the scattering rate becomes

$$\begin{aligned} \Gamma_L &= -\frac{\pi\hbar\omega^2}{4(2\pi)^3\rho^3v_L^2v_T^7} \tilde{c}_{33}c_{44} \int d\phi' H^{-1/2}(\phi') \\ &\times \sum_{n=1}^2 |\tilde{\Phi}_{L+ST \rightarrow FT}(\theta_n, \phi')|^2 \int d\omega' \omega'^3 \frac{\partial n(\omega')}{\partial \omega'}. \end{aligned} \quad (22)$$

The integral over ω' can be extended from 0 to infinity and then

$$-\int_0^\infty d\omega' \omega'^3 \frac{\partial n(\omega')}{\partial \omega'} = 6\zeta(3)(k_B T/\hbar)^3, \quad (23)$$

where $\zeta(n)$ is Riemann's zeta function.

We are left to calculate the integral in Eq. (22) with respect to ϕ' . In the calculation of the Herring processes, we

have $\mathbf{q}' \approx \mathbf{q}'' \parallel [001]$. The associated polarization vectors \mathbf{e}' and \mathbf{e}'' of the ST and FT phonons lie in the (001) plane. Thus, we can make the approximation that $\mathbf{e}' \cdot \mathbf{e}'' = \mathbf{q}' \cdot \mathbf{e}' = \mathbf{q}'' \cdot \mathbf{e}'' = \mathbf{q}'' \cdot \mathbf{e}' = 0$. Next we write the wave vector \mathbf{q} of the L phonon as

$$\mathbf{q} = q(l, m, n), \quad (24)$$

where (l, m, n) are the direction cosines of \mathbf{q} . The polarization vector \mathbf{e} of the initial L phonon is, in general, not collinear with \mathbf{q} except for high symmetry directions due to the presence of the anisotropy. However, $\mathbf{q} \cdot \mathbf{e}/q$ is close to unity ($\mathbf{q} \cdot \mathbf{e}/q \geq 0.9713$ for TeO₂) at least when \mathbf{q} is oriented in the (100) and (110) planes. Consequently, as an approximation, we take \mathbf{e} to be parallel to \mathbf{q} and write

$$\mathbf{e} = (l, m, n). \quad (25)$$

This approximation becomes exact in the [100], [001], [110] and equivalent directions.⁹ Also we put the polarization vectors of the ST and FT phonons as

$$\begin{aligned} \mathbf{e}' &= \mathbf{e}_{ST} = (\cos\phi', \sin\phi', 0), \\ \mathbf{e}'' &= \mathbf{e}_{FT} = (\sin\phi', -\cos\phi', 0). \end{aligned} \quad (26)$$

Thus with Eq. (A1) we have

$$\begin{aligned} |\tilde{\Phi}_{L+ST \rightarrow FT}|^2 &= \frac{1}{4(1 + \delta^2)} [(c_{155} - c_{144} + 2c_{44})(l^2 - m^2) \\ &\quad - 4\delta(c_{456} + c_{44})lm]^2, \end{aligned} \quad (27)$$

where

$$\delta = \frac{\tilde{c}_{13}^2 - \tilde{c}_{11}\tilde{c}_{33}}{\tilde{c}_{12}\tilde{c}_{33} - \tilde{c}_{13}^2} \cot 2\phi'. \quad (28)$$

Executing the integral over ϕ' in Eq. (22), we finally obtain

$$\begin{aligned} \Gamma_L &= \tau_L^{-1} = \frac{3\zeta(3)k_B^3}{8\pi^2\hbar^2\rho^3v_L^2v_T^7} \omega^2 T^3 \\ &\times \frac{c_{44}[\tilde{c}_{11}\tilde{c}_{33} - \tilde{c}_{13}^2]}{(\tilde{c}_{11} - \tilde{c}_{12})[\tilde{c}_{33}(\tilde{c}_{11} + \tilde{c}_{12}) - 2\tilde{c}_{13}^2]} \times \{ (c_{155} \\ &\quad - c_{144} + 2c_{44})^2(l^2 - m^2)^2 [E(h) - (1 - h^2)K(h)] \\ &\quad + 16(c_{456} + c_{44})^2 l^2 m^2 [K(h) - E(h)] \}, \end{aligned} \quad (29)$$

where

$$h^2 = 1 - \frac{(\tilde{c}_{33}\tilde{c}_{12} - \tilde{c}_{13}^2)^2}{(\tilde{c}_{33}\tilde{c}_{11} - \tilde{c}_{13}^2)^2} = -\tilde{h}^2, \quad (30)$$

and $E(h)$ and $K(h)$ are the integrals defined by

$$K(h) \equiv \int_0^{\pi/2} \frac{d\phi}{[1 - h^2 \sin^2 \phi]^{1/2}} = \int_0^{\pi/2} \frac{d\phi}{[1 + \tilde{h}^2 \sin^2 \phi]^{1/2}}, \quad (31)$$

$$E(h) \equiv \int_0^{\pi/2} [1 - h^2 \sin^2 \phi]^{1/2} d\phi = \int_0^{\pi/2} [1 + \tilde{h}^2 \sin^2 \phi]^{1/2} d\phi. \quad (32)$$

Here we note that for TeO_2 , $h^2 = -\tilde{h}^2 < 0$ because $c_{66} > c_{11}$. However, this is an exceptional case. For the majority of tetragonal crystals $c_{66} < c_{11}$, and $h^2 > 0$ and the integral equations (31) and (32) become complete elliptic integrals of the first and second kinds, respectively.

It is interesting to compare formula (29) with the corresponding expression for cubic crystals derived by Simons^{3,7} which can be written in the form

$$\begin{aligned} \Gamma_L^{(\text{cubic})} &= \frac{3\zeta(3)k_B^3}{8\pi^2\tilde{h}^2\rho^3v_L^2v_T^7} \omega^2 T^3 \\ &\times \frac{(c_{11} + c_{12})c_{44}}{(c_{11} + 2c_{12} + c_{44})|c_{11} - c_{12} - 2c_{44}|} \\ &\times \{(c_{155} - c_{144} + 2c_{44})^2[(l^2 - m^2)^2 \\ &+ (m^2 - n^2)^2 + (n^2 - l^2)^2][E(h) - (1 - h^2)K(h)] \\ &+ 16(c_{456} + c_{44})^2(l^2m^2 + m^2n^2 + n^2l^2)\} \\ &\times [K(h) - E(h)]. \end{aligned} \quad (33)$$

In this formula

$$h^2 = 1 - \frac{(c_{12} + c_{44})^2}{(c_{11} + c_{12})^2} \quad (34)$$

and in the derivation of Eq. (33), it has been assumed that h^2 is positive so that $K(h)$ and $E(h)$ are the complete elliptic integrals. Note that v_L in both Eqs. (29) and (33) depends on the propagation direction.

Here we note that Eq. (29) reduces to $\Gamma_L^{(\text{cubic})}$ [Eq. (33)] when cubic symmetry is applied to the elastic constants and $(m^2 - n^2)^2 + (n^2 - l^2)^2$ is added to $(l^2 - m^2)^2$ and also $m^2n^2 + n^2l^2$ to l^2m^2 . These added contributions come from scatterings with transverse phonons propagating close to the directions along the fourfold $[100]$, $[010]$, $[\bar{1}00]$, and $[0\bar{1}0]$ axes in cubic crystals. However, for tetragonal crystals the fourfold symmetry is absent except about the $[001]$ (and $[00\bar{1}]$) axis and the degeneracy of the transverse phonons along those four directions is lost. This is the reason that the transverse phonons propagating close to the $[100]$, $[010]$, $[\bar{1}00]$, and $[0\bar{1}0]$ directions do not contribute to the scattering of L phonons in tetragonal crystals. This point will be discussed again in Sec. IV.

Now the most important result we can find from Eq. (29) is the fact that the rate of scattering by the Herring process in tetragonal TeO_2 vanishes for longitudinal phonons propagating in the $[001]$ and $[00\bar{1}]$ directions. To be more precise, we see that the coefficient of the component of the scattering rate that varies as $\omega^2 T^3$ is zero.¹⁴ Thus in the experiment by Damen *et al.*⁵ the attenuation must arise from some effect other than Herring processes.

Incidentally, at temperatures higher than the Debye temperature, for which most phonon modes are thermally excited, the difference of phonon occupation numbers [Eq. (21)] is replaced with

$$n' - n'' = \frac{k_B T}{\hbar} \frac{\omega}{\omega' \omega''}, \quad (35)$$

and the expression for the attenuation rate becomes

$$\begin{aligned} \Gamma_L &= \frac{k_B^3 \Theta_D^2}{32\pi^2 \tilde{h}^2 \rho^3 v_L^2 v_T^7} \omega^2 T \times \frac{c_{44} |\tilde{c}_{11} \tilde{c}_{33} - \tilde{c}_{13}^2|}{(\tilde{c}_{11} - \tilde{c}_{12}) [\tilde{c}_{33} (\tilde{c}_{11} + \tilde{c}_{12}) - 2\tilde{c}_{13}^2]} \\ &\times \{(c_{155} - c_{144} + 2c_{44})^2 (l^2 - m^2)^2 \\ &\times [E(h) - (1 - h^2)K(h)] + 16(c_{456} + c_{44})^2 \\ &\times l^2 m^2 [K(h) - E(h)]\}, \end{aligned} \quad (36)$$

where we have put the upper limit of the integral of Eq. (13) with respect to q' as $q_{max} = k_B \Theta_D / \hbar v_T$. Thus the attenuation rate proportional to $\omega^2 T$ is obtained in agreement with Herring.¹ Of course, Eq. (36) is only an approximate result because it based on the use of elasticity theory.

III. NUMERICAL RESULTS

For the evaluation of the attenuation rate we used the second- and third-order elastic constants of TeO_2 measured at low temperatures, i.e., $c_{11} = 55.9$, $c_{33} = 105.5$, $c_{44} = 26.6$, $c_{66} = 66.3$, $c_{12} = 51.6$, $c_{13} = 23.9$, $c_{111} = -160$, $c_{112} = -600$, $c_{113} = -140$, $c_{123} = -110$, $c_{133} = +180$, $c_{144} = -41$, $c_{155} = +36$, $c_{166} = -640$, $c_{333} = -2110$, $c_{344} = -54$, $c_{366} = -260$, and $c_{456} = -250$ (in units of 10^{10} dyn cm^{-2}).¹⁰ The mass density is $\rho = 5.99$ g cm^{-3} and these values give bulk sound velocities in the $[001]$ direction $v_L = 4.20 \times 10^5$ cm s^{-1} and $v_T = 2.11 \times 10^5$ cm s^{-1} .

We can write Eq. (29) as

$$\begin{aligned} \Gamma_L &= [B(l^2 - m^2)^2 + 4C l^2 m^2] \omega^2 T^3 \\ &= (B \cos^2 2\phi + C \sin^2 2\phi) \sin^4 \theta \omega^2 T^3 \\ &= A(\theta, \phi) \omega^2 T^3. \end{aligned} \quad (37)$$

In this expression, the coefficients B and C depend on θ and ϕ only through the variation of v_L . The scattering is strongest for propagation at $\theta = \pi/2$, and there is a variation with azimuthal angle since B is not equal to C . From the elastic constants of TeO_2 , we find $h^2 = -3.11$ and the values of the integrals of Eqs. (31) and (32) are $E(h) = 2.446$ and $K(h) = 1.070$. The values of A for selected directions are given in Table I and in Fig. 2.

These magnitudes of A can be compared with the corresponding coefficient for cubic crystals given by

$$\begin{aligned} \Gamma_L^{(\text{cubic})} &= \{\tilde{B}[(l^2 - m^2)^2 + (m^2 - n^2)^2 + (n^2 - l^2)^2] \\ &+ 4\tilde{C}(l^2 m^2 + m^2 n^2 + n^2 l^2)\} \omega^2 T^3 \\ &= A(\theta, \phi) \omega^2 T^3, \end{aligned} \quad (38)$$

TABLE I. Calculated values of coefficient A for TeO_2 , and for the cubic crystals Si, GaAs and Ne in units of s K^{-3} .

I	$A(\theta=0, \phi=0)$	$A(\theta=\pi/2, \phi=0)$	$A(\theta=\pi/2, \phi=\pi/4)$
TeO_2	0	1.97×10^{-22}	7.69×10^{-22}
Si	1.29×10^{-24}	1.29×10^{-24}	2.78×10^{-25}
GaAs	7.67×10^{-24}	7.67×10^{-24}	1.64×10^{-24}
Ne	2.48×10^{-19}	2.48×10^{-19}	5.65×10^{-20}

where \tilde{B} and \tilde{C} are the coefficients corresponding to B and C . Values of A for silicon,¹¹ GaAs,¹² and Ne (Ref. 13) are included in Table I. As might be expected based on the relative magnitudes of the sound velocities in these materials, the scattering is largest in neon and least in silicon.

When the temperature is increased so that it is no longer much less than Θ_D , it is necessary to evaluate Eq. (13) numerically. We have taken the Debye temperature as 200 K,⁵ and set the upper limit of the integral over q' as $q_{max} = k_B \Theta_D / \hbar v_T$. The results for a frequency of a 5 GHz phonon propagating in the [110] direction are shown in Fig. 3. It can be seen that there is a crossover from $\omega^2 T^3$ to $\omega^2 T$ behavior at around 50 K.

IV. DISCUSSIONS

The Herring process is usually considered as a scattering process in which a low energy L phonon scatters from an ST thermal phonon to produce an FT phonon. But since the Herring process concerns the scattering of a low energy phonon, it should be possible to treat this phonon macroscopically, i.e., as a longitudinal sound wave. The scattering rate is then equivalent to the rate at which this sound wave is damped by interactions with thermal phonons.¹⁵

To discuss this qualitatively, consider what happens when through the presence of a sound wave, a small, time-dependent, elastic strain η_{ij} is applied to a crystal containing a distribution of thermal phonons. Because of anharmonicity, the application of the strain results in a small change in the effective elastic constants. This, in turn causes a change in the phonon frequencies and in the polarization vectors. If the

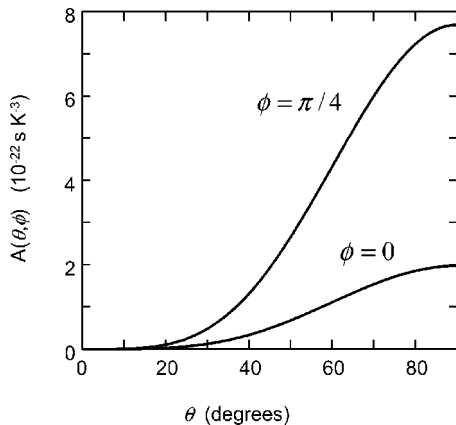


FIG. 2. Angular dependence of the coefficient A of the attenuation rate in the (100) ($\phi=0$) and (110) ($\phi=\pi/4$) planes of TeO_2 .

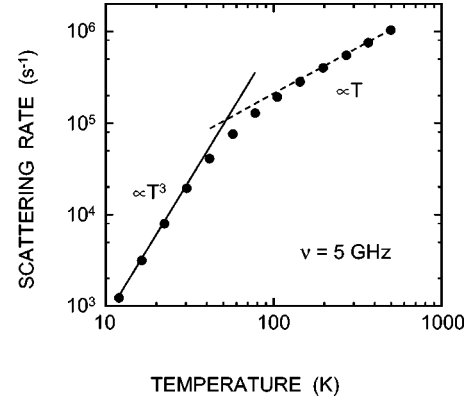


FIG. 3. Temperature dependence of the attenuation rate of L phonons of frequency 5 GHz propagating in the [110] direction. The solid circles are the results of numerical calculations based on Eq. (13). The solid line is the analytical result [Eq. (29)] for low temperatures and the dotted line is the high temperature formula [Eq. (36)].

strain varies very slowly with time, the phonons will adjust adiabatically, i.e., their frequencies and polarization vectors will be modified but if the strain is removed, the phonon distribution will return to its initial condition. However, if the strain changes rapidly, the phonons will not adjust adiabatically, irreversible effects will take place, and the phonon system will absorb energy. We can expect that these effects will be largest for those phonons whose frequency ω' or polarization vector \mathbf{e}' change rapidly when a strain is applied. Based on this qualitative picture we can understand the vanishing of the scattering rate for longitudinal phonons propagating in the [001] and $[00\bar{1}]$ directions.

For most of the thermal phonons, the fractional change in ω' or \mathbf{e}' is of the same order of magnitude as η_{ij} . However, for thermal phonons with wave vectors lying close to a direction in which there is a degeneracy there can be a much larger change in \mathbf{e}' . As a specific example, consider an ST thermal phonon in TeO_2 with wave vector $(\epsilon q, 0, q)$, where ϵ is very small. This phonon has \mathbf{e}' parallel to [100].⁶ Now suppose that the propagation direction of the longitudinal sound is along [100] so that there is an oscillating strain component η_{11} . Such a strain lowers the symmetry of the TeO_2 crystal from tetragonal to orthorhombic and so c_{44} is no longer equal to c_{55} . Thus the strain removes the degeneracy of the transverse phonons propagating in the [001] direction. During the part of the cycle of the sound wave in which the strain is such that $c_{55} > c_{44}$, the polarization of the ST phonon remains parallel to [100], but when the sign of the strain is such that $c_{55} < c_{44}$, the polarization changes to become parallel to [010]. The strain needed to accomplish this large change in \mathbf{e}' is of the same order of magnitude as ϵ . Thus if ϵ is small, the change in \mathbf{e}' is very much larger than the magnitude of the strain of the sound wave, i.e., there is an anomalously large variation of \mathbf{e}' with strain. Our idea is that the Herring processes arise from this effect.

If this is correct, there will be Herring processes provided that the strain of the L phonon (or the strain of the equivalent sound wave) lifts the degeneracy of some transverse

phonons. For a cubic crystal, the transverse phonons are degenerate along all directions of type $\langle 100 \rangle$, and so there will always be some Herring processes regardless of the direction of propagation of the L phonon. This is in agreement with the results of Simons³ as given in Eq. (33). However, in tetragonal TeO₂ the degeneracy occurs only along $[00\bar{1}]$ and $[00\bar{1}]$. A strain of type η_{33} does not lift this degeneracy because $c_{344} = c_{355}$ and so an L phonon propagating in these direction undergoes no Herring scattering.

We want to emphasize that the above ideas are only suggestive. It would be very interesting to see if it is possible to derive the expression for the rate of Herring processes using an elaboration of this approach.

Accepting this result, one can ask what is the origin of the attenuation seen by Damen *et al.*⁵ One possibility is that it is a relaxation process. When a low frequency phonon ω interacts with thermal phonons that have a lifetime τ such that $\omega\tau < 1$, the damping of the phonon varies as ω^2 which is the frequency-dependence observed by Damen *et al.* Under these conditions, the picture of the low energy phonon making momentum and energy-conserving collisions with individual thermal phonons no longer applies.⁷ However, for this to be the explanation of the observed results, it would be necessary for the thermal phonons to have lifetimes less than $(2\pi \times 5\text{GHz})^{-1} = 30$ ps. The thermal conductivity of TeO₂

has been measured by Ewbank and Newman¹⁶ down to 77 K. At this temperature, they found a conductivity of $0.09 \text{ W cm}^{-1} \text{ K}^{-1}$, and from this they estimated a thermal phonon lifetime of around 4 ps. Thus, at this temperature it is certainly true that $\omega\tau < 1$ and so the experiment should be analyzed in terms of a relaxation process. Unfortunately, we have been unable to find experimental data for the thermal conductivity at lower temperatures and so cannot make a reliable estimate of the thermal phonon lifetime.

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APPENDIX

In this appendix we give explicit expressions for the matrix elements $\Phi_{\lambda\lambda'\lambda''}$:

$$\begin{aligned} \Phi_{\lambda\lambda'\lambda''} = & c_{13}[(\mathbf{q} \cdot \mathbf{q}')(\mathbf{q}'' \cdot \mathbf{e}'')(\mathbf{e} \cdot \mathbf{e}') + (\mathbf{q}' \cdot \mathbf{q}'')(\mathbf{q} \cdot \mathbf{e})(\mathbf{e}' \cdot \mathbf{e}'') + (\mathbf{q}'' \cdot \mathbf{q})(\mathbf{q}' \cdot \mathbf{e}')(\mathbf{e}'' \cdot \mathbf{e})] + c_{44}\{(\mathbf{q} \cdot \mathbf{q}')[(\mathbf{q}'' \cdot \mathbf{e})(\mathbf{e}' \cdot \mathbf{e}'') + (\mathbf{q}'' \cdot \mathbf{e}') \\ & \times (\mathbf{e}'' \cdot \mathbf{e})] + (\mathbf{q}' \cdot \mathbf{q}'')[(\mathbf{q} \cdot \mathbf{e}')(\mathbf{e}'' \cdot \mathbf{e}) + (\mathbf{q} \cdot \mathbf{e}'')(\mathbf{e} \cdot \mathbf{e}')] + (\mathbf{q}'' \cdot \mathbf{q})[(\mathbf{q}' \cdot \mathbf{e}'')(\mathbf{e} \cdot \mathbf{e}') + (\mathbf{q}' \cdot \mathbf{e})(\mathbf{e}' \cdot \mathbf{e}'')]\} + (c_{11} - c_{13} \\ & - 2c_{44}) \sum_{i=1}^3 q_i q'_i q''_i [e_i(\mathbf{e}' \cdot \mathbf{e}'') + e'_i(\mathbf{e}'' \cdot \mathbf{e}) + e''_i(\mathbf{e} \cdot \mathbf{e}')] + (c_{33} - c_{11}) q_3 q'_3 q''_3 [e_3(\mathbf{e}' \cdot \mathbf{e}'') + e'_3(\mathbf{e}'' \cdot \mathbf{e}) + e''_3(\mathbf{e} \cdot \mathbf{e}')] \\ & + 2(c_{12} - c_{13}) [(q_1 q'_2 q''_2 e_1 + q_2 q'_1 q''_1 e_2)(\mathbf{e}' \cdot \mathbf{e}'') + (q'_1 q''_2 q_2 e'_1 + q'_2 q''_1 q_1 e'_2)(\mathbf{e}'' \cdot \mathbf{e}) + (q''_1 q_2 q'_2 e''_1 + q''_2 q_1 q'_1 e''_2)(\mathbf{e} \cdot \mathbf{e}')] \\ & + (c_{66} - c_{44}) [(q_1 q'_2 + q'_1 q_2)(q''_1 e''_2 + q''_2 e''_1)(\mathbf{e} \cdot \mathbf{e}') + (q'_1 q''_2 + q''_1 q'_2)(q_1 e_2 + q_2 e_1)(\mathbf{e}' \cdot \mathbf{e}'') + (q''_1 q_2 + q_1 q''_2)(q'_1 e'_2 \\ & + q'_2 e'_1)(\mathbf{e}'' \cdot \mathbf{e})] + (c_{112} - 2c_{144})(\mathbf{q} \cdot \mathbf{e})(\mathbf{q}' \cdot \mathbf{e}')(\mathbf{q}'' \cdot \mathbf{e}'') + c_{144}\{(\mathbf{q} \cdot \mathbf{e})[(\mathbf{q}' \cdot \mathbf{q}'')(\mathbf{e}' \cdot \mathbf{e}'') + (\mathbf{q}' \cdot \mathbf{e}'')(\mathbf{q}'' \cdot \mathbf{e}')] + (\mathbf{q}' \cdot \mathbf{e}') \\ & \times [(\mathbf{q}'' \cdot \mathbf{q})(\mathbf{e}'' \cdot \mathbf{e}) + (\mathbf{q}'' \cdot \mathbf{e})(\mathbf{q} \cdot \mathbf{e}'')] + (\mathbf{q}'' \cdot \mathbf{e}'')[(\mathbf{q} \cdot \mathbf{q}')(\mathbf{e} \cdot \mathbf{e}') + (\mathbf{q} \cdot \mathbf{e}')(\mathbf{q}' \cdot \mathbf{e})]\} + \frac{1}{2}(c_{166} - c_{144})\{(\mathbf{q} \cdot \mathbf{e}')(\mathbf{q}' \cdot \mathbf{e}'') \\ & \times (\mathbf{q}'' \cdot \mathbf{e}) + (\mathbf{q} \cdot \mathbf{e}'')(\mathbf{q}' \cdot \mathbf{e})(\mathbf{q}'' \cdot \mathbf{e}') + (\mathbf{q} \cdot \mathbf{q}')[(\mathbf{q}'' \cdot \mathbf{e})(\mathbf{e}' \cdot \mathbf{e}'') + (\mathbf{q}'' \cdot \mathbf{e}')(\mathbf{e}'' \cdot \mathbf{e})] + (\mathbf{q}' \cdot \mathbf{q}'')[(\mathbf{q} \cdot \mathbf{e}')(\mathbf{e}'' \cdot \mathbf{e}) + (\mathbf{q} \cdot \mathbf{e}'') \\ & \times (\mathbf{e} \cdot \mathbf{e}')] + (\mathbf{q}'' \cdot \mathbf{q})[(\mathbf{q}' \cdot \mathbf{e}'')(\mathbf{e} \cdot \mathbf{e}') + (\mathbf{q}' \cdot \mathbf{e})(\mathbf{e}' \cdot \mathbf{e}'')]\} + (c_{111} - c_{112} - 4c_{166}) \sum_{i=1}^3 q_i q'_i q''_i e_i e'_i e''_i + [c_{333} - c_{111} \\ & - 3(c_{113} + c_{133}) + 6c_{112}] q_3 q'_3 q''_3 e_3 e'_3 e''_3 + (2c_{144} - c_{112} + c_{123})(q_1 q'_2 q''_3 e_1 e'_2 e''_3 + q'_1 q''_2 q_3 e'_1 e''_2 e_3 + q''_1 q_2 q'_3 e''_1 e_2 e'_3 \\ & + q_1 q''_2 q'_3 e_1 e''_2 e'_3 + q'_1 q_2 q''_3 e'_1 e_2 e''_3 + q''_1 q'_2 q_3 e''_1 e'_2 e_3) + \frac{1}{2}(c_{144} - c_{166} + 2c_{456}) \times \{(q_1 e_2 + q_2 e_1)[(q'_2 e'_3 + q'_3 e'_2)(q''_3 e''_1 \\ & + q''_1 e''_3) + (q''_2 e''_3 + q''_3 e''_2)(q'_3 e'_1 + q'_1 e'_3)] + (q'_1 e'_2 + q'_2 e'_1)[(q''_2 e''_3 + q''_3 e''_2)(q_3 e_1 + q_1 e_3) + (q_2 e_3 + q_3 e_2)(q'_3 e''_1 \\ & + q'_1 e''_3)] + (q''_1 e''_2 + q''_2 e''_1)[(q_2 e_3 + q_3 e_2)(q'_3 e'_1 + q'_1 e'_3) + (q'_2 e'_3 + q'_3 e'_2)(q_3 e_1 + q_1 e_3)]\} + (c_{113} - c_{112}) \end{aligned}$$

$$\begin{aligned}
& \sum_{i=1}^3 [q_i q'_i q''_i e_i e'_i e''_i + q'_i q''_i q_3 e'_i e''_i e_3 + q''_i q_i q'_3 e_i e'_i e''_3] + (c_{133} - c_{112}) [q_3 q'_3 e_3 e'_3 (\mathbf{q} \cdot \mathbf{e}'') + q'_3 q''_3 e'_3 e''_3 (\mathbf{q} \cdot \mathbf{e}) + q''_3 q_3 e''_3 e_3 (\mathbf{q}' \cdot \mathbf{e}')] \\
& + (c_{155} - c_{166}) [q_1 e_1 (q'_1 e'_3 + q'_3 e'_1) (q''_1 e''_3 + q''_3 e''_1) + q'_1 e'_1 (q''_1 e''_3 + q''_3 e''_1) (q_1 e_3 + q_3 e_1) + q''_1 e''_1 (q_1 e_3 + q_3 e_1) (q'_1 e'_3 + q'_3 e'_1) \\
& + q_2 e_2 (q'_2 e'_3 + q'_3 e'_2) (q''_2 e''_3 + q''_3 e''_2) + q'_2 e'_2 (q''_2 e''_3 + q''_3 e''_2) (q_2 e_3 + q_3 e_2) + q''_2 e''_2 (q_2 e_3 + q_3 e_2) (q'_2 e'_3 + q'_3 e'_2) + q_3 e_3 (q'_1 e'_2 \\
& + q'_2 e'_1) (q''_1 e''_2 + q''_2 e''_1) + q'_3 e'_3 (q''_1 e''_2 + q''_2 e''_1) (q_1 e_2 + q_2 e_1) + q''_3 e''_3 (q_1 e_2 + q_2 e_1) (q'_1 e'_2 + q'_2 e'_1)] + (c_{344} - c_{166}) \\
& \times \{q_3 e_3 [(q'_1 e'_2 + q'_2 e'_1) (q''_1 e''_2 + q''_2 e''_1) + (q'_2 e'_3 + q'_3 e'_2) (q''_2 e''_3 + q''_3 e''_2) + (q'_3 e'_1 + q'_1 e'_3) (q''_3 e''_1 + q''_1 e''_3)] + q'_3 e'_3 [(q''_1 e''_2 \\
& + q''_2 e''_1) (q_1 e_2 + q_2 e_1) + (q''_2 e''_3 + q''_3 e''_2) (q_2 e_3 + q_3 e_2) + (q''_3 e''_1 + q''_1 e''_3) (q_3 e_1 + q_1 e_3)] + q''_3 e''_3 [(q_1 e_2 + q_2 e_1) (q'_1 e'_2 + q'_2 e'_1) \\
& + (q_2 e_3 + q_3 e_2) (q'_2 e'_3 + q'_3 e'_2) + (q_3 e_1 + q_1 e_3) (q'_3 e'_1 + q'_1 e'_3)]\} + (c_{366} - c_{344} + 2c_{166} - c_{155} - c_{144}) [q_3 e_3 (q'_1 e'_2 + q'_2 e'_1) \\
& \times (q''_1 e''_2 + q''_2 e''_1) + q'_3 e'_3 (q''_1 e''_2 + q''_2 e''_1) (q_1 e_2 + q_2 e_1) + q''_3 e''_3 (q_1 e_2 + q_2 e_1) (q'_1 e'_2 + q'_2 e'_1)]. \tag{A1}
\end{aligned}$$

¹C. Herring, Phys. Rev. **95**, 954 (1954).

²For other collision processes of phonons, see, for example, J. W. Tucker and V. W. Rampton, *Microwave Ultrasonics in Solid State Physics* (North-Holland, Amsterdam, 1972); H. J. Maris and S. Tamura, Phys. Rev. B **47**, 727 (1993); S. Tamura and H. J. Maris, *ibid.* **51**, 2857 (1995).

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⁷H. J. Maris, in *Physical Acoustics* (Academic, New York, 1971), Vol. 8, p. 279.

⁸The magnitude of v_L depends also on the direction of \mathbf{q} . For example, $v_L = (c_{33}/\rho)^{1/2} = 4.20 \times 10^5$ cm s⁻¹ in the [001] direction and $v_L = (c_{11}/\rho)^{1/2} = 3.06 \times 10^5$ cm s⁻¹ in the [100] direction.

⁹However, this approximation becomes poor for directions of \mathbf{q} near the $\langle 100 \rangle$ axis in the (001) plane ($\theta = \pi/2$ and $\phi \sim 0$), though $\mathbf{e} \parallel \mathbf{q}$ holds exactly in the $\langle 100 \rangle$ direction. See Ref. 6.

¹⁰A. G. Every and A. K. McCurdy in *Second and Higher Order*

Elastic Constants, edited by D. F. Nelson, Landolt-Börnstein, New Series, Group III, Vol. 29-a (Springer, Berlin, 1992).

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¹²J. R. Drabble and A. J. Brammer, Solid State Commun. **4**, 467 (1966).

¹³For Ne we employed the parameters $c_{11} = 1.61$, $c_{44} = 0.92$, and $c_{12} = 0.85$ (in units of 10^{10} dyn cm⁻²) and the mass density $\rho = 1.51$ g cm⁻³. [See J. Eckert, W. B. Daniels, and J. D. Axe, Phys. Rev. B **14**, 3649 (1976).] For the third-order elastic constants of Ne, we used a Grüneisen constant $\gamma = 2.7$ and assumed the relationships valid for a fcc model; $c_{123} = c_{144} = c_{456} = 0$ and $c_{111} = 2c_{122} = 2c_{166}$. This leads to $c_{166} = -7.92 \times 10^{10}$ dyn cm⁻². See R. A. Coldwell-Horsfall, Phys. Rev. **129**, 22 (1962).

¹⁴The scattering process L+ST→FT also happens for the wave vectors of both ST and FT phonons near the directions where their constant-frequency surfaces intersect each other. Such a process leads to the scattering rate proportional to $\omega^3 T^2$, which is much smaller than the dominant rate $A \omega^2 T^3$ for $\hbar \omega \ll k_B T$ unless $A = 0$.

¹⁵These two alternate ways of considering the scattering of low energy phonons have been discussed in Ref. 7. However, the discussion there does not include the Herring processes.

¹⁶M. D. Ewbank and P. R. Newman, Solid State Commun. **39**, 303 (1981).