This process can be repeated until the predicted and corrected values of $\xi_i^{(n+1)}$ differ by less than a prescribed value. However, the procedure adopted was to make trial runs on the system of 864 particles with one and with two repetitions of this predictor-corrector procedure. A comparison of the results in terms of the correlations discussed in this paper showed no observable difference. As a further check, the motion of a diatomic system was calculated with one and with two repetitions of this procedure. The two particles were initially at a distance $\rho_{12}=1.9$ and were allowed to oscillate; their positions at 2000 successive intervals Δu were recorded covering a little over three periods of oscillation and the following is a summary of the results to show the degree to which the approximations involved in using the difference equations affect the motion.

(a) At the end of three successive oscillations the separations were: 1.8958, 1.8932, 1.8890, when the predictor-corrector procedure was used only once and 1.9018, 1.9016, and 1.9044 when it was used twice, thus giving improved results.

(b) The distance of closest approach was successively 1.0039, 1.0040, 1.0041 in the first case and 1.0038, 1.0038, 1.0038 in the other.

(c) The mean-square velocity in °K while going through the minimum of the potential was 36.65, 36.61, 36.60, 36.59, 36.59, 36.54, in one case and 36.65, 36.67, 36.67, 36.68, 36.67, 36.70, in the other.

(d) The period of oscillation was (in units of 10^{-12} sec) 6.27, 6.22, 6.17, in one case, and 6.31, 6.32, 6.33 in the other.

This gives an idea of the errors involved in using the difference equations given above. The results given in the paper were all obtained in a run with two passes through the predictor-corrector procedure.

There are five factors which determine the time for computing one step Δu, namely, N, R, the number of predictor-corrector cycles, the manner of writing the program, and the computer used. For $N=864, R=2.25\sigma$, using floating point arithmetic each cycle takes 45 sec on the CDC-3600 computer. For $N=250$, $R=2.0\sigma$, using fixed point arithmetic each cycle takes 40 sec on the IBM-704 machine. For the most time consuming part the program was written in machine language and in FORTRAN for the rest.

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Interactions Between Elastic Waves in an Isotropic Solid*

J. D. CHILDRESS AND C. G. HAMBRICK†
Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana
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Interactions between elastic waves in an isotropic solid are studied in the elastic-continuum approximation. The analysis is carried out completely in a wave-packet formalism, i.e., scattering of wave packets by wave packets. The maximum amplitude (or intensity) and width of the scattered wave packet are expressed in terms of the maximum amplitudes, frequencies, widths, polarizations, and relative propagation directions of the primary-wave packets. The polarization relations and frequency ranges for the allowed interaction processes are obtained; these are essentially identical to the ones given by Jones and Kobett. The results are shown to be in good order-of-magnitude agreement with the experiments of Rollins. Possible application of elastic-wave scattering to the determination of third-order elastic constants is discussed.

I. INTRODUCTION

ELASTIC waves in solids have attracted much theoretical and experimental interest; but surprisingly, the interaction between elastic waves (phonon-phonon scattering) has been investigated experimentally only recently. Last year, Rollins[1] observed directly the production of "sum" and "difference" frequency waves from the interaction of two ultrasonic pulses in aluminum. Somewhat earlier, Gedroits and Krasil'nikov[2] demonstrated the effects of such interactions on the attenuation and harmonic distortion of an ultrasonic wave interacting with itself. Mahler, Mahon, Miller, and Tantilla[3] and Shiren[4] later reported observations of the same phenomena by different experimental means. At about the same time, Jones and Kobett[5] (classical approach) and Childress and Fried[6] (quantum-mechanical approach) discussed elastic-wave interactions and found that such processes should indeed be experimentally observable.

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† Present address: Department of Physical Sciences, McNeese State College, Lake Charles, Louisiana.

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[5] G. L. Jones and D. Kobett, J. Acoust. Soc. Am. **35**, 5 (1963). An erratum notes the omission of a term

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

in the expression for $\mathbf{I}^{\pm}$ below Eq. (4) (their notation).

[6] J. D. Childress and Z. Fried, Bull. Am. Phys. Soc. **8**, 16 (1963).

Elastic waves in a lattice interact one with the other through the lattice potential expansion terms of cubic and higher order in particle displacements, the anharmonic terms. Similarly, terms nonlinear in the deformations obtained in the general theory of elastic continua allow interactions, i.e., scattering of elastic waves by elastic waves. In the following, the development is based on the continuum approximation; this is not much of a restriction since the highest frequencies available experimentally are well within the region of validity of the approximation. The same reason justifies neglect of dispersion. Additional approximations limit the validity of the present study to interactions in which the scattered-wave amplitude is small relative to the primary wave amplitudes.

The present study considers the interaction of two elastic-wave wave packets in an isotropic solid. Results are obtained in terms of quantities of direct experimental interest. We choose the wave-packet treatment as most suitable for comparison with the experimental results of Rollins.[1] A similar treatment is indicated for comparison with Shiren's work; however, the "strong" effects and particular experimental conditions of Shiren[4] cannot be handled directly in the present analysis. Further, the wave-packet approach avoids the awkwardness, particularly in amplitude and intensity expressions, inherent in the plane-wave approach of Jones and Kobett.[5]

Since many experiments in solid-state physics, either for convenience or from necessity, use pulse excitations and consequently would be described most naturally and faithfully in the wave-packet formalism, the present study is perhaps of some interest as an example of a treatment completely in terms of wave packets.

II. APPROXIMATE SOLUTION FOR THE SCATTERED WAVE PACKET

A. The Wave Packets

The two incident wave packets are assumed for mathematical convenience to have the spherically symmetric Gaussian form

$$\mathbf{u}_i^{\pm}(\mathbf{x},t)=(2\pi)^{-3/2}\mathbf{A}_i^{\pm}\int d^3k_i'\,\exp[-\tfrac{1}{2}(\mathbf{k}_i'-\mathbf{k}_i)^2\Delta_i^{-2}]\times\exp[\pm i(\mathbf{k}_i'\cdot\mathbf{x}-\omega_i(k_i')t)],\quad(1)$$

$$\mathbf{A}_i^{\pm}=\mathbf{U}_i^{\pm}/\Delta_i^3,$$

where $\mathbf{u}_i$ is the deformation vector, $\mathbf{U}_i$ is the maximum of $\mathbf{u}_i$, $\mathbf{k}_i'$ is the wave vector of the wave of angular frequency $\omega_i(k_i')$, and Δ_i is the k-space width of the ith wave packet with mean wave vector $\mathbf{k}_i$. As a convention, we take the angular frequency of the first wave $\omega_1(k_1)$ to be greater than or equal to that of the second wave, $\omega_2(k_2)$; this involves no loss in generality, being merely a labeling for the waves.

The $(\pm)$ superscript on $\mathbf{u}$ and the corresponding $(\pm)$ signs on the right-hand side of Eq. (1) have the following significance: If the wave produced in the interaction (hereafter called the scattered wave) is a sum wave, i.e., angular frequency $\omega_1+\omega_2$, the interaction involves an absorption of energy from both primary waves and requires use of the $(+)$ sign for both $\mathbf{u}_1$ and $\mathbf{u}_2$. If the scattered wave is a difference wave, angular frequency $\omega_1-\omega_2$, the interaction takes energy from the ω_1 wave and delivers part of that energy to the ω_2 wave; thus, the $(+)$ sign must be taken for $\mathbf{u}_1$ but the $(-)$ sign taken for $\mathbf{u}_2$.

The scattered wave in the interaction region is written by means of the four-dimensional Fourier transform as

$$\mathbf{u}_{\pm}(\mathbf{x},t)=\frac{1}{(2\pi)^{3/2}}\int\frac{d^3k_{\pm}'d\omega_{\pm}'}{2\omega_{\pm}'}\mathbf{C}_{\pm}(\mathbf{k}_{\pm}',\omega_{\pm}')\times\exp[i(\mathbf{k}_{\pm}'\cdot\mathbf{x}-\omega_{\pm}'t)],\quad(2)$$

where the $2\omega_{\pm}'$ is introduced in the denominator to give $\mathbf{C}_{\pm}$ the same dimensions as $\mathbf{A}_i$ and the $(+)$ and $(-)$ subscripts label sum and difference wave quantities, respectively. Outside the interaction region, the scattered wave is another free wave in the medium and, in the Gaussian approximation, should have the form of Eq. (1), i.e.,

$$\frac{\mathbf{C}_{\pm}(\mathbf{k}_{\pm}',\omega_{\pm}')}{2\omega_{\pm}'}\rightarrow\mathbf{C}_{\pm}\delta[\omega_{\pm}'-\omega_{\pm}(k_{\pm}')]\exp\left[-\frac{(\mathbf{k}_{\pm}'-\mathbf{k}_{\pm})^2}{2\Delta^2}\right].$$

The directions of $\mathbf{A}_i$ and $\mathbf{C}_{\pm}$ vectors give the directions of the respective deformations. These can be expressed as

$$\mathbf{A}_i=A_i\boldsymbol{\varepsilon}_i,\quad\mathbf{C}_{\pm}=C_{\pm}\boldsymbol{\varepsilon}_{\pm},$$

where $\boldsymbol{\varepsilon}_i$ and $\boldsymbol{\varepsilon}_{\pm}$ are the polarization vectors (unit vectors) of the corresponding waves.

Suitable factors could be introduced into the wave-packet expressions so that the propagation of each wave packet would be displayed explicitly; then time-of-flight results would be obtainable. Since these results are essentially trivial, we omit such factors.

B. The Equation of Motion and the Principal Approximation

The equation of motion for a wave in an isotropic elastic medium is

$$\rho_0\frac{\partial^2u_i}{\partial t^2}-\sum_j\left[\mu\frac{\partial^2u_i}{\partial x_j^2}+\alpha\frac{\partial^2u_j}{\partial x_i\partial x_j}\right]$$
$$=\sum_{j,k}\left\{\beta\left[\frac{\partial^2u_j}{\partial x_k^2}\frac{\partial u_j}{\partial x_i}+\frac{\partial^2u_j}{\partial x_k^2}\frac{\partial u_i}{\partial x_j}+2\frac{\partial^2u_i}{\partial x_j\partial x_k}\frac{\partial u_j}{\partial x_k}\right]\right.$$
$$+\gamma\left[\frac{\partial^2u_j}{\partial x_i\partial x_k}\frac{\partial u_j}{\partial x_k}+\frac{\partial^2u_k}{\partial x_j\partial x_k}\frac{\partial u_i}{\partial x_j}\right]+(\gamma-\beta)\frac{\partial^2u_i}{\partial x_k^2}\frac{\partial u_j}{\partial x_j}$$
$$+(\gamma-\alpha)\left[\frac{\partial^2u_k}{\partial x_j\partial x_k}\frac{\partial u_j}{\partial x_i}+\frac{\partial^2u_j}{\partial x_i\partial x_k}\frac{\partial u_k}{\partial x_j}\right]$$
$$\left.+\delta\frac{\partial^2u_k}{\partial x_i\partial x_k}\frac{\partial u_j}{\partial x_j}\right\}\quad(3)$$

to third order in the deformation. Here ρ_0 is the density of the undeformed solid; μ is the shear modulus; and the coefficients α, β, γ, and δ are defined

$$\alpha=K+\tfrac{1}{3}\mu\,,$$
$$\beta=\mu+\tfrac{1}{4}A\,,$$
$$\gamma=K+\tfrac{1}{3}\mu+\tfrac{1}{4}A+B\,,$$
$$\delta=B+2C\,,$$

where K is the modulus of compression and A, B, and C are the third-order elastic constants.[7] The linear equation of motion results if the left-hand side of Eq. (3) is set equal to zero.

For an approximate solution of the nonlinear equation, Eq. (3), we write the total deformation as

$$\mathbf{u}=\mathbf{u}_1{}^{+}+\mathbf{u}_2{}^{\pm}+\mathbf{u}_{\pm}.$$

The principal approximation for this calculation consists of the following: (1) $\mathbf{u}_1{}^{+}$ and $\mathbf{u}_2{}^{\pm}$ are taken to be solutions of the linear equation, hence vanish in the left-hand side of Eq. (3); and (2) terms in $\mathbf{u}_{\pm}$ on the right-hand side of Eq. (3) are neglected. Clearly the approximation is that $\mathbf{u}_{\pm}$ is small relative to $\mathbf{u}_1{}^{+}$ and $\mathbf{u}_2{}^{\pm}$. Further, second-harmonic terms of the primary waves are omitted; these interactions have been treated by Gol'dberg[8] and are not considered here.

C. The Secondary Approximation and the Solution

We substitute Eqs. (1) and (2) into Eq. (3), multiply both sides by $\exp[-i(\mathbf{k}_{\pm}''\cdot\mathbf{x}-\omega_{\pm}''t)]$, integrate over all space and time, then integrate over $\mathbf{k}_{\pm}''$ on the left-hand side by means of the Dirac delta functions which result from the previous step, solve for $\mathbf{C}_{\pm}(\mathbf{k}_{\pm}',\omega_{\pm}')$, and, finally, substitute that into Eq. (2) to obtain

$$\begin{aligned}u_{\pm}(\mathbf{x},t)=&\frac{iA_1{}^{+}A_2{}^{\pm}}{(2\pi)^3\rho_0}\int\frac{d^3k_{\pm}'d\omega_{\pm}'d^3k_1'd^3k_2'}{[\omega_{\pm}'^2-\omega_{\pm}^2(k_{\pm}')]}\delta^3(\mathbf{k}_1'\pm\mathbf{k}_2'-\mathbf{k}_{\pm}')\\&\times\delta[\omega_1(k_1')\pm\omega_2(k_2')-\omega_{\pm}']\mathbf{G}(\mathbf{k}_1',\pm\mathbf{k}_2',\boldsymbol{\varepsilon}_1,\boldsymbol{\varepsilon}_2)\cdot\boldsymbol{\varepsilon}_{\pm}\\&\times\exp[-\tfrac{1}{2}(\mathbf{k}_1'-\mathbf{k}_1)^2\Delta_1{}^{-2}-\tfrac{1}{2}(\mathbf{k}_2'-\mathbf{k}_2)^2\Delta_2{}^{-2}]\\&\times\exp[i(\mathbf{k}_{\pm}'\cdot\mathbf{x}-\omega't)],\end{aligned}\tag{4}$$

where

$$\begin{aligned}\mathbf{G}(\mathbf{k}_1,\pm\mathbf{k}_2,\boldsymbol{\varepsilon}_1,\boldsymbol{\varepsilon}_2)=&\beta\{2(\boldsymbol{\varepsilon}_1\cdot\boldsymbol{\varepsilon}_2)[(\pm k_1{}^2+\mathbf{k}_1\cdot\mathbf{k}_2)\mathbf{k}_2+(k_2{}^2\pm\mathbf{k}_1\cdot\mathbf{k}_2)\mathbf{k}_1]\\&-(\pm k_1{}^2+\mathbf{k}_1\cdot\mathbf{k}_2)(\boldsymbol{\varepsilon}_2\times\mathbf{k}_2)\times\boldsymbol{\varepsilon}_1-(k_2{}^2\pm\mathbf{k}_1\cdot\mathbf{k}_2)(\boldsymbol{\varepsilon}_1\times\mathbf{k}_1)\times\boldsymbol{\varepsilon}_2\}\\&+(2\gamma-\alpha)[(\boldsymbol{\varepsilon}_1\cdot\boldsymbol{\varepsilon}_2)(\mathbf{k}_1\cdot\mathbf{k}_2)(\pm\mathbf{k}_1+\mathbf{k}_2)\pm(\boldsymbol{\varepsilon}_1\cdot\mathbf{k}_1)(\mathbf{k}_1\cdot\mathbf{k}_2)\boldsymbol{\varepsilon}_2+(\boldsymbol{\varepsilon}_2\cdot\mathbf{k}_2)(\mathbf{k}_1\cdot\mathbf{k}_2)\boldsymbol{\varepsilon}_1]\\&+(\gamma-\beta)[(\boldsymbol{\varepsilon}_1\cdot\mathbf{k}_1)k_2{}^2\boldsymbol{\varepsilon}_2\pm(\boldsymbol{\varepsilon}_2\cdot\mathbf{k}_2)k_1{}^2\boldsymbol{\varepsilon}_1]\\&+(\gamma-\alpha)[\pm(\boldsymbol{\varepsilon}_1\cdot\mathbf{k}_1)(\boldsymbol{\varepsilon}_2\times\mathbf{k}_2)\times\mathbf{k}_1+(\boldsymbol{\varepsilon}_2\cdot\mathbf{k}_2)(\boldsymbol{\varepsilon}_1\times\mathbf{k}_1)\times\mathbf{k}_2+(\boldsymbol{\varepsilon}_1\times\mathbf{k}_1)\cdot(\boldsymbol{\varepsilon}_2\times\mathbf{k}_2)(\pm\mathbf{k}_1+\mathbf{k}_2)]\\&+\delta(\boldsymbol{\varepsilon}_1\cdot\mathbf{k}_1)(\boldsymbol{\varepsilon}_2\cdot\mathbf{k}_2)(\pm\mathbf{k}_1+\mathbf{k}_2).\end{aligned}\tag{5}$$

The form of the $\mathbf{G}$ function obtained directly from the right-hand side of Eq. (3) is identical to the corrected expression for $\mathbf{I}^{\pm}$ below Eq. (4) of Jones and Kobett.[5] The above expression for $\mathbf{G}$, obtained by straightforward vector manipulations, is especially easy to interpret for different wave polarizations.

The terms on the left-hand side of Eq. (3) from which $\omega_i(k_i)$ is defined are

$$(\mu/\rho_0)k_i{}^2\boldsymbol{\varepsilon}_i+(\alpha/\rho_0)(\boldsymbol{\varepsilon}_i\cdot\mathbf{k}_i)\mathbf{k}_i.$$

Since only transverse and longitudinal polarizations need be considered in an isotropic solid, the above is set equal to $\omega^2(k)\boldsymbol{\varepsilon}$, where

$$\omega^2(k)=c_t{}^2k^2,c_t{}^2=\mu/\rho_0\tag{6}$$

for a transverse wave and

$$\omega^2(k)=c_l{}^2k^2,\quad c_l{}^2=(\mu+\alpha)/\rho_0\tag{7}$$

for a longitudinal wave.

We can now integrate Eq. (4) provided we assume (the secondary assumption for the approximate solution) that quantities slowly varying relative to the singular functions and the Gaussians can be taken outside the integrals. This is essentially an assumption that the wave packets are "sharp" in k space, i.e.,

$$\Delta_i^2\ll k_i^2.$$

Details of the integration and additional necessary approximations are given in the Appendix; the final result in terms of maximum amplitudes is

$$\begin{aligned}u_{\pm}(\mathbf{x},t)=&\tfrac{1}{4}(\tfrac{1}{2}\pi)^{1/2}\rho_0{}^{-1}U_1{}^{+}U_2{}^{\pm}\mathbf{G}(\mathbf{k}_1,\pm\mathbf{k}_2,\boldsymbol{\varepsilon}_1,\boldsymbol{\varepsilon}_2)\cdot\boldsymbol{\varepsilon}_{\pm}\\&\times[\omega_1(k_1)\pm\omega_2(k_2)]^{-1}\\&\times(\Delta/\Delta_1\Delta_2)^3(c_1{}^{-1}\Delta_1{}^2+c_2{}^{-1}\Delta_2{}^2)\exp[-\tfrac{1}{2}\mathbf{x}^2\Delta^2]\\&\times\exp[i\{(\mathbf{k}_1\pm\mathbf{k}_2)\cdot\mathbf{x}-[\omega_1(k_1)\pm\omega_2(k_2)]t\}].\end{aligned}\tag{8}$$

Here the width in k space of the scattered wave packet is

$$\Delta^2=\frac{(c_1{}^2-2c_1c_2\cos\theta+c_2{}^2)\Delta_1{}^2\Delta_2{}^2}{c_{\pm}{}^2\{1-(1/c_1c_2)[(c_2{}^2\omega_1\pm c_1{}^2\omega_2)/(\omega_1\pm\omega_2)]\cos\theta\}^2(\Delta_1+\Delta_2)^2},\tag{9}$$

[7] The notation for the elastic constants is conventional. The A and C elastic constants are not to be confused with the $A_i{}^{\pm}$ and $C_{\pm}$ amplitude notation.

[8] Z. A. Gol'dberg, Akust. Zh. 6, 307 (1960) [English transl.: Soviet Phys.—Acoust. 6, 306 (1961)].

where

$$\cos\theta = \mathbf{k}_1\cdot\mathbf{k}_2/k_1k_2,$$
$$\neq \pm 1.$$

The derivation of Eq. (9) necessarily excludes the $\cos\theta=\pm1$ values. Hereafter all ω's are $\omega(k)$'s; therefore, the functional notation is omitted in the following.

III. POLARIZATION RELATIONS AND THE ALLOWED SCATTERING PROCESSES

A. Interaction Cases

Inspection of the expression for $\mathbf{G}(\mathbf{k}_1,\pm\mathbf{k}_2,\boldsymbol{\varepsilon}_1,\boldsymbol{\varepsilon}_2)$ given in Eq. (5) shows that the interaction between the two wave packets depends strongly on their polarizations. For analysis of this dependence, we define a convenient coordinate system with respect to the scattering plane, the plane in which the (mean) wave vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_\pm$ lie, as

$$\mathbf{n}_1=\mathbf{k}_1/k_1,$$
$$\mathbf{n}_2=\mathbf{n}_3\times\mathbf{n}_1,$$
$$\mathbf{n}_3=\mathbf{k}_1\times\mathbf{k}_2/|\mathbf{k}_1\times\mathbf{k}_2|.$$

The three unit vectors $\mathbf{n}_1$, $\mathbf{n}_2$, $\mathbf{n}_3$ form a right-handed Cartesian system.

With reference to the scattering plane, each wave has the following three possible polarizations:

(1) longitudinal (denoted by l)

$$\boldsymbol{\varepsilon}=\mathbf{k}/k,$$

(2) transverse in the plane (denoted by t)

$$\boldsymbol{\varepsilon}=\mathbf{n}_3\times\mathbf{k}/k,$$

and

(3) transverse normal to the plane (denoted by τ)

$$\boldsymbol{\varepsilon}=\mathbf{n}_3.$$

The three polarizations taken together with the convention $\omega_1\geq\omega_2$ yield nine interaction cases (a measure of redundancy exists here but the clarity and definiteness is worth the slight cost in elaboration). The interaction cases with the corresponding $\mathbf{G}$ vectors (expressed in most convenient terms) are—

Case I:

$$p_1=l,\quad p_2=l,$$
$$\mathbf{G}_{\mathrm{I}}=c_l^{-3}\omega_1\omega_2[\{[(2K+5/3\mu+A+4B+2C)+(K+7/3\mu+A+2B)\cos^2\theta]\omega_2\cos\theta \pm[(K-\tfrac{2}{3}\mu+2B+2C)+2(K+7/3\mu+A+2B)\cos^2\theta]\omega_1\}\mathbf{n}_1+\{[(K+\tfrac{2}{3}\mu+2B+2C) +(K+7/3\mu+A+2B)\cos^2\theta]\omega_2\pm(K+7/3\mu+A+2B)\omega_1\cos\theta\}\sin\theta\mathbf{n}_2];\quad(10)$$

Case II:

$$p_1=l,\quad p_2=t,$$
$$\mathbf{G}_{\mathrm{II}}=(c_lc_t)^{-1}\omega_1\omega_2[\{-[(K+\tfrac{4}{3}\mu+\tfrac{1}{2}A+B)+(K+7/3\mu+A+2B)\cos^2\theta]c_t^{-1}\omega_2 \mp2(K+7/3\mu+A+2B)c_l^{-1}\omega_1\cos\theta\}\sin\theta\mathbf{n}_1 +\{[(K+\tfrac{1}{3}\mu+\tfrac{1}{4}A+B)-(K+7/3\mu+A+2B)\sin^2\theta]c_t^{-1}\omega_2\cos\theta \pm[(K+\tfrac{4}{3}\mu+\tfrac{1}{2}A+B)-(K+7/3\mu+A+2B)\sin^2\theta]c_l^{-1}\omega_1\}\mathbf{n}_2];\quad(11)$$

Case III:

$$p_1=t,\quad p_2=l,$$
$$\mathbf{G}_{\mathrm{III}}=(c_lc_t)^{-1}\omega_1\omega_2[\{[(\mu+\tfrac{1}{2}A+B)+(K+7/3\mu+A+2B)\cos^2\theta]c_l^{-1}\omega_2 \pm(K+10/3\mu+5/4A+2B)c_t^{-1}\omega_1\cos\theta\}\sin\theta\mathbf{n}_1+\{[(K+\tfrac{4}{3}\mu+\tfrac{1}{2}A+B) +(K+7/3\mu+A+2B)\sin^2\theta]c_l^{-1}\omega_2\cos\theta\pm[(K+\tfrac{1}{3}\mu+\tfrac{1}{4}A+B)+(\mu+\tfrac{1}{4}A)\sin^2\theta]c_t^{-1}\omega_1\}\mathbf{n}_2];\quad(12)$$

Case IV:

$$p_1=t,\quad p_2=t,$$
$$\mathbf{G}_{\mathrm{VI}}=c_t^{-3}\omega_1\omega_2[\{[(\tfrac{1}{4}A+B)+(K+7/3\mu+A+2B)\cos^2\theta]\omega_2\cos\theta \pm[-(\mu-B)+(K+10/3\mu+5/4A+2B)\cos^2\theta]\omega_1\}\mathbf{n}_1+\{[-(\mu-B)+(K+7/3\mu+A+2B)\cos^2\theta]\omega_2 \pm(\mu+\tfrac{1}{4}A)\omega_1\cos\theta\}\sin\theta\mathbf{n}_2];\quad(13)$$

Case V:

$$p_1=\tau,\quad p_2=\tau,$$
$$\mathbf{G}_{\mathrm{V}}=c_t^{-3}\omega_1\omega_2[\{[(\mu+\tfrac{1}{4}A)+(K+\tfrac{4}{3}\mu+A+3B)\cos^2\theta]\omega_2\pm(K+7/3\mu+5/4A+3B)\omega_1\cos\theta\}\mathbf{n}_1 +[(K+\tfrac{4}{3}\mu+A+3B)\omega_2\cos\theta\pm(\mu+\tfrac{1}{4}A)\omega_1]\sin\theta\mathbf{n}_2];\quad(14)$$

Case VI:

$$p_1=l, \quad p_2=\tau,$$
$$\mathbf{G}_{\mathrm{VI}}=(c_lc_t)^{-1}\omega_1\omega_2\{[(K-\tfrac{2}{3}\mu+B)+(\mu+\tfrac{1}{4}A)\cos^2\theta]c_t^{-1}\omega_2\pm(K+\tfrac{4}{3}\mu+\tfrac{1}{2}A+B)c_l^{-1}\omega_1\cos\theta\}\mathbf{n}_3; \tag{15}$$

Case VII:

$$p_1=\tau, \quad p_2=l,$$
$$\mathbf{G}_{\mathrm{VII}}=(c_lc_t)^{-1}\omega_1\omega_2\{(K+\tfrac{4}{3}\mu+\tfrac{1}{2}A+B)c_l^{-1}\omega_2\cos\theta\pm[(K-\tfrac{2}{3}\mu+B)+(\mu+\tfrac{1}{4}A)\cos^2\theta]c_t^{-1}\omega_1\}\mathbf{n}_3; \tag{16}$$

Case VIII:

$$p_1=\tau, \quad p_2=t,$$
$$\mathbf{G}_{\mathrm{VIII}}=c_t^{-3}\omega_1\omega_2[-(\mu+\tfrac{1}{4}A)(\omega_2\pm\omega_1\cos\theta)]\sin\theta\mathbf{n}_3; \tag{17}$$

Case IX:

$$p_1=t, \quad p_2=\tau,$$
$$\mathbf{G}_{\mathrm{IX}}=c_t^{-3}\omega_1\omega_2(\mu+\tfrac{1}{4}A)(\omega_2\cos\theta\pm\omega_1)\sin\theta\mathbf{n}_3. \tag{18}$$

B. Polarization Relations

Since the scalar product $\mathbf{G}\cdot\boldsymbol{\varepsilon}_\pm$ appears in Eq. (8), the direction of the $\mathbf{G}$ vector determines the polarization of the scattered wave. For cases I to V, $\mathbf{G}$ lies in the scattering plane; hence the possible polarizations of the scattered wave are $p_\pm=l$ and $p_\pm=t$. For cases VI to IX, $\mathbf{G}$ is normal to the scattering plane and the only possible polarization of the scattered wave is $p_\pm=\tau$.

The above polarization relations are understandable in cases I to IV. In these cases, both $\boldsymbol{\varepsilon}_1$ and $\boldsymbol{\varepsilon}_2$ lie in the scattering plane and that $\boldsymbol{\varepsilon}_\pm$ also lies in the plane seems reasonable. Cases VI to IX are harder to understand. These cases have one, but not both, of $\boldsymbol{\varepsilon}_1$ and $\boldsymbol{\varepsilon}_2$ normal to the plane and $\boldsymbol{\varepsilon}_\pm$ normal to the plane. Case V appears surprising at first; both $\boldsymbol{\varepsilon}_1$ and $\boldsymbol{\varepsilon}_2$ are normal to the scattering plane but $\boldsymbol{\varepsilon}_\pm$ lies in the plane. Were the wave-packet labels interchanged, the similarity of this case to cases VI to IX would be evident.

TABLE I. Forbidden scattering processes. Processes forbidden by the polarization relations are marked with an X, by the exclusion of $\cos\theta=\pm1$ values with a Y, and by energy-momentum conservation with a Z (some are forbidden on more than one ground but only first is indicated). The quantities p_1, p_2, and $p_\pm$ are the polarizations of the waves of angular frequencies ω_1, ω_2, and $\omega_\pm=\omega_1\pm\omega_2$, respectively; l denotes longitudinal polarization, t transverse in the scattering plane, and τ transverse normal to the scattering plane.

Interaction case	Scattered wave					
	$p_+=l$	$p_+=t$	$p_+=\tau$	$p_-=l$	$p_-=t$	$p_-=\tau$
I: $p_1=l$, $p_2=l$	Y	Z	X	Y		X
II: $p_1=l$, $p_2=t$		Z	X			X
III: $p_1=t$, $p_2=l$		Z	X	Z	Z	X
IV: $p_1=t$, $p_2=t$		Y	X	Z	Y	X
V: $p_1=\tau$, $p_2=\tau$		Y	X	Z	Y	X
VI: $p_1=l$, $p_2=\tau$	X	X	Z	X	X	
VII: $p_1=\tau$, $p_2=l$	X	X	Z	X	X	Z
VIII: $p_1=\tau$, $p_2=t$	X	X	Y	X	X	Y
IX: $p_1=t$, $p_2=\tau$	X	X	Y	X	X	Y

C. Allowed Scattering Processes

The requirements of energy-momentum conservation

$$\omega_\pm=\omega_1\pm\omega_2, \tag{19}$$

$$\mathbf{k}_\pm=\mathbf{k}_1\pm\mathbf{k}_2 \tag{20}$$

must be satisfied by the scattered wave (see the Appendix). From these relations, we derive an expression for $\cos\theta$ in terms of the c's and ω_1 and ω_2

$$\cos\theta=c_\pm^{-2}\{c_1c_2\pm\tfrac{1}{2}[c_2\omega_1(c_1\omega_2)^{-1}(c_1^2-c_\pm^2)+c_1\omega_2(c_2\omega_1)^{-1}(c_2^2-c_\pm^2)]\}. \tag{21}$$

Since $-1<\cos\theta<1$, analysis of Eq. (21) for each interaction case gives conditions for the allowed scattering processes. The exclusion of $\cos\theta=\pm1$ values in the present analysis results in the following restriction: The two primary waves and the scattered wave cannot all be longitudinal or all be transverse.

Use of the polarization relations, the above restriction, and the energy-momentum conservation condition

TABLE II. Frequency ranges for allowed scattering processes. The quantities p_1, p_2, and $p_\pm$ are the polarizations of the waves of angular frequencies ω_1, ω_2, and $\omega_\pm=\omega_1\pm\omega_2$, respectively; l denotes longitudinal polarization, t transverse in the scattering plane, and τ transverse normal to the scattering plane.

Allowed process	Frequency range
$p_1=l$, $p_2=l$, $p_-=t$	$1>\omega_2/\omega_1>(c_l-c_t)/(c_l+c_t)$
$p_1=l$, $p_2=t$, $p_+=l$	$1>\omega_2/\omega_1>0$
	[or $2c_t/(c_l-c_t)>\omega_2/\omega_1>0$ if $c_l\geq3c_t$]
$p_1=l$, $p_2=t$, $p_-=l$	$2c_t/(c_l+c_t)>\omega_2/\omega_1>0$
$p_1=l$, $p_2=t$, $p_-=t$	$(c_l+c_t)/2c_l>\omega_2/\omega_1>(c_l-c_t)/2c_l$
$p_1=t$, $p_2=l$, $p_+=l$	$1>\omega_2/\omega_1>(c_l-c_t)/2c_t$
	[or process forbidden if $c_l\geq3c_t$]
$p_1=t$, $p_2=t$, $p_+=l$	$1>\omega_2/\omega_1>(c_l-c_t)/(c_l+c_t)$
$p_1=\tau$, $p_2=\tau$, $p_+=l$	$1>\omega_2/\omega_1>(c_l-c_t)/(c_l+c_t)$
$p_1=l$, $p_2=t$, $p_+=\tau$	$(c_l+c_t)/2c_l>\omega_2/\omega_1>(c_l-c_t)/2c_l$

reduces greatly the number of possible scattering processes (see Table I). The convention $\omega_1 \geqslant \omega_2$ and the material property $c_l > c_t$ are employed for these results.

The above considerations show that only eight scattering processes are allowed. These processes and the allowed frequency ranges are given in Table II.

For an allowed process with given ω_1, p_1, and ω_2, p_2, the required angle θ between the incident wave-packet propagation directions is given by Eq. (21). The angle $\varphi_\pm$ (relative to $\mathbf{n}_1$) at which the scattered wave packet emerges from the point of interaction is

$$\sin\varphi_\pm = \pm c_\pm \omega_2 [c_2(\omega_1 \pm \omega_2)]^{-1} \sin\theta. \quad (22)$$

IV. DISCUSSION

A. Maximum Amplitude and Peak Intensity of the Scattered Wave Packet

The maximum amplitude $U_\pm$ of the scattered wave packet is obtained from Eq. (8) as

$$U_\pm = \tfrac{1}{4}(\tfrac{1}{2}\pi)^{1/2}\rho_0^{-1} U_1^+ U_2^\pm (\omega_1 \pm \omega_2)^{-1} \mathbf{G}(\omega_1, \pm\omega_2, p_1, p_2)\cdot \boldsymbol{\varepsilon}_\pm \times (\Delta/\Delta_1\Delta_2)^3 (c_1^{-1}\Delta_1^2 + c_2^{-1}\Delta_2^2), \quad (23)$$

where the notational changes are self-evident and where Δ is given by Eq. (9). We use the peak intensity-maximum amplitude relation $I = \frac{1}{2}\rho_0\omega^2 e U^2$ to express the peak intensity $I_\pm$ of the scattered wave packet as

$$I_\pm = \tfrac{1}{16}\pi\rho_0^{-3} I_1^+ I_2^\pm (\omega_1\omega_2)^{-2} [\mathbf{G}(\omega_1, \pm\omega_2, p_1, p_2)\cdot \boldsymbol{\varepsilon}_\pm]^2 \times (\Delta/\Delta_1\Delta_2)^6 (c_\pm/c_1c_2)(c_1^{-1}\Delta_1^2 + c_2^{-1}\Delta_2^2)^2, \quad (24)$$

where I_1^+ and $I_2^\pm$ are the peak intensities of the ω_1 and ω_2 wave packets, respectively.

B. Order-of-Magnitude of the Interaction Effects

We estimate the order-of-magnitude of the elastic-wave interactions for aluminum with

$$\rho_0 = 2.7 \text{ g/cm}^3,$$
$$c_l = 6.4(10^5) \text{ cm/sec},$$
$$c_t = 3.0(10^5) \text{ cm/sec},$$
$$\mu = 2.4(10^{11}) \text{ D/cm}^2,$$
$$K = 8.1(10^{11}) \text{ D/cm}^2,$$

and the other constants assumed to be of the same order-of-magnitude as μ and K. We take Case I ($p_1 = l$, $p_2 = l$, and $p_- = t$) and approximate the factor $(\mathbf{G}_\text{I}\cdot\boldsymbol{\varepsilon}_-)$ roughly as

$$|\mathbf{G}_\text{I}\cdot\boldsymbol{\varepsilon}_-| \sim \tfrac{1}{4}\omega_1\omega_2(\omega_1 - \omega_2)K/c_l^3.$$

The maximum amplitude is then

$$U_- \sim U_1^+ U_2^- \omega_1\omega_2 K T_1 T_2 (T_1^2 + T_2^2)/60\rho_0 c_l^3 T^3 \sim 2(10^{-7})(\text{sec/cm}) U_1^+ U_2^- \omega_1\omega_2 T_1 T_2 (T_1^2 + T_2^2)/T^3, \quad (25)$$

where the T's are pulse (time) lengths, $T_i = 2\pi/c\Delta_i$. For frequencies of the order of 10 Mc/sec and pulse lengths about 10 μsec, the above is about one order-of-magnitude less than the estimate obtained by Jones and Kobett,[5] i.e., for U_1^+, $U_2^- \sim 10^{-10}$ cm, we find $U_- \sim 2(10^{-16})$ cm.

The intensity estimate corresponding to the above is

$$I_- \sim 5(10^{-3}) I_1^+ I_2^- (\omega_1 - \omega_2)^2 K^2 \times [T_1T_2(T_1^2 + T_2^2)/T^3]^2 (\rho_0^3 c_l^2 c_t^5)^{-1} \sim 2(10^{-19})(\text{sec cm}^2/\text{erg}) I_1^+ I_2^- (\omega_1 - \omega_2)^2 \times [T_1T_2(T_1^2 + T_2^2)/T^3]^2; \quad (26)$$

for the same frequency and pulse length magnitudes as before, I_- is of the order of $(10^{-6}\text{ cm}^2/\text{W})\, I_1^+ I_2^-$.

C. Comparison with Experiment

Of the few available experiments, only those of Rollins[1,9] are suitable for comparison with the present calculation. Rollins reports[9] a measurement of the peak intensity of the scattered wave in a magnesium sample. The experimental results were the following: Two transversely polarized wave packets (corresponding to case IV or V) of roughly half-sine shape, of 6-μsec pulse length, of mean frequency 5 Mc/sec, and of peak intensity about 1 W/cm^2 gave a scattered wave of peak intensity roughly $6(10^{-7})$ W/cm^2. Our estimate for this result is about 10^{-6} W/cm^2, quite good agreement. Further, the result from Eq. (26) for aluminum agrees with an earlier measurement of Rollins[1] although the datum was qualified as not being very accurate.

The experiment of Shiren[4] is of great interest although it cannot be analyzed directly in the present theory. Shiren employed 9 Gc/sec pulses propagating colinearly in magnesium oxide. With peak intensities $\sim\frac{1}{2}$ W/cm^2, the "signal" pulse length $\frac{1}{2}$ μsec and the "pump" pulse long enough to bracket the signal pulse, Shiren found as much as 70% of the power in the signal pulse removed by the interaction. These "strong" effects might be expected to lie outside the region of validity of the present approximations; such is not necessarily so. In effect, the two pulses interact "many times," actually continually as they propagate together, so that large total effects are reasonable although the interaction itself may still be weak enough to treat in the present approximation. The experimental details reported are not sufficiently complete to warrant further discussion here.

D. Possible Application to the Determination of Third-Order Elastic Constants

Although the initial plenitude of 54 processes reduced to a mere eight, the eight are sufficient, in principle, for determination of the third-order elastic constants A, B, and C. Study of the relevant $\mathbf{G}$ vectors shows that determination of ten different linear combinations of μ, K, A, B, and C should be possible; these

[9] F. R. Rollins, Jr. (private communication).

would be much more than sufficient to determine the elastic constants.

In general, measurements of third-order elastic constants are quite difficult and the results not very accurate.[10] Measurement of these constants by means of the elastic-wave scattering phenomena would not be easy but probably would be somewhat less difficult than and as accurate, at least, as any presently available method. An elastic-wave scattering method would involve measurements of scattered wave amplitudes (intensities) for various frequencies, polarizations, and relative propagation directions of the input waves. The major experimental difficulty anticipated would lie in the bonding of ultrasonic transducers to samples; bonds would be required with characteristics sufficiently uniform as to allow reproducible measurements. Absolute measurements would not be required since A, B, and C could be obtained to good (relative) accuracy in terms of μ and K and μ and K are readily measurable by other techniques. Even if bonds of suitable uniformity were not obtainable, this difficulty might be circumvented, at least in part, by experimental designs using correlations between a set of transducers.

All in all, these considerations suggest that elastic-wave interaction phenomena may find useful and, perhaps, valuable applications in the measurement of elastic constants and in studies of lattice dynamics.

V. CONCLUSION

The present study agrees in essentials with the results of Jones and Kobett[5] relative to the allowed processes and their frequency ranges. Several discrepancies in their Table I are corrected in our Table II. Our maximum scattered-wave amplitude and theirs differ so greatly in form as to make a detailed comparison impossible. However, one evident and important difference is that our amplitude is proportional to frequency squared (distinction between the different frequencies neglected) but theirs is proportional to frequency cubed. This particular disagreement would be resolved by the inclusion of beam width considerations in the Jones and Kobett analysis.

The present results apply only to maximum amplitudes or peak intensities and widths of experimental wave packets. Our use of Gaussian shape factors does not allow more detailed considerations of pulse shapes.

Extension of this work to anisotropic solids is straightforward but tediously complex. The effort necessary for this task may be warranted if and when elastic-wave interaction measurements prove useful.

APPENDIX

The first step in integrating Eq. (4) is the integration over $\omega_\pm'$ from 0 to $+\infty$. This is done in the complex plane with the contour being closed in the negative half-plane and only the principal value from the pole at $\omega_\pm'=\omega_\pm(k_\pm')$ contributing. The $\mathbf{k}_\pm'$ integration is then carried out by means of the three-dimensional delta function $\delta^3(\mathbf{k}_1'\pm\mathbf{k}_2'-\mathbf{k}_\pm')$. The result after these steps is

$$u_\pm(\mathbf{x},t)=\frac{\pi A_1{}^+A_2{}^\pm}{(2\pi)^3\rho_0}\times\int\frac{d^3k_1'd^3k_2'\delta[\omega_1(k_1')\pm\omega_2(k_2')-\omega_\pm(k_\pm')]}{2\omega_\pm(k_\pm')}\times\mathbf{G}(\mathbf{k}_1',\pm\mathbf{k}_2',\boldsymbol{\varepsilon}_1,\boldsymbol{\varepsilon}_2)\cdot\boldsymbol{\varepsilon}_\pm\times\exp\left[-\frac{(\mathbf{k}_1'-\mathbf{k}_1)^2}{2\Delta_1{}^2}-\frac{(\mathbf{k}_2'-\mathbf{k}_2)^2}{2\Delta_2{}^2}\right]\times\exp\{i[(\mathbf{k}_1'\pm\mathbf{k}_2')\cdot\mathbf{x}-\omega_\pm(k_\pm')t]\},\quad\text{(A1)}$$

where $k_\pm'=|\mathbf{k}_1'\pm\mathbf{k}_2'|$.

Either one of the remaining integrations may be done next; we chose the $\mathbf{k}_2'$ one. The slowly varying parts are taken out of the integral and the integration done in spherical coordinates, the angular parts first. The Gaussian is all that must be considered with $(\mathbf{k}_2'-\mathbf{k}_2)^2$ written as

$$(\mathbf{k}_2'-\mathbf{k}_2)^2=k_2'^2+k_2^2-2k_2'k_2\cos\kappa\,,\quad\cos\kappa=\mathbf{k}_2'\cdot\mathbf{k}_2/k_2'k_2;$$

the relevant integral is

$$\int d\Omega_2'\exp[k_2'k_2\cos\kappa/\Delta_2{}^2]=\frac{2\pi\Delta_2{}^2}{k_2'k_2}\times\left\{\exp\left[\frac{k_2'k_2}{\Delta_2{}^2}\right]-\exp\left[-\frac{k_2'k_2}{\Delta_2{}^2}\right]\right\}.\quad\text{(A2)}$$

We use the remaining delta function for the magnitude k_2' integration. This requires

$$\omega_\pm(k_\pm')=\omega_1(k_1')\pm\omega_2(k_2')\quad\text{(A3)}$$

or

$$c_\pm k_\pm'=c_1k_1'\pm c_2k_2'\quad\text{(A3')}$$

since

$$\omega_i(k_i)=c_ik_i.$$

An earlier result, the integration with $\delta^3(\mathbf{k}_1'\pm\mathbf{k}_2'-\mathbf{k}_\pm')$ is

$$\mathbf{k}_\pm'=\mathbf{k}_1'\pm\mathbf{k}_2'.\quad\text{(A4)}$$

Simultaneous solution of Eqs. (A3′) and (A4) requires

$$k_2'=\eta_\pm k_1',\quad\text{(A5)}$$

where $\eta_\pm$ is a real, positive quantity. After these

[10] A new optical-acoustic technique suggested by J. Melngailis, A. A. Maradudin, and A. Seeger, Phys. Rev. **131**, 1972 (1963), promises a convenient and accurate method for the measurement of elastic constants in transparent materials; thus, these comments do not necessarily apply for such materials.

manipulations, Eq. (A1) becomes

$$u_\pm(\mathbf{x},t)=\frac{A_1{}^+A_2{}^\pm\Delta_2{}^2\mathbf{G}(\mathbf{k}_1,\pm\mathbf{k}_2,\boldsymbol{\varepsilon}_1,\boldsymbol{\varepsilon}_2)\cdot\boldsymbol{\varepsilon}_\pm}{8\pi\rho_0c_2[\omega_1(k_1)\pm\omega_2(k_2)]}\exp[i\{(\mathbf{k}_1\pm\mathbf{k}_2)\cdot\mathbf{x}-[\omega_1(k_1)\pm\omega_2(k_2)]t\}]$$
$$\times\int d^3k_1{}'\exp\left[-\frac{(\mathbf{k}_1{}'-\mathbf{k}_1)^2}{2\Delta_1{}^2}\right]\exp[i(\mathbf{k}_1{}'-\mathbf{k}_1)\cdot\mathbf{x}]\left\{\exp\left[-\eta_\pm{}^2\frac{(k_1{}'-k_1)^2}{2\Delta_2{}^2}\right]-\exp\left[-\eta_\pm{}^2\frac{(k_1{}'+k_1)^2}{2\Delta_2{}^2}\right]\right\}. \quad \text{(A6)}$$

Since $k_1>0$ and $k_1{}'\geq 0$ with the main contribution at $k_1{}'\sim k_1$, the last exponential quantity above is always small and, therefore, is neglected.

The calculation is now forced in a credible fashion to the expected result. The last (remaining) exponential in Eq. (A6) is the source of difficulty. This is written as:

$$\exp[-\eta_\pm{}^2(k_1{}'-k_1)^2/2\Delta_2{}^2]$$
$$=\exp\left[-\eta_\pm{}^2\frac{(\mathbf{k}_1{}'-\mathbf{k}_1)^2}{2\Delta_2{}^2}\right]\exp\left[-\eta_\pm{}^2\frac{k_1{}'k_1(\cos\Psi-1)}{\Delta_2{}^2}\right],$$

where

$$\cos\Psi=\mathbf{k}_1{}'\cdot\mathbf{k}_1/k_1{}'k_1.$$

With the definition

$$\boldsymbol{\kappa}=k_1{}'-k_1,$$

$\Psi_{\max}$ for a given $\kappa\ll k_1$ is obtained for $k_1{}'=k_1$ at

$$\sin\tfrac{1}{2}\Psi_{\max}=\kappa/2k_1$$

or

$$\cos\Psi_{\max}\simeq 1-\kappa^2/2k_1{}^2.$$

Thus, the magnitude of the difficult term is

$$1\leq\exp[-\eta_\pm{}^2k_1{}'k_1(\cos\Psi-1)/\Delta_2{}^2]\leq\exp[\eta_\pm{}^2\kappa^2/2\Delta_2{}^2].$$

The final integral part of Eq. (A6) is now approximately

$$\int d^3\boldsymbol{\kappa}\exp[-\boldsymbol{\kappa}^2/2\Delta^2]\exp[i\boldsymbol{\kappa}\cdot\mathbf{x}]=(2\pi)^{3/2}\Delta^3\exp[-\tfrac{1}{2}\mathbf{x}^2\Delta^2],$$

where

$$\Delta_1{}^{-2}\leq\Delta^{-2}\leq\Delta_1{}^{-2}+\eta_\pm{}^2\Delta_2{}^{-2}. \quad \text{(A7)}$$

Clearly the exact result, were it calculable, would be almost Gaussian for $\Delta^2\ll k_1{}^2$.

The approximate result is

$$u_\pm(\mathbf{x},t)=\frac{A_1{}^+A_2{}^\pm\mathbf{G}(\mathbf{k}_1,\pm\mathbf{k}_2,\boldsymbol{\varepsilon}_1,\boldsymbol{\varepsilon}_2)\cdot\boldsymbol{\varepsilon}_\pm}{4(2/\pi)^{1/2}\rho_0[\omega_1(k_1)\pm\omega_2(k_2)]}$$
$$\times\Delta^3(c_1{}^{-1}\Delta_1{}^2+c_2{}^{-1}\Delta_2{}^2)\exp[-\tfrac{1}{2}\mathbf{x}^2\Delta^2]$$
$$\times\exp[i\{(\mathbf{k}_1\pm\mathbf{k}_2)\cdot\mathbf{x}-[\omega_1(k_1)\pm\omega_2(k_2)]t\}], \quad \text{(A8)}$$

where the above expression has been made symmetric with respect to c_1, Δ_1, and c_2, Δ_2 by replacement of $c_2{}^{-1}\Delta_2{}^2$ by

$$\tfrac{1}{2}(c_1{}^{-1}\Delta_1{}^2+c_2{}^{-1}\Delta_2{}^2).$$

The k-space width Δ of the scattered wave packet is calculated indirectly as follows: The width Δ is determined by the length of time $T=2\pi/c_\pm\Delta$ that the ω_1 and ω_2 wave packets interact. Since the characteristic dimensions of these wave packets are $2\pi/\Delta_1$ and $2\pi/\Delta_2$, respectively, a simple geometric analysis gives

$$\Delta^2=\frac{(c_1{}^2-2c_1c_2\cos\theta+c_2{}^2)\Delta_1{}^2\Delta_2{}^2}{c_\pm{}^2\{1-(1/c_1c_2)[(c_2{}^2\omega_1\pm c_1{}^2\omega_2)/(\omega_1\pm\omega_2)]\cos\theta\}^2(\Delta_1+\Delta_2)^2}, \quad \text{(9)}$$
$$\cos\theta\neq\pm1.$$

The calculation Eq. (9) is not valid for angles nearly equal to 0° or 180°.

Since the wave vectors and frequencies satisfy Eqs. (A3) and (A4), the mean wave vectors and mean frequencies of the wave packets must likewise satisfy

$$\omega_\pm(k_\pm)=\omega_1(k_1)\pm\omega_2(k_2), \quad \text{(19)}$$
$$\mathbf{k}_\pm=\mathbf{k}_1\pm\mathbf{k}_2, \quad \text{(20)}$$

i.e., the energy-momentum conservation condition.