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INNOVATIVE TECHNIQUES FOR STUDYING NEW MATERIALS AND NEW DEVELOPMENTS IN SOLID STATE 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 Solid State 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 past year include the publication of three refereed papers and two book chapters, and the presentation of six contributed papers at meetings. Seven invited talks were given, including two colloquia and two seminars at universities, and three 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. During the past year the total research group has consisted of three graduate students (some with only partial ONR support), but a new graduate student and postdoc have joined the group in June, 1994; 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 brief summary of the research accomplishments will be 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 T_2 phase). The T_2 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 T_2 and R phases apart on the basis of diffraction patterns alone is rather difficult. However, if the T_2 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 RUS is perhaps the only technique currently available that is sufficiently sensitive to measure the difference unambiguously. As reported earlier, we compared the isotropies of the two phases of AlCuLi and found that the R-phase had an anisotropy $\epsilon = |1 - 2C_{44}/(C_{11} - C_{12})|$ equal to 0.017 ± 0.001 , eight times greater than that of the T_2 phase ($\epsilon = 0.0019 \pm 0.0004$).

We have recently prepared and measured a second sample of R-AlCuLi, and found it to have $\epsilon = 0.022 \pm 0.002$, in good agreement with the first. In addition, we have 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 have plotted the data fit for various orientations and are currently orienting 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 should make orientation and structure verification possible. A second T_2 sample has also been measured and is currently undergoing analysis.

Attenuation Measurements on Spherical Ceramic Particles

Earlier we reported results on spherical ceramic particles, called "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. However, we recently 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 our previous result.

Anderson Localization and Nonlinear Pulses in Disordered Media

As discussed in the original proposal, 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 the recent APS meeting 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

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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.

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. The consequences of this are illustrated in Fig. 1, which shows the log of the pulse transmission as a function of distance traveled. 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, as shown by the upper line in Fig. 1. If the pulse width is sufficiently greater than the localization length, then transmission is exponentially decreased, as shown by the lower line. 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, as shown by the middle line.

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 shown in Fig. 1.

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. The results are shown in Fig. 2.

Fig. 2a and 2b show results for one transducer in the periodic system at two different drive levels. It can be seen that the normalized nonlinear pulse does not increase in amplitude with additional drive level, but only changes in time-of-flight. Similar results are seen on the bare substrate. However, for the disordered system, the size of the normalized nonlinear pulse increases with increasing drive level, as shown in Fig. 2c and 2d. Since the data are normalized with the linear signal, which by rigorous theory must be exponentially decaying because of localization, then the increase in the nonlinear signal with amplitude means that the nonlinear pulse is being less localized. This effect as a function of distance is shown by taking the log of the nonlinear enhancement (at the highest drive level) over the exponentially decaying linear pulse and plotting versus transducer distance. The results are given by the circles in Fig. 1. The scatter in the data is not due to noise in the experiment, but rather is due to the process of normalization to the linear pulse, which in the single realization of disorder in the experiment, is undergoing the analog of conductance fluctuations. Despite the fluctuations, the data clearly are best fit by the theoretically predicted intermediate behavior.

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

In our previous report, we announced a measured infrared optical absorption coefficient of $7 \times 10^{-7} \text{ cm}^{-1}$, with a precision of $5 \times 10^{-8} \text{ cm}^{-1}$. 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 ($10 \text{ } \mu\text{V/pm}$). 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. Recently we have switched to LiNb transducers, which have sensitivities of $\sim 2 \text{ } \mu\text{V/pm}$ with the CaF_2 in the resonant photoacoustic apparatus. We have now been able to measure an optical absorption coefficient of $2 \times 10^{-7} \text{ cm}^{-1}$ in the non-piezoelectric CaF_2 . This should establish our technique as the record holder for arbitrary samples. We plan to submit a patent for the resonant photoacoustic method using the LiNb transducers.

Developments in the study of fracture

In the fracture experiment we have 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 $\sim 700 \text{ m/s}$ and an average foam cell size of 1 mm, the identification of individual cells breaking would require a time resolution of $\sim 1 \text{ } \mu\text{s}$. 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 ~ 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, as illustrated in Fig. 3a. This process created a graded stress field across a section of the sample, as illustrated in Fig. 3b. 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, as illustrated in Fig. 3d and 3e. For the two different stress fields, the measurements of the sequences of individual bond breaking events were consistently different, as exemplified in Fig. 3c and 3f. 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 the sharp onset shown in Fig. 3c. 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, as shown in Fig. 3f. 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 are underway.

Current and Other Funding

It is anticipated that there will be no remaining funds at the end of the contract period.

Other research grants include:

1. NSF Division of Materials Research, Condensed Matter Physics Program, DMR 93-06791, which includes 1 man-month of the principal investigators time.
2. ONR, Physics Division, September 15, 1993 to September 14, 1996, 180,000/3 yr, "Anisotropic heat exchanger/stack configurations for thermoacoustic heat engines"; includes 1 man-month of time for the principle investigator, distributed over 12 months.

**APPENDIX: OFFICE OF NAVAL RESEARCH
PUBLICATIONS / PATENTS / PRESENTATIONS / HONORS REPORT
for
1 OCTOBER 1993 through 30 MAY 1994**

R&T Number: 4126941—09

Contract/Grant Number: G N00014-92-J-1186 and N00014-93-1-0779

Contract/Grant Title: Innovative Acoustics Techniques for Studying New Materials and new Developments in Solid State Physics, includes ASSERT

Principal Investigator: Julian D. Maynard

Mailing Address: 104 Davey Lab
University Park, PA 16802

Phone Number: (814) 865-6353

E-Mail Address: maynard@phys.psu.edu

- a. Number of Papers Submitted to Refereed Journals: 0
- b. Number of Papers Published in Refereed Journals: 3
- c. Number of Books or Chapters Submitted but not yet Published: 3
- d. Number of Books or Chapters Published: 2
- e. Number of Printed Technical Reports & Non-Refereed Papers: 0
- f. Number of Patents Filed: 0
- g. Number of Patents Granted: 0
- h. Number of Invited Presentations at Workshops or Prof. Society Meetings: 7
- i. Number of Presentations at Workshops or Prof. Society Meetings: 6
- j. Honors/Awards/Prizes for Contract/Grant Employees: 1
- k. Total Number of Graduate Students and Post-Docs Supported at least 25 % this year on this contract/grant:
 - Grad Students: 3
 - Grad Student Female: 0
 - Grad Student Minority: 0
 - Post-Docs: 1
 - Post_Docs Female: 0
 - Post-Docs Minority: 0

PUBLICATIONS, PRESENTATIONS, ETC.

PAPERS SUBMITTED TO REFEREED JOURNALS

(Not yet published)

None as of June 1994.

PAPERS PUBLISHED IN REFEREED JOURNALS

1. J. D. Maynard, "Classical analogs of mesoscopic quantum phenomena", *Physica B* **194-196**, 231-238 (1994)
2. M. J. McKenna, Justin Keat, Jun Wang, and J. D. Maynard, "Experiments on non-linear wave propagation in disordered media", *Physica B* **194-196**, 1039-1040 (1994)
3. V. A. Hopkins, M. J. McKenna, and J. D. Maynard, "Anderson localization of ^3He with variable disorder provided by a ^4He solid/liquid interface", *Physica B* **194-196**, 1137-1138 (1994)

BOOKS OR CHAPTERS SUBMITTED FOR PUBLICATIONS

1. J. D. Maynard, "Acoustical Holography", to be published as a chapter in *Handbook of Acoustics*, ed. M. J. Crocker (John Wiley and Sons, New York)
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

1. J. D. Maynard, "Learning about phonons with frequencies below one KHz", in *Phonon Scattering in Condensed Matter VII*, ed M. Meissner and R. O. Pohl, (Springer-Verlag, Berlin, 1993) pp. 239-243
2. J. D. Maynard, "Nearfield acoustic holography and arbitrarily shaped sources", Proceedings of the International Meeting on Acoustical Imaging, Lyon, France, in *Journées Imagerie Acoustique*, ed. Jean-Louis Chauray (1994)

INVITED PRESENTATION AT WORKSHOPS OR PROFESSIONAL SOCIETY MEETINGS

1. Colloquium, University of California, Irvine, CA, January 28, 1994 "Tuning-up a quasicrystal"

2. Colloquium, University of Washington, Seattle, WA, January 31, 1994 "Tuning-up a quasicrystal"
3. Invited Symposium Lecture, 125th Meeting of the Acoustical Society of America, Ottawa, Ontario, May, 1993, "Pulses, nonlinearity, and Anderson localization"
4. Invited Lecture, International Meeting on Acoustical Imaging, Lyon, France, March 1, 1994, "Nearfield acoustic holography and arbitrarily shaped sources"
5. Invited Symposium Lecture, APS Meeting, Washington, DC, April 1994 "Disorder and nonlinearity: Studies in liquid helium"
6. Seminar, Penn State University, Department of Engineering Science and Mechanics, University Park, PA, January 26, 1994 "Tuning-up a quasicrystal"
7. Seminar, University of Maryland, Department of Physics, May 3, 1994 "Experimental studies of nonlinearity and disorder", John D. Weeks, host

CONTRIBUTED PRESENTATIONS AT WORKSHOPS OR PROFESSIONAL SOCIETY MEETINGS

1. P. S. Spoor, M. J. McKenna, and J. D. Maynard, "A comparison of elastic constants of the quasicrystalline and cubic approximant phases of AlCuLi, using resonant ultrasound spectroscopy", J. Acoust. Soc. Am. **93**, 2276 (1993).
2. 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
3. M. J. McKenna, J. Keat, J. Wang, and J. D. Maynard, "Experiments on nonlinear wave propagation in disordered media, presented at the 20th International Meeting of Low Temperature Physics, Eugene, Oregon, August 1993
4. S. R. Savitski and J. D. Maynard, "Observation of Individual Bond Breaking Events in Precursors, Cascades, etc. in the Onset and Progression of Fracture", Bull. Am. Phys. Soc. **39**, 860 (1994)
5. P. S. Spoor and J. D. Maynard, "Measurements and Analysis of the anisotropy of the AlCuLi quasicrystal and the related R-phase", Bull. Am. Phys. Soc. **39**, 862 (1994)
6. V. A. Hopkins, M. J. McKenna, and J. D. Maynard, "Vortex generation in a capillary array: A reciprocity calibration", Bull. Am. Phys. Soc. **39**, 1049 (1994)

HONORS/AWARDS/PRIZES

Silver Medal in Physical Acoustics, to be awarded November 30, 1994

**GRADUATE STUDENTS SUPPORTED UNDER
CONTRACT FOR YEAR ENDING 31 MAY 1994**

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), Infrared resonant photoacoustics
3. Vern Hopkins (Ph.D. candidate, physics), Nonlinear pulses in disordered array of scatterers

**POSTDOCTORALS SUPPORTED UNDER
CONTRACT FOR YEAR ENDING 31 MAY 1994**

Tian-ming Zhang, Postdoctoral Scholar, began June, 1994

MISCELLANEOUS

Undergraduates Involved in Research:

1. Michael Baloh, Senior, 1994
2. Steve Savitski, Senior 1994
3. Rob Bailis, Senior, 1994
4. Jason White, NSF Research Experience for Undergrad. student 1993
5. Joseph Buck, Junior, summer 1994
6. Robert McNeese, NSF Research Experience for Undergrad. student 1994

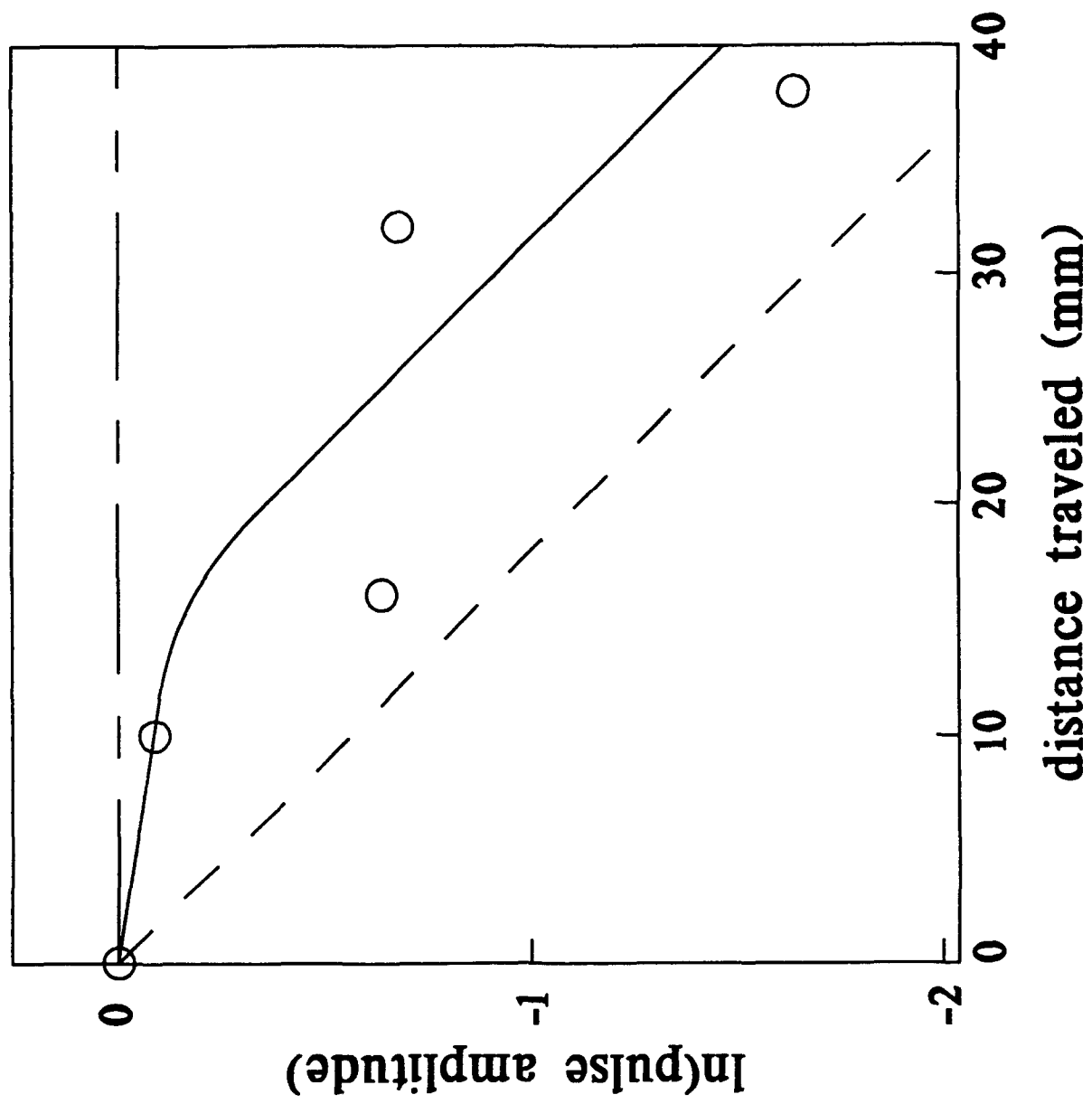


Figure 1.

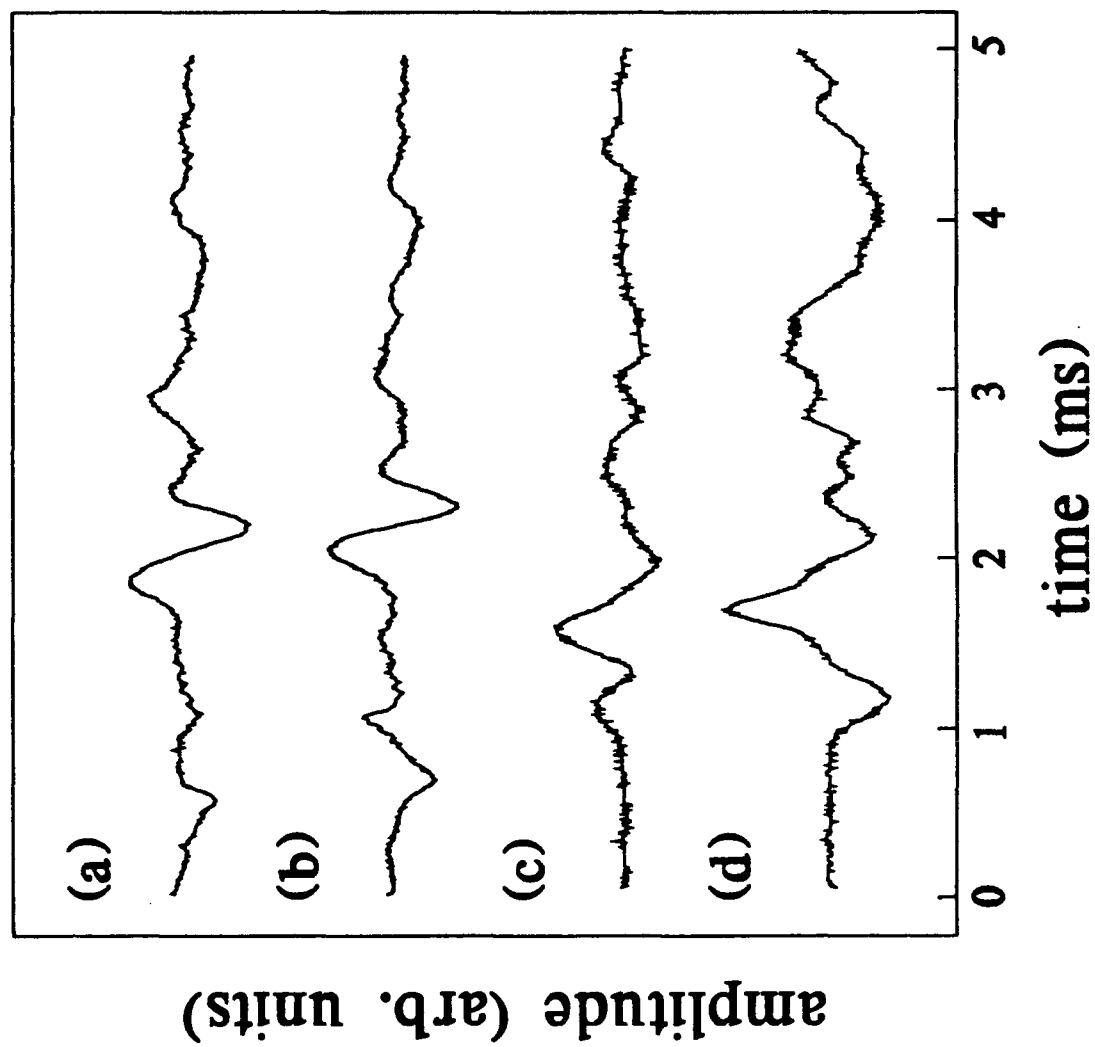
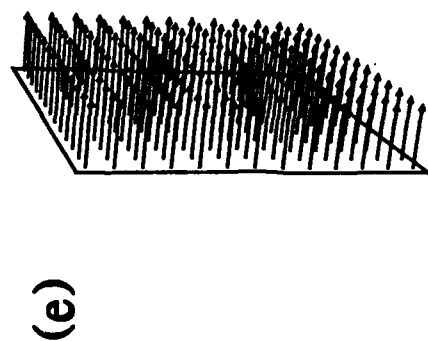
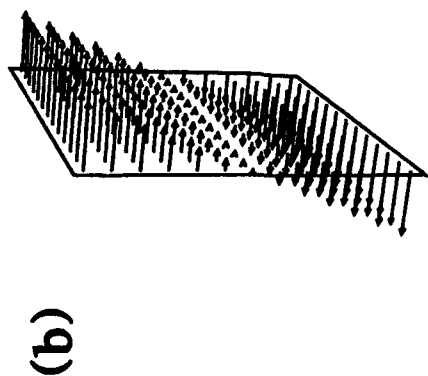
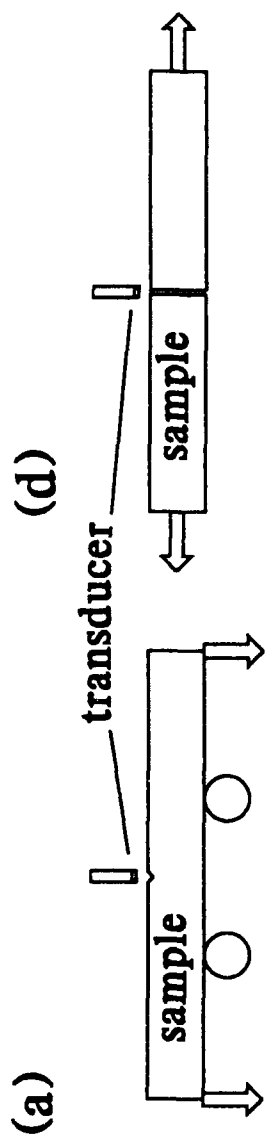


Figure 2.



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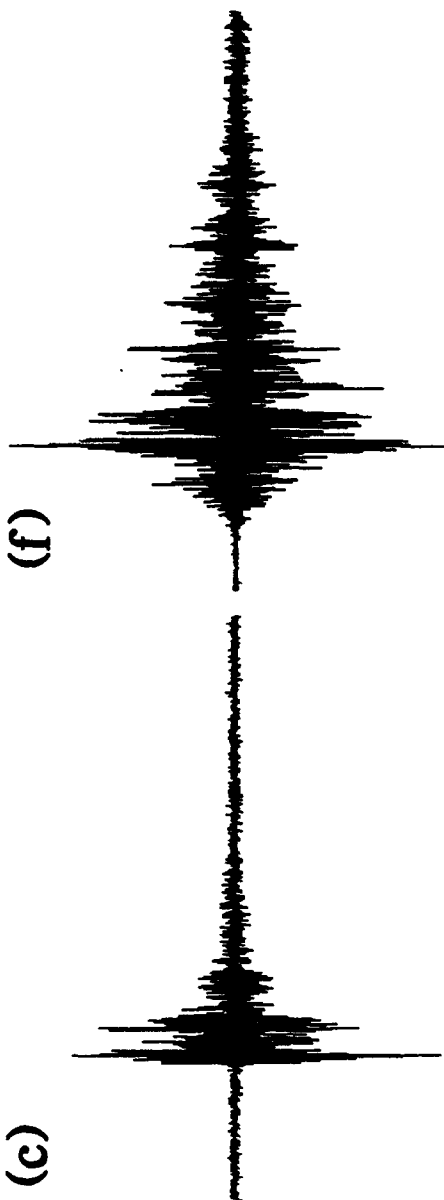


Figure 3.