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Physics and Engineering Principles of Nonlinear Acoustics

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The nonlinear differential equation describing propagation of finite amplitude ultrasonic waves in gases, liquids, and solids is shown to have the same mathematical form. The nonlinearity parameter in each case is defined in terms of fundamentally significant physical quantities. Waveform distortion is described, and the discontinuity distances are compared. Recent results are given for gases, liquids, and solids and their significance is evaluated in terms of modern condensed matter physics and engineering.

§1. Introduction

Over the past two decades it has become apparent not only that natural phenomena are distinctly nonlinear, but also that such nonlinearity can be described in a consistent way. No longer is it necessary to abandon a measurement or an investigation at the point that the amplitude becomes great enough that nonlinear behavior is observed. This statement is valid for essentially all branches of physics, but it is especially true of investigations in which acoustical phenomana are used. The subject of nonlinear acoustics now can be understood to the point that it is being used in such diverse fields as hardness testing of steels,[1] improvement of resolution in acoustical microscopy,[2] and the investigation of the relationship between lattice anharmonicity and bulk nonlinearity in crystalline solids.[3] Such a wide range of investigations more and more requires knowledge about the general features of nonlinear physics in general, and nonlinear acoustics in particular. The purpose of this study is to identify some nonlinear phenomena and to show that the nonlinear behavior of different media—gases, liquids, and solids—is similar, although the acoustical nonlinearities have different thermodynamic origins. The approach is to show that the same nonlinear differential equation describes gases, liquids, and solids, then to identify coefficients. The different coefficients allow one to define the origin of the nonlinearity in each case. A perturbation analysis becomes the most useful approach, as will become apparent.

§2. Nonlinear Fluids

2.1 *Equation of state*

The same treatment can be used to derive the nonlinear wave equation in gases and liquids. The difference in the theory appears in the form of the equation of state. Let us write the equation of state in the generally applicable form of a Taylor expansion of the pressure in terms of the condensation (the fractional change in density):

$$P-P_0=A\left(\frac{\rho-\rho_0}{\rho_0}\right)+\frac{B}{2!}\left(\frac{\rho-\rho_0}{\rho_0}\right)^2+\cdots \tag{1}$$

In this form the coefficients A and B are defined by:

$$A=\rho_0\left(\frac{\partial P}{\partial \rho}\right)_{\rho=\rho_0}$$

$$B=\rho_0^2\left(\frac{\partial^2 P}{\partial \rho^2}\right)_{\rho=\rho_0}. \tag{2}$$

Since the propagation of a sound wave involves perturbations of the thermodynamic variables about equilibrium values, the form of Eq. (1) is especially useful for describing sound propagation in general, and nonlinear acoustical phenomena in particular. It is for this reason that the ratio B/A has become an essential parameter in nonlinear acoustics, and its evaluation a prime objective of many research efforts.

Although Eq. (1) is applied in this form to the description of the nonlinear behavior of fluids, this has not always been the case. In fact, many of the nonlinear acoustical phenomena were first described in terms of ideal gases. The equation of state for an isentropic process in an ideal gas usually is written

$$P=P_0\left(\frac{\rho}{\rho_0}\right)^\gamma \tag{3}$$

where $\gamma=C_P/C_V$ is the ratio of specific heats. Substituting Eq. (3) into Eq. (1) leads to the conclusion that for an ideal gas the value of

$$B/A=\gamma-1. \tag{4}$$

Thus, one now has a connection between the nonlinear behavior of liquids and ideal gases through the equation of state.

2.2 *Finite amplitude waves in fluids*

The recent increase in interest in nonlinear acoustics has been brought about by the increase in the number of practical occurrences and applications of waves of finite amplitude and the increasing possibilities for the evaluation of numerical solutions to complicated nonlinear equations. One can derive a nonlinear differential equation capable of describing a number of nonlinear acoustical phenomena by applying a perturbation analysis to the equation of state, the equation of motion, and the equation of continuity, and keeping second order terms in the dependent variables. Assuming an isentropic

process and combining the equation of motion and the equation of continuity with the equation of state, Eq. (1), one obtains an equation in particle displacement ξ:

$$\frac{\partial^2\xi}{\partial t^2}=C_0^2\left(1-B/A+\frac{B/A}{1+\dfrac{\partial\xi}{\partial a}}\right)\frac{\dfrac{\partial^2\xi}{\partial a^2}}{\left(1+\dfrac{\partial\xi}{\partial a}\right)^2} \tag{5}$$

where a is the space coordinate measured along the propagation direction and $C_0^2=A/\rho_0$. Expanding the function $(1+\partial\xi/\partial a)^{-1}$ into a power series one obtains

$$\frac{\partial^2\xi}{\partial t^2}=C_0^2\left(1-B/A\,\frac{\partial\xi}{\partial a}+\cdots\right)\frac{\dfrac{\partial^2\xi}{\partial a^2}}{\left(1+\dfrac{\partial\xi}{\partial a}\right)^2}. \tag{6}$$

The bracketed term is the power series expansion of the function $[1+(\partial\xi/\partial a)]^{-B/A}$. Thus, the nonlinear equation can be written in the familiar form

$$\frac{\partial^2\xi}{\partial a^2}=C_0^2\frac{\dfrac{\partial^2\xi}{\partial a^2}}{\left(1+\dfrac{\partial\xi}{\partial a}\right)^{B/A+2}}.$$

This equation describes the propagation of a finite amplitude wave in a fluid, and with the identification of B/A given in Eq. (4), it also describes an ideal gas. For comparison with the equation describing solid state nonlinearity it can be expanded in the form

$$\frac{\partial^2\xi}{\partial t^2}=C_0^2\left(\frac{\partial^2\xi}{\partial a^2}-(B/A+2)\frac{\partial\xi}{\partial a}\frac{\partial^2\xi}{\partial a^2}+\cdots\right) \tag{8}$$

§3. Nonlinear Solids

The derivation of the nonlinear wave equation to describe finite amplitude wave propagation in solids proceeds along different directions from that used for fluids, although for special circumstances the resulting differential equation has the same form, with a set of slightly more complicated coefficients. For many situations, then, the same mathematical description of the propagation of finite amplitude waves is appropriate for gases, liquids, and solids. These are the phenomena we will focus our attention on.

The derivation begins with a definition of the elastic constants in terms of an expansion of the elastic strain energy $\phi(\eta)$ in terms of the strains η_{ij}

$$\phi(\eta)=\frac{1}{2!}\sum_{ijkl=1}^{3}C_{ijkl}\eta_{ij}\eta_{kl}+\frac{1}{3!}\sum_{ijklmn=1}^{3}C_{ijklmn}\eta_{ij}\eta_{kl}\eta_{mn}. \tag{9}$$

The C_{ijkl} are the ordinary elastic constants. For linear elasticity theory they are the only constants defined. Since we are concerned with nonlinear effects that can be explained only by introducing higher order terms, we will call the C_{ijkl} the second order elastic (SOE) constants since they are coefficients of the second powers in strains. The C_{ijklmn}, then, are the third order elastic (TOE) constants.

To derive the nonlinear differential equation for describing finite amplitude waves in a solid, Lagrange's equations for continuous media are used. The derivation requires only the definition of the strain and the assumption that the elastic energy is a function of the strain alone, which means that the solid is assumed to be a lossless continuum in which attenuation and dispersion are negligible.

To orient the a_1 axis along the direction of propagation one must use a rotation matrix to transform the strain matrix because the elastic constants defined by Eq. (9) are defined with respect to the crystalline symmetry axes. By choosing to write the displacements ξ_i parallel (or perpendicular to) the propagation direction, one can use the Lagrangian procedure followed by rotation operations appropriate to the crystalline symmetry and write the nonlinear wave equation in the form

$$\rho\frac{\partial^2\xi_i}{\partial t^2}=\sum_{j=1}^{3}A_{ij}\frac{\partial^2\xi_j}{\partial a_1^2}+\sum_{j,l=1}^{3}B_{ijl}\frac{\partial\xi_j}{\partial a_1}\frac{\partial^2\xi_l}{\partial a_1^2}, \tag{10}$$

where the A_{ij} are known linear combinations of SOE constants and the B_{jjl} are known linear combinations of third order elastic constants. Although the mathematical form of Eq. (10) already can be recognized as being the same as that in Eq. (8), the comparison can be facilitated by specializing Eq. (10) to cubic symmetry.

If one considers wave propagation along the pure mode directions in a cubic crystal, then one finds that pure longitudinal waves propagating along the principal directions [100], [110], and [111] are described by a nonlinear wave equation of the form

$$\frac{\partial^2\xi}{\partial t^2}=C_0^2\left(\frac{\partial^2\xi}{\partial a^2}+\frac{3K_2+K_3}{K_2}\frac{\partial\xi}{\partial a}\frac{\partial^2\xi}{\partial a^2}+\cdots\right), \tag{11}$$

where $C_0^2=K_2/\rho_0$, and K_2 is a linear combination of SOE constants and K_3 is a linear combination of TOE constants as given in Table 1. Written in this form it is apparent that the mathematical form of Eqs. (11) and (8) is the same. Thus, longitudinal waves propagating along pure mode directions in crystals (or in isotropic solids) exhibit nonlinear effects which are similar to those exhibited by liquids and gases. To emphasize the similarity of the behavior of finite amplitude waves in gases, liquids, and solids, we will define the ratio of the coefficients of the nonlinear term to the linear term in Eqs. (8) and (11) as the nonlinearity parameter β and list it along with the square of the speed of a small amplitude wave C_0^2 in Table 2. These two quantities are fundamental to nonlinear acoustics. Using them one can write Eqs. (8) and (11) in the final form

Table I. K_2 and K_3 for [100], [110], and [111] directions.

Direction	K_2	K_3
[100]	C_{11}	C_{111}
[110]	$\frac{1}{2}[C_{11}+C_{12}+2C_{44}]$	$\frac{1}{4}[C_{111}+3C_{112}+12C_{166}]$
[111]	$\frac{1}{3}[C_{11}+2C_{12}+4C_{44}]$	$\frac{1}{9}[C_{111}+6C_{112}+12C_{144}+24C_{166}+2C_{123}+16C_{456}]$

Table II. Parameters entering into the description of wave propagation in nonlinear gases, liquids, and solids.

Parameter	Ideal Gas	Liquid	Solid
C_0^2	$\gamma P_0/\rho_0$	A/ρ_0	K_2/ρ_0
β	$\gamma+1$	$B/A+2$	$-\left(\frac{K_3}{K_2}+3\right)$

$$\frac{\partial^2\xi}{\partial t^2}=C_0^2\left(\frac{\partial^2\xi}{\partial a^2}-\beta\frac{\partial\xi}{\partial a}\frac{\partial^2\xi}{\partial a^2}+\cdots\right) \tag{12}$$

which can be applied to gases, liquids, and solids by using Table 2. Generally speaking, the magnitudes of β are such that nonlinear effects are noticeable at lower frequencies in gases, then become important at higher frequencies in liquids and solids.

§4. Solution of Nonlinear Equation

The nonlinear equation can be solved under the assumption of a sinusoidal driver at $a=0$. An exact solution for the particle velocity $u=\partial\xi/\partial t$ is

$$u=2\delta\sum_{n=1}^{\infty}\frac{J_2(na/L)}{na/L}\sin n(\omega t-ka), \tag{13}$$

where ω and k are the frequency and propagation constant of the wave, δ is an arbitrary constant, and J_n is the nth order Bessel function. The quantity L is the discontinuity distance, a parameter of considerable significance to nonlinear acoustics.

By making a power series expansion of the solution, one can obtain an expression for the particle displacement ξ as a function of the distance a from a sinusoidal driver in the form

$$\xi=A\sin(ka-\omega t)+\beta\frac{A^2k^2a}{8}\cos 2(ka-\omega t)+\cdots \tag{14}$$

which is especially useful for the study of solids. Equation (14) shows that a second harmonic is generated whose amplitude is proportional to the nonlinearity parameter β. Knowing the proportionality constant and measuring the amplitudes of the fundamental and second harmonic allows one to evaluate the nonlinearity parameter for pure mode directions in solids.

4.1 *Waveform distortion*

The growth of the second harmonic (and higher harmonics) in Eq. (14) leads one to expect a waveform distortion to occur as the wave propagates. Such waveform distortion also can be predicted from the phase velocity

$$C_{ph}=C_0\left(1+\frac{3u}{2C_0}\beta\right)^{1/3} \tag{15}$$

which depends on the particle velocity u and the nonlinearity parameter β. Because of the variation, the waveform distorts as indicated in Fig. 1.

4.2 *Discontinuity distance*

The waveform distortion shown in Fig. 1 occurs because the phase velocity is a function of the particle velocity u (Eq. (15)); however, this situation can exist only

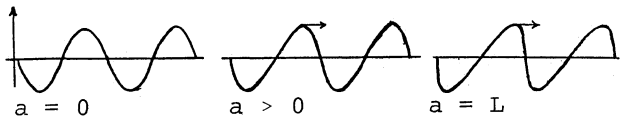

Fig. 1. Progressive waveform distortion in a finite amplitude ultrasonic wave.

Table III. Discontinuity distances for common substances.

Substance	Discontinuity Distance L	L in Meters if $\xi_0=1$ Å at 30 MHz
Air	$\frac{7.2\times10^4}{\omega^2\xi_0}$ (m²/sec²)	0.02
Water (30°C)	$\frac{6.4\times10^5}{\omega^2\xi_0}$	0.2
Copper [100]	$\frac{2\times10^7}{\omega^2\xi_0}$	5.6
Germanium [100]	$\frac{1.9\times10^7}{\omega^2\xi_0}$	5.3
Fused Silica	$-\frac{2.9\times10^6}{\omega^2\xi_0}$	−0.8

for distances smaller than the distance required for the leading edge of the waveform to approach a vertical tangent. This propagation distance is called the discontinuity distance L:

$$L=\frac{2C_0^2}{\beta\omega u_0} \tag{16}$$

where u_0 is the particle velocity at $a=0$. A more useful form for present purposes is

$$L=\frac{2C_0^2}{\beta\omega 2\xi_0} \tag{17}$$

where ξ_0 is the particle displacement amplitude at $a=0$.

An impression of relative magnitudes can be obtained if one uses Eq. (17) and the definitions of Table 2 to evaluate the discontinuity distance for some known substances. The results are shown in Table 3. In the second column the discontinuity distance is expressed as a number divided by $\omega^2\xi_0$. These numbers would be quite realistic if one used frequencies ω and amplitudes characteristic of experiments in the various substances. For example, audio frequencies would be appropriate for air. However, a comparison of the relative nonlinearity can best be obtained by using the same magnitudes for each substance. For this reason we have assumed an amplitude of 1 Å at 30 MHz for all substances listed, even though attenuation in air would make an actual experiment at this frequency very difficult. The values 1 Å and 30 MHz actually are appropriate for investigations in solids. The magnitudes of the discontinuity distances given in Column 3 give a good impression of the fact that gases and liquids are much more nonlinear than solids. The negative magnitude of the discontinuity distance for fused silica results from its inherently negative nonlinearity parameter and implies that fused silica is one of the few materials that can sustain a rarefaction shock as the discontinuity distance is approached.

§5. Recent Results

Nonlinear acoustics has made possible the measurement of physical quantities that heretofore had not been measured. At present scientists are correlating physical behavior with these measured quantities. The coefficients A and B in the adiabatic equation of state for fluids

$$P-P_0=A\left(\frac{\rho-\rho_0}{\rho_0}\right)+\frac{B}{2}\left(\frac{\rho-\rho_0}{\rho_0}\right)^2+\cdots$$

now are available for a significant number of fluids. A typical value of the ratio B/A for diatomic gases is 0.4. This is considerably smaller than that for liquids. Values of B/A, or the nonlinearity parameter $B/A+2$, are measures of the relative nonlinearity of fluids. As shown in Table 4, B/A for water changes from 4.16 to 6.11 as the temperature increases from 0°C to 100°C at atmospheric pressure.[4] As water is one of the principal constituents of biological tissue B/A values for biological tissues are being evaluated and compared with that for water. For example, the B/A values for cat liver are between 6.5 and 7.0, both *in vivo* and *in vitro*, so it is implied that blood circulation in liver has little influence upon its B/A value.[5]

Table IV. Values of B/A for various fluids at atmospheric pressure.

Liquid	Temperature °C	B/A
Distilled water	0	4.16
	10	4.63
	20	4.96
	30	5.22
	40	5.38
	50	5.55
	60	5.67
	80	5.96
	100	6.11
Acetone	30	9.44
Benzene	30	9.03
Benzyl alcohol	30	10.19
Carbon tetrachloride	30	11.54
Carbon disulphide	10	6.4
	25	6.2
	40	6.1
Chlorobenzene	30	9.33
Cyclohexane	30	10.07
Diethylamine	30	10.30
Glycerine	20	8.80
Mercury	40.5	8.33
Methyl acetate	30	9.66
Nitrobenzene	30	9.88
Methyl alcohol	30	9.62
Ethyl alcohol	30	10.57
Propyl alcohol	30	10.70
Butyl alcohol	30	10.72
Toluene	30	8.30
Heptane	40	11.14
Octane	40	11.34
Nonane	40	11.27
Dodecane	40	11.33
Hexadecane	40	11.40
Liquid nitrogen	−199	9.69
Liquid oxygen	−199	9.56
Liquid methane	−158	10.27
Monatomic gas	20	0.67
Diatomic gas	20	0.40

Values of the nonlinearity parameters of a number of solids are now available. As indicated in Fig. 2, the nonlinearity parameters of solids, especially for the [100] direction, have proved to be virtually independent of temperature. This fact is very significant to improving our understanding of the relationship between the nonlinearity of a solid measured by acoustical techniques and anharmonicity of the interatomic potential function determined from thermal expansion, neutron scattering, or other techniques.

Nonlinearity parameters of solids have been correlated with their interatomic bonding with considerable success. For example, in Table 5 are listed solids having cubic symmetry.[6] By examining a number of examples in each bonding category it has become apparent that covalent bonding produces solids with small nonlinearity parameters. Thus solids with zincblende (diamond lattice) structure such as silicon, germanium, etc. are found to have the smallest nonlinearity parameters. The largest nonlinearity parameters to date are found among the solids with ionic bonding of the NaCl type. In NaCl one also finds a large thermal expansion coefficient at room temperature. It has not yet been firmly established that a large thermal expansion coefficient always accompanies a large nonlinearity parameter.

Finally, technologically important correlations are be-

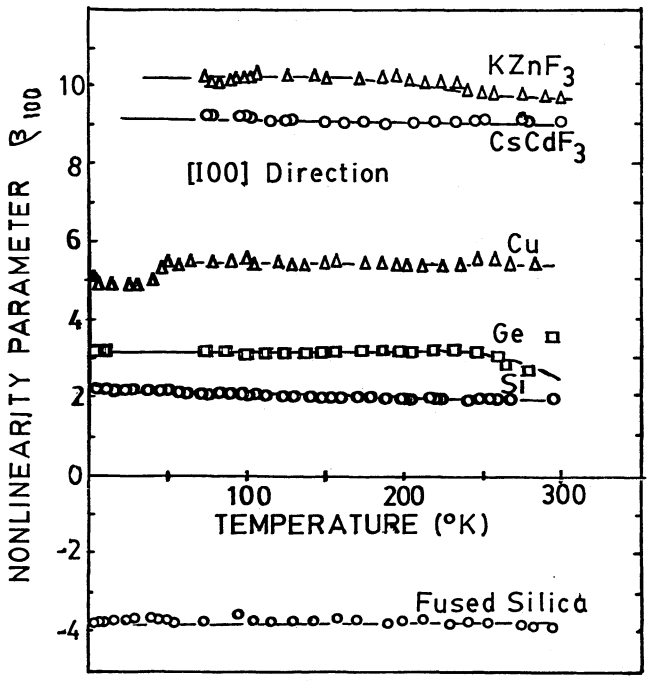

Fig. 2. Measured temperature dependence of nonlinearity parameters.

Table V. Comparison of structure, bonding, and nonlinearity.

Structure	Bonding	β_{av}	Range of β
NaCl	Ionic	14.6	14.0-15.4
BCC	Metallic	8.2	7.4- 8.8
FCC	van der Waals	6.4	5.8- 7.0
FCC	Metallic	5.8	4.0- 7.0
Fluorite	Ionic	3.8	3.4- 4.6
Zincblende	Covalent	2.2	1.8- 3.0

ing made between the nonlinearity parameters of solids as well as fluids. The most recent one is the correlation of nonlinearity parameter magnitude with hardness in steels.[1] Such correlations are defining the value of the new nonlinearity parameter. They also are helping to define the ultimate role to be played by nonlinear acoustics in science and technology.

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