Innovative Acoustic Techniques for Studying New Materials and New Developments in Condensed Matter Physics

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This project involved the use of innovative acoustic techniques to study new materials and new developments in solid state physics. Major accomplishments include a) the preparation and publication of a number of papers and book chapters, b) the measurement and analysis of an aluminum alloy quasicrystal and its cubic approximant c) the use of resonant ultrasound to measure acoustic attenuation and determine the effects of heat treatment on ceramics, d) the extension of our technique for measuring even lower (possibly the lowest) infrared optical absorption coefficient, and e) the measurement of the effects of disorder on the propagation of a nonlinear pulse, and f) the observation of statistical effects in measurements of individual bond breaking events in fracture.

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INNOVATIVE TECHNIQUES FOR STUDYING NEW MATERIALS 
AND NEW DEVELOPMENTS IN CONDENSED MATTER PHYSICS

This report summarizes the goals and accomplishments for ONR grant N00014-92-J-1186, "Innovative Acoustic Techniques for Studying New Materials and New Developments in Condensed Matter Physics". The goals of the project are a) to use resonant ultrasound spectroscopy to study new materials, such as quasicrystals and ceramics, b) to use a resonant photoacoustic technique to measure infrared optical absorption in highly transparent materials, c) to use acoustic analogs to study effects analogous to those of mesoscopic electronic systems, and d) the measurement of individual bond breaking events during the fracture of brittle materials.

Published papers, submitted papers, talks, etc.

Accomplishments during the three year period covered by this report include the publication of ten refereed papers and six book chapters, and the presentation of 22 contributed papers at meetings. Twenty invited talks were given, including six colloquia and three seminars at universities, and eleven symposium talks at international meetings. One paper for Phys. Rev. Lett. (on nonlinear pulses in disordered media) is nearly ready for submission, and a second Phys. Rev. Lett. on the isotropy of quasicrystals should be submitted during this summer. The total research group has consisted of three Graduate students involved in the research include Phil Spoor, Wei-li Lin (with ASSERT support), and Vern Hopkins (with only summer ONR support). Spoor and Lin are currently writing their Ph.D. theses. The research has benefited from two post doctoral scholars, Mark McKenna and Tianming Zhang. A number of undergraduates have made significant contributions to our research. A list of the publications, personnel, etc. is presented in the appendix. In the sections which follow, a summary of the research accomplishments is presented.

Probing the Elastic Isotropy of a Quasicrystal

Since the discovery of quasicrystalline symmetry in solids, there has been considerable interest in alloys such as AlCuLi, from which large (several millimeter-sized), stable crystals that exhibit a five-fold symmetric diffraction pattern can be grown (the T2 phase). The T2 phase is thought to be very closely related to the well-known R-phase, which consists of a bcc lattice of icosahedra and has a nearly fivefold-symmetric diffraction pattern. Distinguishing T2 and R phases apart on the basis of diffraction patterns alone is rather difficult. However, if the T2 crystals are indeed true quasicrystals, they should be elastically isotropic. Comparison of the isotropies of the two phases can therefore provide a vital clue to the underlying structures, and Resonant Ultrasound Spectroscopy (RUS) is perhaps the only technique currently available that is sufficiently sensitive to measure the difference unambiguously. In our research, we have compared the isotropies of the two phases of AlCuLi and found that the R-phase had an anisotropy \( \epsilon = \frac{1 - 2C_{44}}{(C_{11} - C_{12})} \) equal to 0.017 \pm 0.001, eight times greater than that of the T2 phase \( \epsilon = 0.0019 \pm 0.0004 \).

For the analysis of the measured data, we developed a computer program which allows us to fit our frequency data to different rotations of the elastic tensor. If the anisotropy in the
R-phases is due to the structural difference between the two phases, as opposed to internal defects or sample preparation errors, the data fit should have a preferred orientation which should agree with the orientation found from x-ray diffraction. We plotted the data fit for various orientations and oriented the samples using transmission Laue diffraction. The samples were originally oriented using external morphology only, as early attempts to orient them using reflection Laue were unsuccessful (the unusually large unit cell of the R-phase leads to a high degree of surface smearing during cutting and polishing, which is difficult to remove, even with etching). However, transmission Laue photos showed the high quality of the samples under study and made orientation and structure verification possible.

The research using RUS will form the basis for the Ph.D. thesis for Phil Spoor. A paper describing the technique and results for the quasicrystal is currently being prepared for submission to Physical Review Letters.

**Attenuation Measurements on Spherical Ceramic Particles**

In addition to the quasicrystal measurements, we used the RUS technique to study “proppants”, used for oil recovery and solar receivers. We measured the elastic constants and attenuation for various heat treatments, and discovered a peak in the attenuation at 1100 C that coincided with a minimum in Young’s modulus, thought to be due to the appearance and subsequent healing of internal microcracks. Our attenuation measurements were based on graphical determinations of Q for a representative mode of vibration, which was not straightforward since the imperfect, spheroidal proppants tended to have multiple overlapping resonances. Subsequently we developed nonlinear curve-fitting software to satisfactorily model all the peaks in each cluster, determining the Q of each very precisely. We were thus able to unambiguously confirm the correlation between the heat-treatment effects and the acoustic properties. A paper on these results will be prepared in the near future, in collaboration with John Helmman of the College of Earth and Mineral Science.

**Anderson Localization and Nonlinear Pulses in Disordered Media**

The study of systems which are both disordered and nonlinear is a relatively new frontier. Most of the research to date is theory, with significant contributions by mathematicians. A fundamental question is whether or not Anderson localization is weakened by the effects of nonlinearity. A survey of the theory papers shows that about half of the papers predict that Anderson localization is weakened by nonlinearity, and about half predict that it is not. In our current research we have used nonlinear surface waves on films of superfluid helium (third sound) to address this question. Our results were presented in an invited symposium lecture at a meeting of the American Physical Society in Washington, DC.

While the different predictions by the theory papers would seem to indicate a controversy, there is in fact no contradiction, because the conclusion depends on how the question is posed. For example, one may study the wave mechanics of a system by exciting it with a continuous wave, \( \cos (\omega t) \), and examining the transmission spectrum, \( S(\omega) \). On the other hand, one may launch a pulse into the system and study the temporal response
at an exit point, $T(t)$. In a linear system the two results would be simply related by a Fourier transform. However, this is no longer true for a nonlinear system, and different results may be obtained. Current theory predicts that for continuous wave excitation of a 1-D nonlinear disordered system of length $L$, eigenstates remain localized and $S(\omega)$ for $\omega > 0$ decreases exponentially with $L$. Our experimental results verifying the prediction for continuous waves (rather than pulses) were published in Phys. Rev. Lett. 69, 1087 (1992).

The theoretically predicted behavior for a pulse in a nonlinear disordered system is more interesting, and might be described with a simple picture as follows. For a linear 1-D disordered system, the behavior of a pulse is rigorously found by making a product of matrices from one end of the system to the other, and as predicted by rigorous theory, the eigenstates would be localized and the transmitted pulse energy would decrease exponentially with $L$. However, a nonlinear pulse has an extra degree of freedom which may be adjusted to satisfy conditions locally, over some characteristic length, i.e. the “width” of the pulse. If the width of the pulse is much less than the Anderson localization length, then the disorder has no effect and the pulse is transmitted without the exponential decrease. If the pulse width is sufficiently greater than the localization length, then transmission is exponentially decreased. When the width of the pulse is on the order of the localization length, then the pulse travels some distance with a slight decrease before an exponential decrease begins.

In order to study nonlinear pulse propagation experimentally, we used surface waves on a fluid because they are intrinsically nonlinear (the speed of the surface wave depends on depth, which is modified by the presence of a finite amplitude wave). We used surface waves in superfluid helium films to reduce viscous damping which would weaken long range phase coherence in the linear regime; water surface wave experiments suffer dramatically from the limitations of damping. In our experiment, the superfluid film coats a glass substrate, with a 1-D array of scatterers provided by grooves cut into the glass surface with a diamond wire saw. These scatterers yield about 30% reflection, which, as required for our studies, is found to be independent of wave amplitude. For the disordered system, an array of about 40 scatterers was used, with the spacing randomly varied about periodic positions within the limits of plus or minus one half of a lattice constant (1 mm). With this amount of disorder, the Anderson localization length was calculated to be about the same as the width of the nonlinear pulses, so that our experiment probed the interesting intermediate regime of behavior.

The behavior of the pulse as a function of distance was obtained using pairs of drive and receive third sound transducers (superconducting aluminum bolometers) with spacings of 6, 10, 16, 26, 32, and 38 lattice constants. At each distance, recordings of received pulse signals were made with typically 20 different drive levels, covering more than two orders of magnitude, from linear to nonlinear. At sufficiently high drive levels, a pulse with a time of flight depending on drive level (a clear nonlinear effect) was observed. The effect was similar to that found for nonlinear third sound on a bare substrate. The initial analysis of the nonlinear pulse data in the current experiment was the same as
that used in our previously published research. In the current experiment, in order to eliminate the necessity of calibrating all of the transducers, the result from the nonlinear signal was normalized with the linear signal at the same transducer. In addition, similar measurements were made with linear and nonlinear pulses traveling on a bare substrate and on a substrate with a periodic array of scatterers.

An important aspect of our experiment is that data were taken for more than one realization of the disorder for each distance, so that we could examine ensemble averaged results. This greatly reduced the scatter in the data, and we were able to verify the theoretical predictions for low amplitudes (showing the exponential decay), and for intermediate amplitudes (showing slight decay, followed by exponential decay). A paper on these results is currently being prepared for submission to Physical Review Letters.

Measurement of an extremely low (possibly the lowest) bulk infrared optical absorption

One of the acoustic innovations developed in our research program is a resonant photoacoustic technique for measuring optical absorption in highly transparent crystals and glasses. Such highly transparent materials are important for applications in optical fiber long distance transmission lines, in lenses and windows for high-power laser systems, and in electro-optic, magneto-optic, and acousto-optic components for optical computers, etc. The optical absorption in new materials is so small that it has become difficult to measure in conveniently sized (~1 cm) samples. One of the most sensitive methods for measuring optical absorption is the photoacoustic technique, which usually requires a high power pulse laser. Our resonant photoacoustic technique uses a continuous (CW) laser which has less power than a pulse laser, but nevertheless has orders of magnitude improved sensitivity in measuring optical absorption. This technique may be used to measure optical absorption coefficients as small as $10^{-8}$ cm$^{-1}$.

The difference between the conventional photoacoustic technique and the resonant photoacoustic technique is as follows: In the conventional technique, a high power laser pulse, containing several hundred mJ of energy, passes through a sample, and the energy absorbed generates an acoustic pulse; the acoustic pulse is detected with a piezoelectric transducer attached to the sample. The amount of optical energy which was absorbed may be determined from the amplitude of the acoustic pulse. The sensitivity of the technique is limited by the noise (electrical, thermal) in the acoustic transducer. In the resonant photoacoustic technique, the pulse laser is replaced with a continuous wave (CW) laser, which may have less power by a factor of 1000. However, the CW laser may be modulated at a frequency corresponding to an acoustic resonance of the sample, and when driven at resonance, the sample itself acts as a natural amplifier with a gain equal to the quality factor (Q) of the resonance. This gain occurs before the transduction, so that the transducer noise is not amplified. Since highly transparent samples are often made with high purity material, such samples will also have high mechanical Q's, at least $10^4$ and as high as $10^6$. The gain which arises by driving the sample at a high Q resonance more than compensates for the lower laser power, and results in ~100 times improved sensitivity. Furthermore, with CW
modulation, very low noise phase sensitive detection may be employed.

The minimum in the absorption of electromagnetic radiation occurs at about 1 \( \mu \text{m} \); indeed the absorption is so small here that no previous measurement has been able to detect any absorption. With the use of a Nd:YAG laser, with a wavelength just above 1 \( \mu \text{m} \), we have applied the resonant photoacoustic technique to study infrared absorption, and have observed the lowest measured optical absorption.

In our early research, we measured an infrared optical absorption coefficient of \( 7 \times 10^{-7} \) cm\(^{-1} \), with a precision of \( 5 \times 10^{-8} \) cm\(^{-1} \). This result was with a quartz sample, which being piezoelectric, induced an extra electrical signal in the PVDF transducers, resulting in a greatly enhanced transducer sensitivity (10 \( \mu \text{V/pm} \)). Attempts to measure a non-piezoelectric CaF\(_2\) sample were found to be quite difficult; without the piezoelectric effect, the transducer sensitivities were orders of magnitude smaller than those with the quartz. We switched to LiNb transducers, which have sensitivities of \( \sim 2 \) \( \mu \text{V/pm} \) with the CaF\(_2\) in the resonant photoacoustic apparatus. We have now been able to measure an optical absorption coefficient of \( 2 \times 10^{-7} \) cm\(^{-1} \) in the non-piezoelectric CaF\(_2\). This should establish our technique as the record holder for arbitrary samples. This research forms the basis for the Ph.D. thesis of Wei-li Lin. A paper describing our latest technique and results will be prepared in the near future.

**Developments in the study of fracture**

In our early research with the fracture we had been studying a certain type of polystyrene foam as a large scale model of a fracturing material. A fracture test system was developed and initial measurements were made with a bandwidth up to 100 KHz, using conventional accelerometers. However, it was realized that with a measured sound speed of \( \sim 700 \) m/s and an average foam cell size of 1 mm, the identification of individual cells breaking would require a time resolution of \( \sim 1 \) \( \mu \text{s} \). We next tried 2.25 MHz piezoelectric NDE transducers, but decided that the face of the transducer was too broad to correctly receive widely spaced high frequency signals. We next tried \( \sim 1.5 \) mm diameter 10 MHz “pinducers”, but found that the process of gluing the transducers to the sample destroyed the transducers. We then tried putting the sample and an unmounted pinducer under water, and obtained excellent results, with individual bond breaking events readily recorded.

Our first measurements involved fracturing a notched sample with transverse forces, and this process created a graded stress field across a section of the sample. A theoretician in our department, Jayanth Banavar, pointed out that this was analogous to having a phase transition in an external field (e.g. a liquid-vapor transition in the presence of a gravitation field), and the statistical physics of this situation was not as interesting as a system with no external field. As a result we changed our fracture mode to uniform tensile stress. For the two different stress fields, the measurements of the sequences of individual bond breaking events were consistently different. We believe that for the graded stress field, the fracture is governed by the statistical distribution of strengths in only one line of bonds (the one with the maximum tensile stress), and being only one-dimensional, this
can fail relatively suddenly, resulting in a sharp onset. With the uniform stress field, the
distribution of bond strengths is two-dimensional, and a broader cascade of weaker bond
breaking events precedes the complete fracture. In any case, our experiment quantitatively
reveals the fundamental differences in the statistical physics of the system with and without
an external field. Further measurements and analysis will be continued.
APPENDIX. PUBLICATIONS, PRESENTATIONS, ETC.

PAPERS PUBLISHED IN REFEREED JOURNALS


BOOKS OR CHAPTERS SUBMITTED FOR PUBLICATIONS


2. J. D. Maynard, “Phonons in Crystals, Quasicrystals, and Anderson Localization” to be published as a chapter in Handbook of Acoustics, ed. M. J. Crocker (John Wiley and Sons, New York)

3. J. D. Maynard, “Tutorial on Acoustic Imaging”, to be published by the Acoustical
Society of America

BOOKS OR CHAPTERS PUBLISHED


INVITED PRESENTATION AT WORKSHOPS OR PROFESSIONAL SOCIETY MEETINGS

1. Colloquium, Department of Physics, University of Pittsburgh, April 13, 1992, “Nonlinearity and Disorder”


5. Invited Lecture, 1992 Physical Acoustics Summer School, Asilomar Conference Center,
Pacific Grove, CA, June, 1992, “Linear and nonlinear wave propagation in periodic, random, and quasiperiodic media”

6. Invited lecture, Seventh International Conference on Phonon Scattering in Condensed Matter, Cornell University, August, 1992, “Learning about phonons with frequencies below one KHz”

7. Seminar, Cornell University, Department of Physics, September 8 1992 “Acoustic analogs of mesoscopic systems”, David Lee, host

8. Colloquium, Department of Physics, West Virginia University, December 3, 1992, “Tuning-up a Quasicrystal”, Thomas Myers, host

9. Colloquium, Department of Physics, Washington University, St. Louis, MO January 13, 1993, “Tuning-up a Quasicrystal”, J. E. Shrauner, host


13. Colloquium, Department of Physics, University of Oregon, Eugene, OR, October 7, 1993, Martin Weybourne, host

14. Colloquium, University of California, Irvine, CA, January 28, 1994 “Tuning-up a quasicrystal”

15. Colloquium, University of Washington, Seattle, WA, January 31, 1994 “Tuning-up a quasicrystal”


19. Seminar, Penn State University, Department of Engineering Science and Mechanics, University Park, PA, January 26, 1994 “Tuning-up a quasicrystal”
CONTRIBUTED PRESENTATIONS AT WORKSHOPS OR PROFESSIONAL SOCIETY MEETINGS


18. V. A. Hopkins, M. J. McKenna, and J. D. Maynard, “Anderson localization of 3He with variable disorder provided by a 4He solid/liquid interface, presented at the 20th International Meeting of Low Temperature Physics, Eugene, Oregon, August 1993


HONORS/AWARDS/PRIZES

Silver Medal in Physical Acoustics, awarded to J. D. Maynard, November 30, 1994
GRADUATE STUDENTS SUPPORTED

1. Philip Spoor (Ph.D. candidate, acoustics), Elastic Constants for Aluminum Alloy Quasicrystals and High Tc Superconductors

2. Wei-Li Lin (Ph.D. candidate, physics; ASSERT support), Infrared resonant photoacoustics

3. Vern Hopkins (Ph.D. candidate, physics, Summer support), Nonlinear pulses in disordered array of scatterers

POSTDOCTORALS SUPPORTED UNDER CONTRACT FOR YEAR ENDING 31 MAY 1994

Mark McKenna, Research Associate, July 1, 1989 to July 31, 1993

Tian-ming Zhang, Postdoctoral Scholar, June, 1994 to present

MISCELLANEOUS

Meetings attended:


2. University of California Summer School on Nonlinear Science, “Slips, cracks, and tears” (attended by student Wei-li Lin, August 1992)

Undergraduates Involved in Research:

1. Ron Stanley, Summer 1991
2. Brian Pudliner, Summer 1991
3. Chris Koeppen, Spring 1992
4. Brian S. Wilson, Spring 1992
5. Justin Keat, Summer 1992
7. Steve Savitski, Senior 1994
8. Rob Baillis, Senior, 1994
9. Jason White, Summer 1993
10. Joseph Buck, Summer 1994
11. Robert McNeese, Summer 1994