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Ultrasonic Study of Three-Phonon Interactions. I. Theory*

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The nonlinear interaction of two ultrasonic waves in a homogeneous, isotropic medium is investigated by using the first-order time-dependent perturbation theory of quantum mechanics to calculate transition probabilities between available phonon states. Excluding collinear interactions, it is shown that there are two general types of possible interactions, depending on whether the zeros of the scattered wave displacement amplitude do or do not depend on the third-order elastic constants. Using correspondences between phonon densities and classical displacement amplitudes, and between generated phonons and Huygens wave sources, the theoretical displacement amplitudes for the scattered waves are derived. The amplitudes agree exactly with those derived from classical theory and are plotted for various materials and interaction geometries.

I. INTRODUCTION

MANY authors[1–4] have considered the absorption of ultrasonic waves in an ideal crystal as a result of the sound quanta interacting, via the anharmonic terms of the Hamiltonian, with the lattice vibration quanta. With the advent of recent experimental techniques, it is now possible to study three-phonon interactions in detail. This is done by experimentally generating two noncollinear beams of ultrasonic phonons and, by standard experimental procedures, detecting the phonon beam created by the interaction of the initial phonon beams.[5] In this work the experimentalist uses the language of classical waves, whereas the theorist uses the language of quantum mechanics. This paper bridges the gap between the two disciplines.

The first section describes the first-order time-dependent perturbation theory used to calculate the transition probabilities between different phonon states in a homogeneous isotropic solid. Although this approach is described elsewhere,[1] it is included to make this paper self-contained and to correct some minor errors which have appeared in the literature. It is then shown that, for isotropic media, there are two general types of possible interactions, depending on whether the zeros of the scattered wave amplitude do or do not depend on the third-order elastic constants. The detailed expressions for the scattered wave displacement amplitudes are derived using some interesting relationships between the quantum and classical theories. The results, expressed in classical terms and agreeing exactly with those derived from the classical approach of Jones and Kobett,[6] are plotted for various interaction geometries and for five materials. The theoretical plots are compared with experimental results in Paper II.[7]

II. GENERAL THEORY

Following Slonimskii's approach,[1] the deformation of a solid under stress is described by the components $w_{\alpha\beta}$ of the deformation tensor:

$$w_{\alpha\beta}=\tfrac{1}{2}(u_{\alpha,\beta}+u_{\beta,\alpha}+u_{\gamma,\alpha}u_{\gamma,\beta}), \quad (1)$$

$$u_{\alpha,\beta}=\partial u_\alpha/\partial x_\beta, \quad (2)$$

where u_α is the displacement of a point in the x_α direction, and the Einsteinian notation is used, i.e., repeated indices denote a summation over those indices. The deformation tensor defined by Eq. (1) is quite general and is even valid for finite deformations. In terms of the

* Supported by U. S. Air Force Materials Laboratory, Wright-Patterson Air Force Base, Ohio.

[1] G. L. Slonimskii, Zh. Eksperim. i Teor. Fiz. **12**, 1457 (1937).
[2] L. Landau and G. Rumer, Physik Z. Sowjetunion **11**, 18 (1937).
[3] S. Simons, Proc. Cambridge Phil. Soc. **53**, 702 (1957).
[4] R. L. Orbach, Doctoral thesis, University of California, 1960 (unpublished).
[5] Fred R. Rollins, Jr., Appl. Phys. Letters **2**, 147 (1963).
[6] G. L. Jones and D. R. Kobett, J. Acoust. Soc. Am. **35**, 5 (1963).
[7] Fred R. Rollins, Jr., Lyle H. Taylor, and Paul Todd, following paper, Phys. Rev. **136**, A597 (1964).

deformation tensor, the elastic energy density $\mathcal{H}$ of an isotropic solid is written[8] as

$$\mathcal{H}=\mu w_{\alpha\beta}^2+(\tfrac{1}{2}K-\tfrac{1}{3}\mu)w_{\alpha\alpha}^2+\tfrac{1}{3}A w_{\alpha\beta}w_{\beta\gamma}w_{\gamma\alpha} +Bw_{\alpha\beta}^2w_{\gamma\gamma}+\tfrac{1}{3}Cw_{\alpha\alpha}^3, \quad (3)$$

where μ is the modulus of rigidity, K is the modulus of compression, and A, B, C are the third-order elastic constants. For the purposes of this paper, all terms of fourth order or higher are assumed to be negligible.

Defining two new tensors:

$$u_{\alpha\beta}=\tfrac{1}{2}(u_{\alpha,\beta}+u_{\beta,\alpha}), \quad v_{\alpha\beta}=\tfrac{1}{2}(u_{\alpha,\beta}-u_{\beta,\alpha}), \quad (4)$$

the Hamiltonian density can be written as a sum of two Hamiltonian densities:

$$\mathcal{H}_0=\mu u_{\alpha\beta}^2+(\tfrac{1}{2}K-\tfrac{1}{3}\mu)u_{\alpha\alpha}^2, \quad (5)$$

$$\mathcal{H}'=\tfrac{1}{3}Cu_{\alpha\alpha}^3+(B+\tfrac{1}{2}K-\tfrac{1}{3}\mu)u_{\alpha\beta}^2u_{\gamma\gamma}+(\tfrac{1}{3}A+\mu)u_{\alpha\beta}u_{\beta\gamma}u_{\gamma\alpha} -(\tfrac{1}{2}K-\tfrac{1}{3}\mu)u_{\gamma\gamma}v_{\alpha\beta}v_{\beta\alpha}-\mu u_{\alpha\beta}v_{\beta\gamma}v_{\gamma\alpha}, \quad (6)$$

where the symmetry properties of $u_{\alpha\beta}$ and $v_{\alpha\beta}$ have been used. Using time-dependent perturbation theory, $\mathcal{H}_0$ is the unperturbed Hamiltonian density, and $\mathcal{H}'$ is the perturbation Hamiltonian density which produces nonzero probabilities for transitions between available phonon states.

In general, the displacement vector $\mathbf{u}(\mathbf{r})$ is the sum of the displacement vectors associated with each harmonic wave:

$$\mathbf{u}(\mathbf{r})=\sum_{n=1}^{3}\mathbf{e}_n(a_n e^{i\mathbf{k}_n\cdot\mathbf{r}}+a_{n*}e^{-i\mathbf{k}_n\cdot\mathbf{r}}), \quad (7)$$

where $\mathbf{e}_n$ is a unit vector in the direction of polarization, a_n is the amplitude of the nth phonon, and $\mathbf{k}_n$ is the propagation vector. It follows that

$$u_{\alpha\beta}=u_{\alpha\beta}^1+u_{\alpha\beta}^2+u_{\alpha\beta}^3, \quad v_{\alpha\beta}=v_{\alpha\beta}^1+v_{\alpha\beta}^2+v_{\alpha\beta}^3, \quad (8)$$

where the superscript refers to the number of the phonon state (for typographical convenience the phonon state number will appear as a subscript when tensor component subscripts do not appear). The perturbation Hamiltonian density can now be written as[9]

$$\begin{aligned}\mathcal{H}'=&2Cu_{\alpha\alpha}^1u_{\alpha\alpha}^2u_{\alpha\alpha}^3+(2B+K-\tfrac{2}{3}\mu)\\&\times(u_{\gamma\gamma}^1u_{\alpha\beta}^2u_{\beta\alpha}^3+u_{\gamma\gamma}^2u_{\alpha\beta}^1u_{\beta\alpha}^3+u_{\gamma\gamma}^3u_{\alpha\beta}^1u_{\beta\alpha}^2)\\&+(2A+6\mu)u_{\alpha\beta}^1u_{\beta\gamma}^2u_{\gamma\alpha}^3+(K-\tfrac{2}{3}\mu)\\&\times(u_{\gamma\gamma}^1v_{\alpha\beta}^2v_{\alpha\beta}^3+u_{\gamma\gamma}^2v_{\alpha\beta}^1v_{\alpha\beta}^3+u_{\gamma\gamma}^3v_{\alpha\beta}^1v_{\alpha\beta}^2)\\&+2\mu(u_{\alpha\beta}^1v_{\beta\gamma}^2v_{\alpha\gamma}^3+u_{\alpha\beta}^2v_{\beta\gamma}^1v_{\alpha\gamma}^3+u_{\alpha\beta}^3v_{\beta\gamma}^1v_{\alpha\gamma}^3), \end{aligned} \quad (9)$$

where all terms containing two or three functions with the same superscript have been discarded since they do not pertain to three-phonon interactions.

In quantum theory, the amplitudes of Eq. (7) are the annihilation and creation operators of the linear harmonic oscillator whose only nonzero matrix elements are

$$\langle N\pm1|\binom{a^*}{a}|N\rangle=\left[\frac{\hbar}{2m\omega}\binom{N+1}{N}\right]^{1/2}e^{\pm i\omega t}, \quad (10)$$

where N is the initial number of phonons, $\hbar\omega$ is the phonon energy, t is the time, and m is the mass of the volume of interaction V. To conserve space the large parenthesis notation is used where the upper term is taken when the upper signs of the equation are used, etc.

The matrix elements of the components $u_{\alpha\beta}$ and $v_{\alpha\beta}$ are obtained by differentiating the displacement components:

$$\langle N\pm1|u_{\alpha\beta}|N\rangle=\frac{i}{2}\mp e^{\mp i\mathbf{k}\cdot\mathbf{r}}(e_\alpha k_\beta+e_\beta k_\alpha)\langle N\pm1|\binom{a^*}{a}|N\rangle, \quad (11)$$

$$\langle N\pm1|v_{\alpha\beta}|N\rangle=\frac{i}{2}\mp e^{\mp i\mathbf{k}\cdot\mathbf{r}}(e_\alpha k_\beta-e_\beta k_\alpha)\langle N\pm1|\binom{a^*}{a}|N\rangle. \quad (12)$$

Since $\mathbf{e}\times\mathbf{k}=0$ for longitudinal phonons and $\mathbf{e}\cdot\mathbf{k}=0$ for transverse phonons, $\langle N\pm1|v_{\alpha\beta}|N\rangle$ and $\langle N\pm1|u_{\alpha\alpha}|N\rangle$ vanish, respectively, for longitudinal and transverse phonons.

Assuming that the initial phonons interact for a sufficiently long time, the transition probability (the rate of occurrence of a process per unit time) P between the initial i and final f states is given by[10]

$$P=(2\pi/\hbar)H_{if}'^2\mathfrak{D}_f(\mathcal{E}_i), \quad H_{if}'^2=|\langle f|H'|i\rangle|^2, \quad (13)$$

where $\mathcal{E}_i$ is the energy of the initial state, and $\mathfrak{D}_f(\mathcal{E}_i)$ is the density of final states about $\mathcal{E}_i$. The perturbing Hamiltonian H' is obtained by integrating the perturbing Hamiltonian density over the volume of interaction. The integration yields a V factor provided the total momentum is conserved and zero otherwise.

III. GENERAL INTERACTIONS

From Eq. (11) and the conservation of momentum, it is apparent that the first term of Eq. (9) represents three collinear longitudinal waves. Although Shiren[11] has observed this interaction, it is generally forbidden if dispersion is present and will not be considered further.

Investigating all possible three-phonon interactions satisfying the conservation laws of energy and momentum and recalling that the speed c_l of the longitudinal phonons (L) is always greater than the speed c_t of the

[8] L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon Press, Inc., New York, 1959).

[9] Several perturbing Hamiltonian densities that appear in the literature are in error.

[10] J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1962).

[11] N. S. Shiren, Phys. Rev. Letters **11**, 3 (1963).

transverse phonons (T), the only nonzero interactions are given by

$$L \leftrightarrow T+T, \quad L \leftrightarrow L+T. \tag{14}$$

These interactions will be called the α and β interactions, respectively, with the corresponding matrix elements labeled ${}_\alpha H_{if}'$ and ${}_\beta H_{if}'$. (In anisotropic solids, Herring[12] has shown that the $T \leftrightarrow L+T$ interaction also exists and must be considered.)

To evaluate the foregoing equations, use rectangular Cartesian coordinates, let $\mathbf{u}_1$ be a longitudinal wave traveling in the x_1 direction, let $\mathbf{u}_3$ be a transverse wave, and define the x_2 direction to be in the plane of interaction formed by the three propagation vectors. With these restrictions and letting θ_n be the polarization angle between $\mathbf{e}_n$ and the plane of interaction, the matrix elements can be evaluated from the interaction geometries of Fig. 1 to give

$$\begin{aligned}{}_\alpha H_{if}' = \tfrac{1}{2}(\Omega/c_t^2)\{&\cos\theta_2 \cos\theta_3 \\ &\times[(K+\tfrac{1}{3}\mu)+(2B+K+A+7\mu/3)\cos(2\Phi+2\Psi)] \\ &+\sin\theta_2 \sin\theta_3[(B+K-\tfrac{2}{3}\mu)2\cos(\Phi+\Psi) \\ &+(A+4\mu)\cos\Phi\cos\Psi]\},\end{aligned} \tag{15}$$

$${}_\beta H_{if}' = (\Omega/c_t^2)(2B+K+A+7\mu/3)\cos\theta_3\cos\Phi \times[\cos\Phi \sin 2\Psi+\sin\Phi\cos 2\Psi], \tag{16}$$

where

$$\Omega = V^2\frac{\hbar^3\omega_1\omega_2\omega_3}{8m^3c_l^2c_t^2}\binom{N_1+1}{N_1}\binom{N_2+1}{N_2}\binom{N_3+1}{N_3}. \tag{17}$$

The N_n terms emphasize that the interaction geometries of Fig. 1 do not specify which are the initial and final phonons. When this specification is made, the N_n+1 terms are used for the created phonons and the N_n terms for the annihilated phonons.

It is apparent from Eq. (16) that the zeros of ${}_\beta H_{if}'$ depend only on the interaction geometry. In fact, the matrix element vanishes whenever the polarization vector is perpendicular to the plane of interaction or whenever the longitudinal phonons intersect at right angles. It is even more interesting that the zeros of ${}_\alpha H_{if}'$ depend on the material as well as the interaction

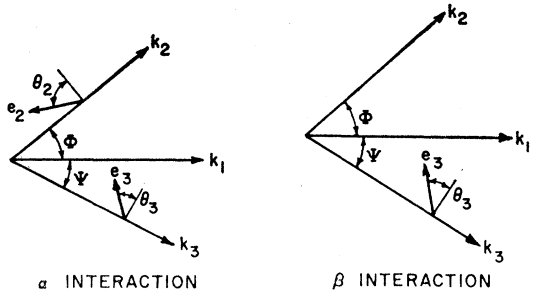

FIG. 1. Phonon vector relationships.

[12] Conyers Herring, Phys. Rev. **95**, 954 (1954).

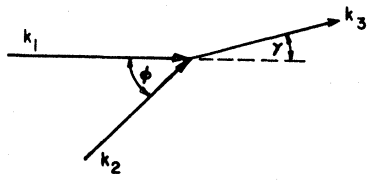

FIG. 2. Interaction geometry.

geometry. This presents the possibility of determining the third-order elastic constants of isotropic solids by experimentally determining the geometry that produces the zeros of this interaction. This determination would be very accurate since it does not depend on any multiplicative factors such as Ω. It is expected that in anisotropic solids the β interaction zeros will also depend on the material constants.

IV. SCATTERED WAVE DISPLACEMENT AMPLITUDES

It is now convenient to change the above notation for labeling the phonon states and their interaction geometries. As diagrammed in Fig. 2, let the first- and second-phonon states represent the experimentally generated input phonons interacting at an angle φ. The third-phonon state then represents the created phonon emitted at an angle γ.

Envisioning a classical elastic wave as being an ideally dense homogeneous beam of phonons, the beam intensity I_n is given by

$$I_n = 10^3\hbar c_n\omega_n\eta_n, \quad I_n = \tfrac{1}{2}10^3\rho c_n\omega_n^2X_n^2, \tag{18}$$

where c_n is the phonon speed, η_n is the phonon density, and the 10^3 factor is the conversion factor from the mks to the cgs units used in this paper. The second equation is the classical expression where X_n is the displacement amplitude. Consequently, the relationship between the phonon density and the displacement amplitude is

$$\eta_n = (\rho\omega_n/2\hbar)X_n^2. \tag{19}$$

Of all the interactions occurring between the η_1 phonons and the η_2 phonons, only a small number, given by the transition probability, will generate η_3 phonons. Each newly created η_3 phonon may be visualized as the center of a Huygens' spherical wave which interacts with adjacent Huygens waves to produce a diffraction pattern. In other words, diffraction from a small volume element, or single aperture, is produced and the appropriate classical equations can be applied. For example, if the scattered beam has a circular cross section of diameter d, the classical expression $15.3\ c_3/\omega_3 d$ gives the angular beam width of the main lobe. We will determine the displacement amplitude at the center of the main lobe in the far field.

Integrating the Huygens' spherical waves over all angles, the transition probability per unit time is evaluated as

$$P = 4\pi r^2\eta_3 c_3, \tag{20}$$

TABLE I. All possible interactions and related factors.

Case No.	Interaction	$\cos\varphi$	$\tan\gamma$	Λ
I	$T(\omega_1)+T(\omega_2)\rightarrow L(\omega_1+\omega_2)$	$c^2-\dfrac{(1-c^2)(a^2+1)}{2a}$	$\dfrac{a\sin\varphi}{1+a\cos\varphi}$	$\dfrac{\sin\theta_2\sin\theta_3}{c_l^3(1+a)^2}\{(B+K-\frac{2}{3}\mu)c^2(1+a)^2\cos\varphi$ $+(2\mu+\frac{1}{2}A)[a+(1+a^2)\cos\varphi+a\cos^2\varphi]\}$ $+\dfrac{\cos\theta_2\cos\theta_3}{c_tc_l^2}\left[\left(2B+K+A+\dfrac{7\mu}{3}\right)\cos^2\varphi-(B+\mu+\frac{1}{2}A)\right]$
II	$L(\omega_1)\rightarrow L(\omega_2)+T(\omega_1-\omega_2)$	$\dfrac{1}{c^2}-\dfrac{(1-c^2)(a^2+1)}{2ac^2}$	$\dfrac{-a\sin\varphi}{1-a\cos\varphi}$	$\dfrac{\cos\theta_3\sin 2\varphi}{2c_l^3}\left(2B+A+K+\dfrac{7\mu}{3}\right)$
III	$L(\omega_1)+T(\omega_2)\rightarrow L(\omega_1+\omega_2)$	$c-\dfrac{a(1-c^2)}{2c}$	$\dfrac{a\sin\varphi}{c+a\cos\varphi}$	$\dfrac{\cos\theta_2\sin\varphi}{c_tc_l^2(1+a)^2}\left(2B+A+K+\dfrac{7\mu}{3}\right)[a+c(2+2a+a^2)\cos\varphi]$
IV	$L(\omega_1)\rightarrow T(\omega_2)+L(\omega_1-\omega_2)$	$c+\dfrac{a(1-c^2)}{2c}$	$\dfrac{-a\sin\varphi}{c-a\cos\varphi}$	$\dfrac{\cos\theta_2\sin\varphi}{c_tc_l^2(1-a)^2}\left(2B+A+K+\dfrac{7\mu}{3}\right)[-a+c(2-2a+a^2)\cos\varphi]$
V	$L(\omega_1)\rightarrow T(\omega_2)+T(\omega_1-\omega_2)$	$\dfrac{1}{c}-\dfrac{(1-c^2)}{2ac}$	$\dfrac{-a\sin\varphi}{c-a\cos\varphi}$	$\dfrac{\sin\theta_2\sin\theta_3}{c_l^3(1+a)}[(B+K-\frac{2}{3}\mu)(c\cos\varphi-a)$ $+(2\mu+\frac{1}{2}A)(c-a\cos\varphi)\cos\varphi]$ $+\dfrac{\cos\theta_2\cos\theta_3}{c_l^3(1-a^2)}\left[\left(2B+K+A+\dfrac{7\mu}{3}\right)(c\cos\varphi-a)^2\right.$ $\left.-(B+\mu+\frac{1}{2}A)(1-a)^2\right]$

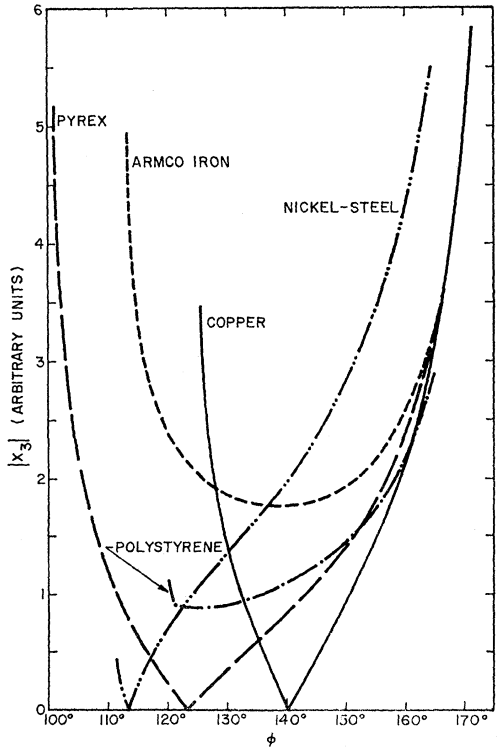

FIG. 3. Interaction case I ($\theta_1=\theta_2=90°$) for five solids.

where r is the distance from the point of observation to the origin of the η_3 phonons and η_3 is evaluated at r. In other words,

$$X_3^2=\hbar P/2\pi r^2\rho\omega_3 c_3. \tag{21}$$

The density of final states can be found in the usual manner to be[13]

$$\mathfrak{D}_f=V\omega_3^2/2\pi^2\hbar c_3^3, \tag{22}$$

where the conservation of energy has been used. Consequently Eqs. (13) and (21) give

$$X_3^2=(V\omega_3/2\pi^2r^2\rho\hbar c_3^4)\,|\langle\eta_1V-1,\eta_2V,1\,|H'|\,\eta_1V,\eta_2V,0\rangle|^2, \tag{23}$$

where appropriate values for N_1, N_2, N_3 are used and it is assumed that

$$\eta_2V\gg 1. \tag{24}$$

Evaluating the matrix elements in Eq. (23), the displacement amplitude of the scattered beam can be written as

$$X_3=X_1X_2V\Lambda\omega_1^3a(1+a)/8\pi r\rho c_tc_l, \tag{25}$$

where $a=\omega_2/\omega_1$ and $c=c_t/c_l$. The value of Λ is a function of the particular interaction and is listed in Table I for the five possible cases arising from the interactions

[13] Leonard I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955).

TABLE II. Elastic constants and c_t/c_l for five materials.

Material	μ 10^{10} dyn/cm^2	λ 10^{10} dyn/cm^2	$c=c_t/c_l$	K 10^{10} dyn/cm^2	A 10^{10} dyn/cm^2	B 10^{10} dyn/cm^2
REX-535 Nickel-Steel[a]	78.00	90.90	0.562	142.9	− 730	− 225
Copper[b]	46.00	131.0	0.454	161.7	−1590	170
Armco iron[c]	82.00	110.0	0.547	164.7	1100	−1580
Polystyrene[c]	1.381	2.889	0.495	3.810	− 10	− 8.3
Pyrex[c]	27.50	13.53	0.633	31.86	420	− 118

[a] See Ref. 15.
[b] See Ref. 16.
[c] See Ref. 14.

depicted in (14). The factors determining the interaction geometries (see Fig. 2) are also included in Table I. Cases I and V are α interactions and cases II, III, and IV are β interactions. (Note that cases II, IV, and V could be utilized to make an ultrasonic amplifier.) It can be shown that for case I, Eq. (25) is completely symmetric in ω_1 and ω_2, i.e., for this case the scattered wave displacement amplitude is independent of the labeling of the two input waves. It can also be shown from Eq. (25) that case V is just the reverse interaction of case I and that cases II, III, and IV are all reverse interactions of each other. This is as it should be.

Jones and Kobett[6] investigate this same problem by using the classical wave approach and applying the Fourier transform method for solving the inhomogeneous vector wave equation. However, the following term, in their notation,

$$-(K-\tfrac{2}{3}\mu+B)[(\mathbf{A}_0\cdot\mathbf{k}_1)(\mathbf{k}_1\cdot\mathbf{k}_2)\mathbf{B}_0\pm(\mathbf{B}_0\cdot\mathbf{k}_2)(\mathbf{k}_1\cdot\mathbf{k}_2)\mathbf{A}_0], \tag{26}$$

where $\mathbf{A}_0$ and $\mathbf{B}_0$ are the input wave displacement amplitudes, was inadvertently omitted from their $\mathbf{I}^{\pm}$ vector. Including this term and calculating the scattered wave displacement amplitudes from their results, Eq. (25) is again obtained. This is most gratifying.

V. NUMERICAL RESULTS

There are only five isotropic materials for which the third-order elastic constants have been published.[14–16] The difficulties encountered in these measurements can be appreciated by the large experimental errors—on the order of 100% for some of the constants. The available third-order elastic constants and other pertinent data are tabulated in Table II.

Using the data in Table II, the value of $|X_3|$ as given by Eq. (25) can be calculated as a function of the interaction angle φ. The only unknown is the value of the volume of interaction V. We will use, as an approximation, the volume common to two completely intersecting cylinders, each of diameter l, i.e.,

$$V\approx\tfrac{2}{3}l^3\csc\varphi. \tag{27}$$

Only the absolute value of Eq. (25) should be calculated since the sign has been ignored in the derivation and since only X_3^2 is pertinent to experimental measurements.

Figure 3 is a plot, for the five materials listed in Table II, of $|X_3|$ versus φ for case I with $\theta_1=\theta_2=90°$. In order to present all of the calculations in a readable manner without circuitous explanations, the values of $|X_3|$ are given in arbitrary units and *each* curve has been arbitrarily shifted in the vertical direction. In other words, only the shapes of the curves are accurately retained. This is also true for Figs. 4 and 5. The actual values of the displacement amplitudes for all interactions in iron, copper, polystyrene, Pyrex, and nickel-steel, are available from the authors.

Case I is an α interaction and it is quite apparent in Fig. 3 that the zeros of the displacement amplitude do indeed depend on the material constants. It is interesting that the curves for polystyrene and iron do not have

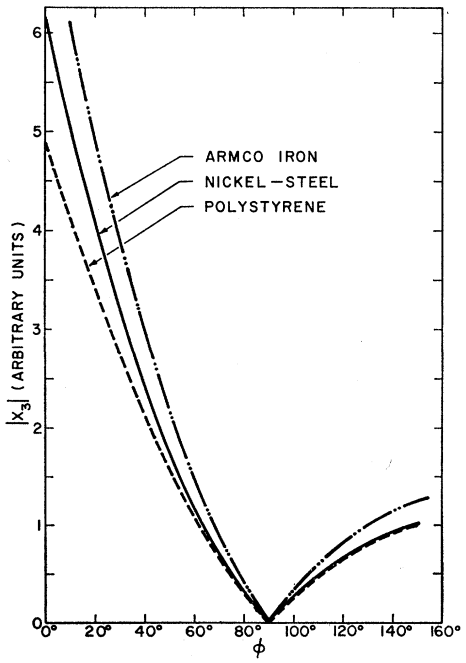

FIG. 4. Interaction case II ($\theta_3=0°$) for three solids.

[14] D. S. Hughes and J. L. Kelly, Phys. Rev. **92**, 1145 (1953).
[15] D. I. Crecraft, Nature **195**, 1193 (1962).
[16] A. Seeger and O. Buck, Z. Naturforsch. **15a**, 1056 (1960).

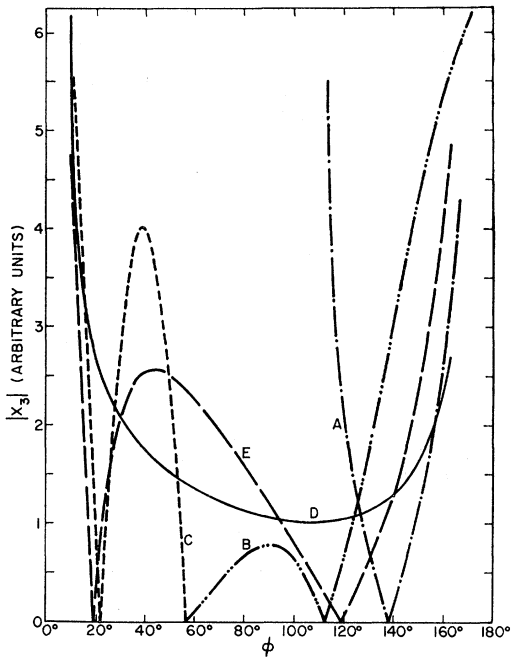

FIG. 5. Five interaction cases for iron: (A) case I ($\theta_1=\theta_2=0°$); (B) case III ($\theta_2=0°$); (C) case IV ($\theta_2=0°$); (D) case V ($\theta_2=\theta_3=90°$); (E) case V ($\theta_2=\theta_3=0°$).

zeros but do exhibit minimums. However, zeros may exist for a different choice of polarization angles. The end points of each curve essentially represent the angular relationship at which the conservation laws of energy and momentum can no longer be satisfied. These points can be easily found from Table I and the requirement that $|\cos\varphi|\leq 1$.

Figure 4 is a plot, for iron, polystyrene, and steel, of case II with $\theta_3=0°$. This case is a β interaction and, of course, the zeros of the curves are independent of the material. Caution must be exercised in interpreting these curves since the approximation in Eq. (27) dominates each curve shape near interaction angles of 0 and 180°.

Figure 5 is a plot for iron of the displacement amplitude curves for several additional interaction cases. All of these figures illustrate the strong dependence of the scattered wave displacement amplitude on the properties of the solid and on the interaction geometry.

VI. SUMMARY

The nonlinear interaction of two elastic waves in a homogeneous, isotropic solid has been investigated by using the first-order time-dependent perturbation theory of quantum mechanics to calculate transition probabilities between available phonon states. Excluding collinear interactions, it has been shown that there are two general types of possible interactions, depending on whether the zeros of the scattered wave displacement amplitude do or do not depend on the third-order elastic constants. The former type could be used to evaluate the third-order elastic constants. A relationship between phonon density and displacement amplitude was then derived. Using the concept that newly created phonons are sources of Huygens waves, the exact scattered wave displacement amplitudes were then derived for all possible interactions. The resultant displacement amplitudes agree exactly with those derived from the classical approach of applying the Fourier transform method for solving the inhomogeneous vector wave equation. The predicted displacement amplitudes were then plotted for various materials and interaction geometries. The results will be compared to experimental measurements in paper II.[7]

ACKNOWLEDGMENTS

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