OFFICE OF NAVAL RESEARCH
CONTRACT NO. N00014-81-K-0229
TASK NO. 4126-306
Technical Report No. 27 (Final)
September 1983 to June 1987
NONLINEAR ACOUSTICS
M. A. Breazeale
Principal Investigator
August 1987

UNIVERSITY OF TENNESSEE

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Technical Report No. 27 (Final), NONLINEAR ACOUSTICS (Unclassified)

This is the final report summarizing results obtained during more than 23 years of research on Nonlinear Acoustics. The report contains a general overview of research performed and a general summary statement. Lists of publications, lectures, and technical reports are provided, as well as a list of personnel.
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I. INTRODUCTION

This is the final report on Contract No. N00014-81-K-0229 entitled "Nonlinear Acoustics" covering the period December 1, 1980, to June 30, 1987. As the research performed under this contract is a continuation of the program supported under other contracts, a summary of the entire program is given for completeness. The series of contracts in question is summarized as follows:

<table>
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<th>Contract No.</th>
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<tr>
<td>Nonr 4289(01)</td>
<td>1 September 1963 to 31 August 1970</td>
<td>$140,921</td>
</tr>
<tr>
<td>N00014-71-A-0121</td>
<td>1 September 1970 to 31 August 1975</td>
<td>$127,342</td>
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<tr>
<td>N00014-76-C-0177</td>
<td>1 September 1975 to 30 November 1980</td>
<td>$280,733</td>
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<tr>
<td>N00014-81-K-0229</td>
<td>1 December 1980 to 30 June 1987</td>
<td>$342,000</td>
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Throughout the 24 years one main objective was to define conditions under which nonlinear behavior of the propagating medium would be in evidence and to use quantitative evaluation of this nonlinearity to characterize the medium. This approach has taken us into the investigation of many phenomena. The description of some is adequate if one uses only the linear form of the equations. In some instances the nonlinearity was observable but could be ignored in the first approximation. In other instances the existence of nonlinearity forms the basis of the entire investigation.

Throughout the investigations the emphasis has been on the observation and elucidation of physical phenomena, especially nonlinear phenomena, of interest to Naval programs. Attention has been paid to contributions to understanding of fundamental physics, and in some
instances results of very practical significance have been obtained. Whether basic physics or applied physics, the results were considered to be important. A by-product of the research program has been a sizeable number of students with some knowledge of physical acoustics, many of whom received advanced degrees with support from the above contracts. A list of students and a complete bibliography of publications are included in this Final Report.
II. AN OVERVIEW OF RESEARCH PERFORMED

In the course of the ONR Program we issued summary reports to cover special topics on which considerable research was done. These summaries are to be found in Technical Reports Nos. 8, 15, 17, and 18. For convenience of the reader we reproduce prefaces from these reports below to explain the relationship among the different investigations and to provide reference information for more detailed analyses of the physical phenomena discussed in the reports themselves.

Preface to Technical Report No. 8

This is the Final Technical Report under Contract NONR 4289(01). This summary report covers essentially four topics not covered by previous technical reports: (1) In "Simplification of Apparatus to Diffract Light by Ultrasonic Waves," we point out that the laser is ideally suited to such measurements because the laser beam is small, intense, and collimated, each of which simplifies measurements. (2) The next three chapters deal with various aspects of ultrasonic parametric oscillations, including the nonlinear equation which probably describes the situation. The use of parametric oscillations to make a variable frequency transducer has not been practical. On the other hand, measurement of the threshold of parametric oscillation does offer promise of a means of measuring relative attenuation. (3) Diffraction lobes from the beam pattern of transducers can be eliminated quite simply in certain cases. This technique is described in Chapter V. The analogous technique in optics is referred to as "apodization." Therefore, it would be correct to refer to this ultrasonic technique as "apodization of transducers." The final chapter
gives a graphical technique for determining the electrical field inside the apodized transducers. (4) The final chapter shows the way the apodized transducers can be used to make a detailed study of the energy redistribution in an ultrasonic beam reflected from a liquid-solid interface at the angle at which surface waves are generated.

Previous Technical Reports under Contract 4289(01) have been:


Many of these Technical Reports contain M.S. theses or doctoral dissertations. Master's degree students receiving partial or full support under Contract 4289(01) have been: Charles Ross Endsley III, William Ross McCluney, Ching-Tu Chang, Gary Dale McNeely, and Richard Frederick Smith. Doctoral degree students have been: Arnie Lee Van Buren and Laszlo Adler.
Preface to Technical Report No. 15

This technical report comprises publications on the reflection of ultrasonic waves at a liquid-solid interface made between January 1975 and the present. During this period a number of publications have been made on other subjects; however, we prefer to concentrate on the reflection problem in this technical report in order to present a unified picture of the development of this subject. A later technical report will cover other subjects.

The report begins with the English language version of a paper whose Russian language counterpart was included in Technical Report No. 11. It may be worthwhile to point out that our Russian colleagues were impressed with the paper enough to make it the lead article in Akusticheskii Zhurnal for the year 1975.

In this paper we were able to get agreement between theory and experiment at the Rayleigh angle sufficient to permit us to plot them on the same graph. (Up until this time theory and experiment were agreeing only qualitatively so that such a comparison was not possible.) The agreement was made possible by advances both in experiment and in theory. The experimental advance was the use of a transducer which produced a truly Gaussian amplitude distribution in the incident beam (see Technical Report No. 8). The theoretical advance was the retention of higher-order terms in the power series expansion of the phase shift upon reflection than had been used previously by Brekhovskikh. This led to a complicated expression for the reflected beam, a power series expansion in Fresnel integrals, which had to be evaluated by computer.
A mathematical improvement resulted from interaction between the author and Werner G. Neubauer and Larry Flax of NRL. Rather than a series expansion, a closed-form solution in terms of error functions was introduced. This is contained in the second paper.

In the third paper we show that a considerable improvement of the agreement between theory and experiment results from the use of the theory of Bertoni and Tamir, with modification to account for the propagation distance of the incident beam. The data in this paper also are very carefully taken.

The fourth paper, given at the XV International Conference on Acoustics-Ultrasound in Prague, is a summary paper describing the application of the theory of Bertoni and Tamir to the results on stainless steel. In this paper we also indicate that a corrugated interface makes possible coupling to a surface wave which propagates in the negative-x direction along the interface. The fifth paper shows that this negatively directed surface wave causes the reflected beam to be displaced in the opposite direction from that observed at the Rayleigh angle.

The sixth paper shows not only that the displacement observed at liquid-solid interfaces can be observed at water-sediment interfaces, but also that the backward displacement phenomenon might exist because of the periodicity resulting from the granularity of the sediment. Furthermore, the water-sediment interface corresponds to a water-solid interface for which \( V_L > V > V_S \), a situation which previously had not been observed to give a displacement of the reflected beam. However, such a displacement does exist for this class of liquid-solid reflectors,
as we show in the seventh paper. The displacement is observed near the
critical angle for water-plexiglass, not the Rayleigh angle, however,
for there is no Rayleigh angle.

The eighth paper is an application of the surface wave phenomena
to the detection of subsurface flaws in metals. The surface waves
generated at the Rayleigh angle are described in the theory of Bertoni
and Tamir as "leaky waves" because the energy "leaks" from them back into
the liquid to become the reflected beam. The ninth paper shows how these
leaky waves can be used to detect subsurface flaws in solids. The final
paper begins a study of the effect of the introduction of a layer on the
solid.

Preface to Technical Report No. 17

This technical report comprises publications made between September
1974 and the present. During this period the program was expanded to
include the study of the nonlinear acoustics of solids. Because of the
broadening of the range of topics, in February 1978 we decided to separate
out a single topic—Ultrasonic Wave Reflection at Liquid-Solid Interfaces—and present it as Technical Report No. 15. The current technical report
overlaps Technical Report No. 15 in time, but not in subject matter. The
current technical report gives our contributions to subjects other than
reflection of ultrasonic waves at interfaces. For convenience, the
report is divided into three parts.

Part I is made up of two papers dealing with critical mixing of
binary liquid mixtures. These two papers essentially complete our
contribution to this subject for the present.
Part II covers two different measurement techniques. The first paper is a presentation of a comparison of ultrasonic pressure amplitudes measured by three different optical techniques and by a thermocouple probe with calculated amplitudes. The measurements were made simultaneously in the same ultrasonic field in 1958, but previously were unpublished. A revival of interest in calibration at the Miami meeting of the Acoustical Society of America persuaded the author and Floyd Dunn that our results would be of interest to our colleagues and thus prompted the publication. The second paper describes the improvement in measurement of ultrasonic wave velocities in solids which results from the use of capacitive transducer. This technique eliminates the perennial bond problem.

Part III comprises six contributions to the nonlinear acoustics of solids. The first is a short summary paper describing the temperature dependence of the nonlinearity parameters of germanium and of fused silica down to 77°K. The second is an invited paper, given at the 6th International Symposium on Nonlinear Acoustics in Moscow, which describes the measurement technique and presents results on germanium down to 3°K. The third is a more extensive description of both the nonlinearity parameter and the third-order elastic constants of germanium between room temperature and 3°K. The fourth is a short paper given at the ICA in Madrid on the nonlinearity parameter of four different types of fused silica. The fifth is a more extensive account of the results of our experiments on the nonlinearity of fused silica. The sixth and final paper is a summary paper, given at the International Symposium on
Nonlinear Acoustics in Paris, which gives the temperature dependence of the nonlinearity parameters and third-order elastic constants of germanium and copper and makes an attempt to relate them to other quantities of significance in solid state physics.

Preface To Technical Report No. 18

In Technical Report Nos. 15, "Ultrasonic Wave Reflection at Liquid-Solid Interfaces," and 17, "Studies of Linear and Nonlinear Ultrasonic Phenomena," we presented summaries of our contributions to certain subjects. Technical Report No. 18 is intended to expand on the summary and bring it up to date. It is divided into two parts.

Part I. Schlieren Studies of Ultrasonic Waves

The Report begins with the description of a unique goniometer designed by members of the Ultrasonic Group for use in the schlieren system for visualization of ultrasonic waves in liquids. By using the properties of parallelograms we were able to produce a precision goniometer without use of precision machine shop facilities. The second paper presents some photographs made with the goniometer in the schlieren system and shows the effect of a layer of Al₂O₃ on a stainless steel reflector of ultrasonic waves in water. The leaky Rayleigh wave excited in the Al₂O₃ layer has a velocity smaller than that excited either at a water-stainless steel interface or at a water-Al₂O₃ interface.

Part II. Nonlinear Acoustics of Solids

In relatively large single crystal samples (1 inch in diameter and 1 inch long) one can measure such things as "The Nonlinearity Parameters
and Third-Order Elastic Constants of Copper between 300 and 3°K" as reported in Paper No. 3. The fact that the measurements can be made to low temperatures is especially important, as the effect of thermal motion of the atoms is ignored in many theories. This means that they are strictly applicable only at 0°K. For comparison with these theories, then, we measure to the lowest readily obtainable temperature.

A problem encountered in the study of the nonlinear properties of solids is the fact that oftentimes it is difficult to grow large single crystals of interesting substances. Ordinarily we use a 30 MHz ultrasonic wave of finite amplitude to determine the nonlinearity parameters of single crystals 1 inch in diameter and 1 inch long. The amplitude of the second harmonic, which must be measured absolutely, typically is of the order of 10^{-2} Å in these samples. We posed for ourselves a question: Given our desire to measure nonlinearity parameters, what is the smallest sample one can measure with present technique? The fourth paper, "Measurement of Nonlinearity Parameters in Small Solid Samples by the Harmonic Generation Technique," is an attempt to answer the question.

Another question of fundamental importance to nonlinear acoustics of solids is the relationship between the nonlinearity parameter measured acoustically and the Grüneisen parameter which comes from measurement of thermal properties. This question is given a relatively simple, and almost complete, answer in the fifth paper on "Relationship between Solid Nonlinearity Parameters and Thermodynamic Grüneisen Parameters." This paper was based on the oral presentation given at the joint meeting of the Acoustical Society of America and the Acoustical
Society of Japan. This was an especially appropriate audience since two of the authors were from the United States and one was from Japan.

The final paper in this Report, "Quantum Mechanical Theory of Nonlinear Interaction of Ultrasonic Waves," answers in part another fundamental question of nonlinear acoustics. Presumably in the correspondence limit the quantum mechanical description of phonon-phonon interaction would become identical to the description (based on nonlinear elasticity) of the scattering of one acoustical disturbance by another. But this assumption was hard to prove. The paper provides specific examples. It begins with the general quantum mechanical description of phonon-phonon interaction and specializes the description to that of two collinear phonons of frequency \( v \) which interact to produce a phonon of frequency \( 2v \). (This comes from energy conservation: \( hv + hv = 2hv \).) By maintaining the wave description (avoiding quantization), one is able to show that the mathematical result is identical to that previously obtained from a generalization of elasticity. This is true also in the description of third harmonic generation. In third harmonic generation one is able to show, in addition, that the small term in the third harmonic amplitude which contains fourth-order elastic constants in fact comes from four-phonon interactions in the quantum mechanical picture, whereas all of the other terms (those involving second-order and third-order elastic constants) resulted from three-phonon interactions. The advantage of the quantum mechanical approach lies primarily in the fact that the path from the general description to the particular application is explicitly marked, and the point at which one makes any particular simplifying assumption can be located unambiguously.
Since Technical Report No. 18, our technical reports have dealt with the nonlinear acoustics of solids, with one exception. Technical Report No. 19 is an excellent recapitulation of the details necessary to measurement of the third-order elastic constants of cubic crystals. The results on silicon and germanium are good as well. Technical Report No. 20 answers a fundamental question that has plagued us for some time: how small can a solid sample be and still be large enough that diffraction does not cause problems in evaluation of nonlinear distortion? Some crystals can be grown only as small samples. The answer given is that samples greater than 4 mm in length and 5 mm in cross section are required. Number 21 deals with the effect of nonparallelism of surfaces in a solid. Although this is an old topic, new information is available in this report. Technical Report No. 22 gives the third-order elastic constants that enter into nonlinear distortion of an ultrasonic wave in a solid of hexagonal symmetry. A new representation of the nonlinearity is made in this report. Unfortunately, however, the combinations of third-order elastic constants given are in error. The problem has been traced to an error in the expression for the strain energy in a hexagonal crystal given by Einspruch and Manning. A publication by the author of this report gives the correct expression and makes the proper derivation. It will be submitted for publication soon.

Technical Report No. 23 gives the results of measurement of nonlinear distortion of ultrasonic waves in piezoelectric crystals such as quartz and lithium niobate. In Technical Report No. 24 we give an analysis of the propagation of finite amplitude waves in pure mode directions in
hexagonal crystals that were overlooked in No. 22. Also included is the theory for trigonal symmetry. Technical Report No. 25 is an attempt to answer a question of relative accuracy in the evaluation of second-order elastic constants compared with use of data from standard sources.

The final Technical Report No. 26 answers a question about higher order Bragg diffraction of light by finite amplitude ultrasonic waves raised in Technical Report No. 9. It shows that the theory of Blöme and Leroy accounts for observations of multiple images in the higher order. Such information is useful not only in ultrasonic Bragg imaging; it would be useful in the x-ray analogue as well.
III. GENERAL SUMMARY STATEMENT

As can be seen in Technical Reports and publications, the research on nonlinear acoustics has covered a wide range of topics in the physics of deformable media. This is entirely proper, and it has produced a wide range of results. In some instances the phenomenon which initially appeared to result from nonlinear interactions later proved adequately to be explained by a linear theory. This is true, for example, for the interesting observations on higher order Bragg imaging (Technical Report Nos. 9 and 25). The multiple images seen in the higher orders (two in the second order, three in the third, etc.) seem to be accounted for by the theory of Blömme and Leroy, a linear theory (with only one minor discrepancy remaining). To use a nonlinear model in such a case would have been confusing.

In other instances the nonlinearity can be observed, but does not play a dominant role. Such a situation was found in the study of ultrasonic waves incident onto a liquid-solid interface (Technical Report No. 18). We showed at the International Congress on Acoustics in Tokyo (Proceedings, ICA, Tokyo, Paper H-205 (1968) that nonlinear distortion occurs in surface waves generated at the Rayleigh angle, but have not yet defined a situation which would make constructive use of this result.

The third situation, the one that has provided motivation for the entire program, is one in which nonlinearity becomes the key element in making a physical explanation of the observed phenomena. Several such situations were studied during the course of the contract, but two examples suffice to prove my point: parametric interaction of ultrasonic waves in a resonant cavity (Technical Report No. 7) and nonlinear
distortion of ultrasonic waves in a solid (Technical Reports Nos. 14, 19, 20, 22-25). The former example excited considerable interest throughout the physics community and led to our most-requested reprint (Am. J. Phys. 39, 1522-27 (1971).) The latter example has led to a new technique for measuring physical parameters of fundamental importance to solid state physics—third order elastic constants. Since the technique is capable of measurements over a wide range of temperature the data are being reproduced by such reference works as Landolt-Börnstein's Tables of Fundamental Physical Constants and are being used both by experimental and theoretical solid state physicists. This desirable result justifies much of the effort expended on projects undertaken throughout the duration of the program on Nonlinear Acoustics.

Occasionally beginning graduate students approached the research with the impression that "nonlinear acoustics" was in some sense a more restricted study than "acoustics." Only a few weeks in the laboratory always revealed the ubiquity of nonlinearity. They found that all of nature is nonlinear and that the linear approximation is a special case. The nonlinear situation is the general situation.

In this context one confidently can predict that pioneering investigators in acoustics will continue to be in nonlinear acoustics for the foreseeable future. Whether one calls it nonlinear acoustics or something else, the most productive investigations will continue to be ones which result in a more accurate and a more complete description of physical reality.
IV. PUBLICATIONS RESULTING FROM RESEARCH ON ONR CONTRACTS


V. LECTURES GIVEN ON ONR RESEARCH


12. Laszlo Adler, "Ultrasonic Research in Modern Physics" (Invited), Tennessee Section, American Association of Physics Teachers, Memphis, 1970.


17. M. A. Breazeale, "Photon-Phonon Interaction for the Common Man" (Invited), Symposium on Molecular Energy Transfer, University of Mississippi, 1972.


47. Jacob Philip and M. A. Breazeale, "Combinations of the TOE Constants of the Fluoroperovskites CsCdF$_3$ and KZnF$_3$ Measured as a Function of Temperature," 102nd Meeting, Acoustical Society of America, Miami, 1981.


VI. TECHNICAL REPORTS RESULTING FROM RESEARCH ON ONR CONTRACTS


VII. GRADUATE STUDENTS, POSTDOCS, AND FACULTY ASSOCIATES
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4. Ashok Gottipamula, Ph.D., Osmania University, India, 1983-1984

5. Gonghuan Du, 1985-1986 (Assoc. Prof., Nanjing University, PRC)


APRIL 1984

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