Ultrasonic Absorption Microscopy and Spectroscopy
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I. INTRODUCTION

The ultrasonic instrument considered in this paper is capable of resolving structure on a microscale, i.e., down to at least one micron. The term "microscope" has been employed by other investigators to describe devices which incorporate, as a characteristic feature, some method of detecting acoustic field distributions with macroscopic resolution, i.e., of the order of millimeters. A number of these methods were reviewed by Rozenberg several years ago (1955).\(^1\) However, none of the methods described in the above listed reference contain the inherent practical capability of resulting in an instrument to resolve microstructure by ultrasonic means. It will become apparent, from the analysis reviewed here and from the results presented on the production and measurements of ultrasonic fields in the appropriate frequency range, that a device to resolve structure in thin specimens on a scale of resolution comparable to light microscopy is feasible. Further, the ultrasonic method described herein makes possible uhf acoustic absorption spectrometry which is important in the study of the structure of liquids and which should become useful in studies of biological systems for identifying distributions of macromolecular species characterized by different spectra of relaxation frequencies.

The principles of operation and the first stage of instrumentation development of the type of ultrasonic microscope discussed here were first reported in 1958\(^2\) and initial experimental results were presented in 1959.\(^3\) At that time a steady rate of progress in the evolution of instrumentation seemed assured. However, research support was withdrawn after a short but promising initial period of development activity, and the emphasis was then placed on employing and evolving equipment for measuring ultrasonic absorption coefficients of liquids at very high frequencies. This latter effort entails simpler instrumentation problems than does the microscope. To date, measurements have been made at frequencies as high as 2 kMc and amplitude absorption coefficient values as high as 1400 cm\(^{-1}\).
have been accurately determined. Such a high absorption value corresponds to an intensity decrement of $e^{-1}$ in a path length of 3.6$\mu$. These data are the first reported results using coherent acoustic energy in the kilomegacycle frequency range with high absorption liquids. However, it is of interest to note here that absorption measurements at approximately 1 kMc in mercury (a low absorption liquid) were reported in 1947 and that more recently acoustic absorption measurements in piezoelectric solids have been reported at frequencies as high as 24 kMc.

The combination of kilomegacycle frequencies and high absorption coefficient values represents an intractable combination to be handled by methods previously developed for measuring acoustic absorption in liquids. The interferometric, the optical, and the pulse methods suffer from alignment problems which become more critical as the wavelength of sound decreases. The assumptions of plane waves and uniform intensity distributions over the wave fronts are inherent in the optical method. The pulse transmission and pulse reflection methods also suffer from transit time limitations. However, successful measurement of high absorption coefficients in the frequency range of interest can be achieved with an instrument embodying the "transient" thermoelectric probe as a detector of acoustic field parameters. This detection method combines accuracy with high geometric resolution capability for (1) measuring steep field gradients in absorption coefficient determinations and (2) detecting fine scale structure in field distributions for ultrasonic microscope applications. It should be noted that, as shown below, the resolution of microstructure by acoustic energy can be achieved by employing unfocused sound and an appropriate small probe, or array of such probes, immediately adjacent to the thin specimen to determine the field distribution in the transmitted "beam." Such a configuration eliminates the difficulties implicit in the use of coupling and transmission media characterized by inherent high absorption, at the frequencies considered here, which are an integral part of focused beam designs.

A primary objective of this paper is a review of the analysis of the resolution capability of ultrasonic microscopes based upon the use of unfocused sound and thermoelectric detection of the field distribution. A second objective is a description of the form and performance of the instrument which is used at present for obtaining absorption coefficient data at ultrahigh frequencies and an illustration of the type of
data which is obtained. A third objective is an outline of some of the design and fabrication problems associated with visual presentation of ultrasonically observed structures and with speed of information accumulation, which must be solved to make a high resolution ultrasonic microscope available for practical use.

II. PRINCIPLES OF OPERATION AND ANALYSIS

The principle of operation of the type of ultrasonic absorption microscope of interest here is illustrated in Figure 1. High frequency pulsed acoustic waves are generated in a coupling medium by a piezoelectric element such as a circular X-cut quartz plate vibrating in a thickness mode. The coupling liquid conducts the sound to and from the specimen which is interposed between the sound source and a small thermoelectric probe or array of such probes. The array detects the acoustic energy transmitted through the portions of the specimen adjacent to the individual probes. Thus, an acoustic image of the structure can be produced by moving the specimen relative to the probe array.

When this type of device is employed as an ultrasonic absorption spectroscopy for studying liquids, the medium, whose absorption coefficient is to be measured, is in contact with the sound source and fills the chamber. A single thermocouple probe is moved along the direction of sound propagation in the medium, and the acoustic intensity gradient is determined, from which the intensity absorption coefficient per unit path length is computed.

A single piezoelectric element can provide acoustic energy over a very wide range of frequencies by exciting it electrically at odd harmonics of the fundamental thickness resonance frequency. For example, one transducer in use at this laboratory utilizes a 3/4 in. diameter X-cut quartz element to operate from 4 Mc, the fundamental thickness mode of vibration, to 1948 Mc, the 487th harmonic, and this operating frequency span with a single crystal does not represent a practical limit.

For spectroscopy, a single positioning system with three rectangular degrees of freedom is employed to support and move the probe. Two degrees of freedom provide motion of the probe in planes parallel to the crystal face for study of
the field distribution and consequent choice of the coordinates at which movement perpendicular to the wave fronts is made, with the third degree of freedom, to determine the intensity decrement. Precise knowledge of the amount of displacement of the probe in the direction of wave propagation is essential. For example, in the present instrument, readings to $10^{-5}$ inch are possible, and this is essential in determining absorption coefficient values of $1500 \text{ cm}^{-1}$ since the total path length in the liquid over which readings are obtained is approximately $10^{-3}$ inch. Measurements of higher absorption coefficient values will require increased precision in probe displacement measurements. In initiating a set of measurements, the thermocouple junction is moved into close proximity to, or actual contact with, the crystal face. This is accomplished by observing the transient contact potential produced when the probe touches the crystal electrode, and this operation is aided by viewing the junction within the liquid with an optical microscope. The viewing is done through a thin plastic window which is approximately parallel to the crystal face and which constitutes the end wall of the liquid confining chamber. No appreciable interference with the radiation field is produced by placing this window close enough to the crystal to view the junction at a magnification of 40 X or greater because the fraction of the acoustic energy radiated by the transducer which reaches the window is negligible.

For ultrasonic microscopy, the present instrument is provided with two positioning systems - one to support and move the specimen and the other to carry the single probe or array of probes. Such an arrangement is desirable, at least in the development stage, since the relative intensity of the sound arriving at any probe position is dependent upon both the position in the field and upon the fraction of the incident sound energy absorbed by the portion of the specimen immediately adjacent to the probe. Thus, it is essential that the probe position be adjustable to place it at a position where the acoustic field parameters (with the specimen removed from the field) do not exhibit steep gradients. However, during examination of a specimen it is preferred to move the latter while maintaining the probe in a fixed position since then variations in probe output can be directly identified with structure changes in the specimen. Should the probe be moved, with the specimen maintained in a fixed position, changes in probe output may result from two factors - (1) changes in the field distribution undisturbed by the specimen and (2) variations in absorption from one position to another in the specimen. The identification
of structure would then require that the effects of these two factors on the observed field distribution be separated. By comparison, maintenance of the probe in a fixed position in the field and movement of the specimen permit all the variation in the acoustic level at the probe to be interpreted in terms of structural features of the specimen.

Since close proximity of the probe or thermocouple junction and the crystal face or specimen surface are desired, the configuration of the thermocouple wires (copper and constantan) is arranged as illustrated in Figure 2. The stiffer constantan provides the main support and spring characteristic. The junction is formed by butt welding the copper filament to the tip of the small-radius-of-curvature loop of constantan wire. Annealed, commercial wires 0.0005 in. in diameter are used for the thermocouple components. The tip of the copper filament and the portion of the constantan loop nearest to the transducer are etched in acid to result in a junction of minimum size. The thermocouple components are butted into the proper configuration, and the weld is accomplished by repetitive discharges from condensers 2 μ total capacitance charged to 60 volts. Thermocouples in which the maximum dimension of the junction is less than 5 μ are readily constructed.

The ultimate capability, e.g., resolution, of the type of ultrasonic microscope described here depends critically upon the dimensions and values of the physical parameters which characterize the probe materials and the imbedding medium. Accordingly, an analysis relating performance of the instrument to these parameters, the intensity, the duration of the pulse of acoustic energy, and to the sensitivity of the electrical detecting circuitry, will now be given. To accomplish this, the formulas which describe the transient response of thermocouple probes imbedded in fluid media and subjected to pulses of ultrasonic energy are employed.

An estimate of the resolving capability of the ultrasonic microscope can be obtained as follows. Let μ designate the intensity absorption coefficient per unit path length in the specimen. The value of this quantity is in general different from one site to another and may also be dependent upon the direction of propagation of the sound with respect to oriented structure in the specimen. The resolution of the instrument for detecting structure characterized by a value of the absorption coefficient different from the average value for a larger volume of the specimen is accomplished by determining the minimum diameter
of a cylindrically symmetrical structural element (with \( \mu \) a maximum in the center and extending through the thickness of the specimen) that can be detected, Figure 3. Let

\[
\mu = \mu_o + \Delta \mu
\]  

(1)

where \( \mu_o \) is the average value of the absorption coefficient throughout a volume which includes the structural element and \( \Delta \mu \) is the increase in absorption coefficient caused by the presence of the element. The quantity \( \Delta \mu \) can be expressed as

\[
\Delta \mu = \Delta \mu_m f(a)
\]  

(2)

where \( a \) is the radial distance coordinate from the position of maximum absorption and \( f(0) = 1 \). Choose a parabolic form for the function \( f(a) \), i.e.,

\[
f(a) = 1 - Ba^2
\]  

(3)

If \( \Delta \mu \) decreases to 0.7 of \( \Delta \mu_m \) in a distance \( \Delta a \), then

\[
B = 0.3/(\Delta a)^2
\]  

(4)

From the analysis presented in reference 7, it is possible to calculate approximately the ratio of the heat conducted away from the element, \( H_{ct} \), to that generated above the average in the element, \( H_{at} \), both in the time interval \( t \) as

\[
\frac{H_{ct}}{H_{at}} = \frac{Kt}{\rho C_p} \left( \frac{f'(a)}{f(a)} \right)_{a = 0}
\]  

(5)

where \( \rho C_p \) is the heat capacity per unit volume per degree, \( K \) is the coefficient of thermal conductivity and \( t \) is the time duration of exposure. Combining Eqs. 3, 4 and 5 yields

\[
\frac{H_{ct}}{H_{at}} = \frac{Kt}{\rho C_p} \frac{0.6}{(\Delta a)^2}
\]  

(6)

If the numerical values, \( \rho C_p = 4 \) joules/cm\(^3\)/°C and \( K = 0.005 \) w/cm/°C, are inserted into this expression (these values are within the range characteristic of many nonmetallic solids at room temperature) and if an exposure duration \( t = (10)^{-5} \) sec is chosen, then for \( \frac{H_{ct}}{H_{at}} = 1/2 \), the value of \( \Delta a \) computed from

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Eq. 6 is approximately 1 micron. This is the resolution as limited by conduction in the specimen and for the pulse duration indicated.

Since the temperature increment at the probe must lie in the range of detectibility, it is necessary to relate it to the intensity and pulse duration. For the purpose of this calculation one can assume that the medium in which the probe is imbedded has an absorption coefficient value equal to the average value for the specimen and therefore the temperature difference to be detected is just equal to the increment above (or below) the average in the structure element. Integration of Eq. 1 of reference 7 for the initial time rate of temperature change (above the average) as a function of the absorption coefficient, the heat capacity and the intensity yields

\[ \Delta T = \frac{\Delta \mu ml_t}{\rho C_p} \]  

(7)

where \( \Delta T \) designates the sound intensity and the time interval is so short that thermal conduction can be neglected. This calculation also neglects the contribution to the temperature rise contributed by the action of viscous forces at the probe surface. (7) For a fixed time of exposure, the intensity and absorption coefficient determine the temperature increment. Obviously the greater the absorption coefficient difference, the smaller need be the intensity for a desired temperature increment. In order to maintain the intensity within reasonable bounds, it is necessary to operate in the kilomcycle frequency range for many materials of interest. For example, if \( t = (10)^{-5} \) sec as above and if \( \Delta \mu = 0.1 \mu \), then for a value of \( \mu \) equal to 1000 cm\(^{-1}\), the requisite acoustic intensity is \((10)^{4}\) w/cm\(^2\) for a temperature increment of 2.5°C. If the pulse duration is increased to \((10)^{-4}\) sec (constituting a loss in resolution by \(\sqrt{10}\)), then the required intensity is reduced to 1000 w/cm\(^2\). This level, which is consistent with tensile strength limitations of this material, is within a factor of three of the values now generated in this laboratory directly off the face of X-cut quartz crystals operated at the fundamental resonant frequency. Alternately, a pulse duration of \((10)^{-5}\) sec can be employed with an intensity of \((10)^{3}\) w/cm\(^2\) if a temperature rise of 0.25°C is acceptable for measurement purposes. It should be noted in this connection that operation with a probe having a thermoelectric power level of 50 μV/°C results in an output of 12.5 μV for a temperature increment of 0.25°C, and this voltage level can readily be detected with presently available electronic equipment.

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Heat conduction in the thermocouple leads, as it affects the resolution of the instrument, is another factor which must be considered. This results in a limitation on the maximum diameter of the leads in the neighborhood of the junction. From the analysis in reference 7, the fractional decrease in the temperature at the site of the junction is given approximately by

\[
\frac{\delta T}{\Delta T_m} = -\frac{3a_0^2}{2} \left( \frac{K_w}{K} \right) \left( \frac{f'(a)}{h(a)} \right)_{a=0} \tag{8}
\]

where \( \delta T \) is the difference between the temperature of the absorbing structure and that of the thermocouple junction resulting only from heat conduction away from the junction by the wires, \( \Delta T_m \) is the temperature rise in the absorbing structure in the absence of the thermocouple junction, \( K_w \) is the coefficient of thermal conductivity of the wire, \( a_0 \) is its radius and the other symbols have been previously defined. Using numerical values previously employed and a value of \( K_w \) characteristic of iron and assuming that \( \frac{\delta T}{\Delta T_m} \) should be no greater than 0.5 (i.e., choosing it equal 0.5), yields

\[
a_0 = 0.1 \Delta a \tag{9}
\]

If a resolution of one micron is desired, then \( a_0 = 0.1 \mu \) and the diameter of the iron wire in the immediate neighborhood of the junction should be no greater than 0.2 \( \mu \). This value, which determines the area of cross section for thermal conduction, suggests the use of a supporting wire with a much lower coefficient of thermal conductivity and of much larger cross section (e.g., a fused silicon filament of 1 \( \mu \) diameter) upon which a thermocouple of appropriate dimensions is evaporated. The length of the thin evaporated layers should be no greater than a few microns in order to achieve a low value for the electrical resistance. Therefore, tapering of the evaporated layer to a much greater thickness should be accomplished as close to the junction as is consistent with the thermal conduction losses.

The present electronic instrumentation, which provides sufficient power for spectroscopy, includes a commercial rf power oscillator, for driving the piezoelectric element. Coupling to the transducer is accomplished by a 50\( \Omega \) coaxial cable and appropriate impedance matching units, Figure 4. (4) Two methods of electrically exciting X-cut quartz, at high, odd
harmonic frequencies, to radiate sound waves in liquids in the ultra high frequency region have proved successful. One method utilizes one or more short sections of coaxial transmission line to obtain an electrical impedance match between the quartz plate assembly and the 50-Ω line to the power oscillator. In the second method, the quartz plate is placed in the reentrant structure of an appropriately designed electromagnetic cavity. While both methods have been used to excite the piezoelectric plates to produce sound amplitudes of sufficient intensity in liquids for spectroscopic studies (maximum intensities in the range from 0.1 to 1 w/cm²), the fixed dimension cavity method has the inherent disadvantage of being restricted to a narrow frequency range of operation by the electromagnetic Q of the unit.

For detecting the voltage generated by the thermocouple probe (approximately 10Ω resistance) a DC chopper amplifier with a noise level of 0.005 microvolt is currently employed. The frequency response of the amplifier is down approximately 6 db at 10 cps. The output from the amplifier can be fed into an oscilloscope with a calibrated scale on the face for accurate visual reading of spot deflection or the pulse output from the DC amplifier may be recorded. The amplitude stability of the driver and the reproducibility of the duration of the output pulse as well as the gain of the detector amplifier and the recording or presentation system are important in determining the accuracy with which absorption coefficients can now be measured. These same factors will be of equal importance in microscopy-bearing on the resolution of structure in the specimen and preventing artifacts, i.e., apparent detail in the detected field distribution not corresponding to structure in the specimen. This latter would be especially important when a single probe (or a small number of probes) would be used to determine the transmitted field distribution at a large number of sites. Here, drifting of the system gain or of the acoustic energy per pulse might be the cause of mistakenly ascribing structure to the specimen. The overall stability of the present system described in terms of the variation in deflection indicated by the recording system in response to successive pulses of acoustic energy, for a fixed position of the probe in the field, is approximately ± 1%.

Electrical control equipment is provided for sensing and maintaining the temperature of the liquid in the chamber at a desired fixed value. The value of the ultrasonic absorption coefficient changes with the temperature and for the highly
absorbing liquids that have been studied at this laboratory the percentage change with temperature is comparable to the percentage change in the viscosity with the temperature. (The acoustic absorption coefficient is directly proportional to the viscosity for a true Stokes' liquid, however, few liquids exhibit such a behavior, especially if measurements over a wide frequency range are considered, see Fig. 5.)

Figures 5 and 6 illustrate the type of measurements that can be made when the instrument is operated for spectroscopic purposes. Figure 5 shows absorption data, obtained by the method described in this paper, for a silicone fluid to 2 kHz. (4) Figure 6 shows absorption data for castor oil at 30°C obtained by the transient thermoelectric method, together with data obtained by other investigators employing different methods. (4) It is seen that the transient thermoelectric method is capable of determining amplitude absorption coefficients at least as high as 1500 cm⁻¹.

III. FUTURE DEVELOPMENT

As already indicated, because of both expediency and inherent interest, the course of development of an ultrasonic absorption microscope is being pursued by the authors and collaborators through the stage of evolving instrumentation for determining ultrasonic absorption coefficients of liquids over a spectrum of frequencies from values near a megacycle per second into the kilomegacycle per second region. The instrumentation for this purpose is similar, in many features, to that required for ultrasonic microscopy. Although operation has been achieved and measurements have been made over an appropriate frequency range, a number of major instrumentation design and/or construction tasks must be consummated before a practical ultrasonic microscope is realized. These include: (1) transducers to operate at higher intensities, (2) probes of smaller size for achieving the desired resolution, (3) probe arrays for simultaneous detection of acoustic energy at a number of sites, (4) systems for moving the specimen with respect to the probe array while maintaining constant, close, spatial separation, (5) pulsed wide band driver for exciting the transducers to make possible high resolution ultrasonic spectroscopy, (6) presentation systems for making the ultrasonic absorption information accessible in visual form. These problems will now be considered in the order listed.
Calculations included in the previous section of this paper indicate that micron resolution can be achieved by employing short pulses of ultrasound [(10)^-5 sec duration] at intensity levels of approximately 1000 w/cm². Calculations further indicate that this level can be radiated directly from the face of X-cut quartz without exceeding the tensile strength\(^{(9)}\) (direct radiation at 200 w/cm² is in routine use in other work in progress at this laboratory). Therefore, extension to the 1000 w/cm² level does not pose any anticipated difficult problems. Perhaps the method of supporting the crystal will require modification to eliminate or prevent any unessential stressing of the crystal plate. Heating due to acoustic energy dissipation in the holder presents no problem if the duty cycle is sufficiently low since the total acoustic energy per pulse at the required levels is not greater than that currently employed in the absorption work.

To achieve a resolution of 1 micron using the same thermoelectric materials will necessitate a considerable reduction of the probe diameter (or cross section of the metallic conductors) and this might be accomplished by evaporation methods as indicated above. The construction of probe arrays for simultaneous accumulation of information at a multiplicity of sites per acoustic pulse will require new fabrication methods. At the present stage of instrument development it appears that a linear array of probes would constitute a particularly useful arrangement. Such an array would sample, for each acoustic pulse, the sound transmitted through a series of sites along a narrow band of the specimen. The width of the band would be made equal to the limit of resolution of the system, and the structural detail detected along the axis of the band, per acoustic pulse, would be determined by the spacing distance between adjacent probes. Movement of the array along the axis of the band - a position for each of a series of pulses - would provide information up to the resolution limit of the system in this direction. Movement of the array in a direction at right angles to the band axis would provide the sequences of information gathering steps required to obtain a complete two-dimensional picture of the structure of a thin specimen.

The scanning method just described requires that either the probe array move or the specimen be moved in a series of precisely determined sequential steps. Since it appears more practical, as indicated above, to maintain each probe at a fixed position in the sound field generated by the source, a system must be provided to move the specimen in its plane while
maintaining a close spacing distance between the specimen and
the array in order to prevent loss of resolution. The mainte-
nance of the probes in fixed positions in the field provides for
a constant acoustic path length between the piezoelectric
element and the probe as required to prevent variations in the
level of the detected field strength due to differences in the
absorption path. If one-micron resolution is desired, then the
junctions of probes should be within a distance of one micron
from the specimen surface. Such close spacing might best be
achieved by embedding the junctions in a solid absorbing mate-
rial with the junctions within one micron from the surface. The
specimen would then be placed in direct proximity with the
surface of the probe embedding assembly, with a very thin
liquid coupling medium interposed. The small displacements
of the specimen probably can be accomplished best by employ-
ing piezoelectric displacement transducers to move the speci-
men in two mutually perpendicular directions. Such a system
would readily provide movement in micron steps with the essen-
tial accuracy and reproducibility.

The electronic equipment for providing the excitation
for the crystal plates should furnish the requisite pulsed power
over a wide range of frequencies since much information is to
be gained by determining the absorption spectrum of the speci-
men. The output from this equipment must be synchronized
with the movements of the system which supports the specimen
and with the events which control the presentation system.

The presentation system includes the amplifiers neces-
sary to detect the output from the thermocouple probes. A
sensitivity to the millimicrovolt level (on long pulses - 0.1 sec)
is desirable. Obviously, the higher the sensitivity, the lower
the sound intensity of the pulses need be to detect a given change
in the absorption. For the single probe configuration, only one
high sensitivity amplifier need be included. However, incor-
poration of a probe array would probably entail the use of a
series of amplifiers since storage and sampling of signals at the
low levels of interest here is not feasible at present. Since a
multiplicity of appropriate amplifiers would represent a con-
siderable expenditure of funds, the first instruments would
include only a single probe and amplifiers. Since scanning of a
complete specimen would require an extended period of time
for a single probe instrument, the two-dimensional presentation
would probably be accomplished best by first storing the infor-
mation and second subtracting off a constant value from all
levels of probe output, to provide for contrast on the two
dimensional representation. The latter could then take the form of a photograph produced by exposure to a moving light focus with intensity proportional to the amplitude differences.

IV. CONCLUSION

The work already accomplished on instrumentation development and the successful measurements of absorption in high absorption liquids (at kilomacycle frequencies) supports the view that practical ultrasonic instruments to resolve structure of thin specimens on a microscale can be developed along the lines indicated in this paper. Such instruments would also furnish information on identification of structural type since examination of the specimen over a wide spectrum of frequencies would be possible.

V. ACKNOWLEDGEMENT

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LIST OF REFERENCES


Discussion

ULTRASONIC MICROSCOPY

Paul Maycock: Is the DC chopper system a modification of the Doffney chopper?

William J. Fry: I am not sure I recognize that particular classification. It's a commercial amplifier.

Paul Maycock: You didn't build the chopper?

William J. Fry: No, we didn't build the chopper.

Paul Maycock: I am intrigued by your idea of the film deposited thermocouple. Have you tried it?

William J. Fry: No, we haven't made any thermocouples in that fashion.

Leonard Goodman: Has any correlation been evidenced between acoustic properties and thermal conductivity properties?
William J. Fry: The question is asked whether thermal conductivity can be correlated with acoustic properties. Of course one can compute a value for the absorption coefficient from a value for the viscosity. For the data that we showed on the slides, the absorption values for castor oil deviated somewhat from the classical Stokes' absorption values calculated this way. Thermal conductivity also enters into the mechanism of absorption, but in the case of nonmetallic liquids, only a small fraction of the ultrasonic absorption is involved with the heat conductivity. Now mercury, on which data were reported at frequencies as high as 1 kMc in about 1947, is a material such that the thermal conductivity coefficient can be used to calculate quite closely the observed value of the absorption. However, for the liquids on which data are reported here, the conductivity of the mechanism contributes negligibly to the absorption. In the case of the silicone oil for which the ultrahigh frequency dependence of the absorption coefficient was illustrated, it is apparent that a relaxation frequency is present. We haven't correlated the observed relaxation frequency with the structure of the molecule, and, in fact, the samples we are using aren't uniform in terms of molecular size. At present the manufacturer is setting up to fractionate the oil to obtain samples with a much narrower distribution of molecular sizes. One might hope to correlate measurements on such material with features of the structure.

A Participant: Have the infrared detector methods been considered as contrasted with thermoelectric methods?

William J. Fry: We haven't considered this. I'm not sure exactly how we could accomplish this at single sites in the field. It would appear that such a method would have the same limitations that characterize the optical method using visible light to detect refractive index changes--one observes the average effect of the field distribution along a line transverse to the direction of propagation. In addition to the variation in the sound intensity across the field, there is the additional difficulty that the entire transmission path is only a few thousandths of an inch long at the higher frequencies.

Donald W. Oplinger: Are there any diffraction or beam-spreading effects that bother the thermocouple method?

William J. Fry: I think the simplest way to answer the question is to compare the size of the radiator, which is a centimeter or two in diameter--that is, of the order of 10,000
microns, and the length of the path from the crystal face that the sound transverses before essentially complete absorption—of the order of 100 microns. The path length is thus about 1/100 the diameter of the source. The absorption coefficient is so high that, as far as the axial field distribution is concerned, the diameter of the crystal is essentially of infinite extent. There is negligible contribution of acoustic energy to the field along the central axis from the outer or rim area of the source. Of course for lower values of absorption, (lower frequencies or less absorbing liquids) one must separate the effect of diffraction from the effect of absorption on the field distribution.

Paul Edwards: Have you considered thermistor elements as detectors in place of thermocouple elements?

William J. Fry: We have thought about it, but we are unfamiliar with any methods for making the thermistors as small as thermocouple junctions. Perhaps the questioner has some method of accomplishing this. We utilize 5-micron diameter junctions now, but it would be nice to have the thermistor sensitivity; however, we don't know of any way of making them small enough.

John P. Scheuer: What is the diameter of the wire at the junction of the temperature-sensitive element?

William J. Fry: The wire is etched down from 5 or 10 thousandths of an inch, and a butt junction is made so that on completion one has in the junction region a smooth wire of about 5 microns diameter (the minimum size we have accomplished thus far). The etching is done by employing a small loop of platinum wire to support a drop of acid. Under microscopic observation this is introduced over the tips of the individual wires, and the etching process is controlled to produce the desired taper. The junction itself is constructed by employing micromanipulator movement to bring the two ends of the wires into mutual proximity. A convenient way to achieve the butted contact is to produce a potential difference between the wires and let electrostatic attraction achieve the desired results.

As you know, for iron-constantan the temperature sensitivity is about 50 microvolts per degree. I can't produce off-hand a very precise estimate for the maximum temperature sensitivity of the thermocouple-amplifier system. However, I do recall computing an "ultimate sensitivity," for 0.1-second pulses, on the basis of signal amplitude equal to noise. The
value was somewhere between 1/10000 and 1/100000 degree with the present system.

A Participant: Degrees centigrade?

William J. Fry: Yes. It is safe to state that one could readily detect 1/10000 of a degree.

A Participant: Using iron-constantan or copper-constantan?

William J. Fry: We use copper-constantan now; however, we have used both types. Iron is somewhat inconvenient to use because rust spots develop in its surface, and these affect the etching process so that uniform tapering is difficult to achieve. So, although the sensitivity of the copper-constantan couple is somewhat lower, the combination is a better choice for such small diameter wires if exposure to air, water vapor, etc., is envisioned.

Nicholas Grossman: Do you see any applicability of ultrasonic microscopy to metals?

William J. Fry: I would say that one certainly can envision an applicability, but the choice of metals at the start is probably not the best because of the high thermal conductivity which limits resolution. I think the method is applicable, but I would rather examine nonmetallic materials while developing the system.

Harold Dunegan: Do you assume the energy distribution to be constant over the crystal face?

William J. Fry: One can study the acoustic field distribution by moving the probe at right angles to the direction of propagation, and it is not uniform over the entire crystal face. The distribution can be determined with liquids of relatively low absorption characteristics in the chamber, and this is what we do. It is of interest to note here that we have found it no more difficult to generate "usable" sound at harmonics hundreds of times the fundamental thickness resonant frequency, that at say the third or fifth harmonic.

Donald W. Oplinger: Is it desirable to coat the thermocouple junction in order to increase the sensitivity, since one might choose a material of higher absorption coefficient value than that which one would be using as the coupling medium?
William J. Fry: I would like to comment on this question as follows. At low frequencies this is precisely what one does. We use the thermoelectric probes by imbedding the junction in a liquid with a high absorption coefficient value compared to that of the liquid in which the sound is propagating. This is accomplished by suspending the thermocouple junction in a disc of material, usually of circular symmetry. The dimensions of this disc might be, for example, several inches in diameter so that the cross section of the acoustic field is smaller in size than the disc area, and, since the characteristic impedance of the medium in the disc can be chosen to match the impedance of the medium containing the field, the only possible source of appreciable disturbance to the field is presented by the thermocouple junction and supporting wires. These are, as already noted, quite small so that, for example, in the frequency range one to ten megacycles where this method is employed, the junction and neighboring wire diameter are 1/100 wavelength or less. This method is thus very useful for examining the fine structure of ultrasonic fields without introducing disturbances caused by the probe. For work at frequencies in the kilomegacycle range, coupling liquids that one would normally choose have such enormous absorption coefficient values that it is unlikely one would want to imbed the thermocouple junction in a second medium except for the possible advantage that one might realize in ultrasonic microscopy by using a solid material in order to achieve and maintain a close spatial interval between the specimen and the probe or probe array. In this latter case it would be desirable in addition to choosing a material with a higher absorption coefficient value to match the acoustic impedances.

Newbold O. Cross: What solid materials have you studied and what thicknesses are appropriate?

William J. Fry: We haven't studied any solids. What we have done is evolve the instrumentation thus far while confining our attention to measuring the absorption coefficients of liquids, since this represents an easier task than the microscope application. As I indicated earlier, when one doesn't have funds for one job, he can sometimes think of a second which might contribute to the first, and this is what happened. So the liquids are still being studied because we have no money for the solids. As it turned out, we have produced some extremely useful results which will be directly applicable to the microscope applications and to measurements on solids. Now, with respect to the question of appropriate thicknesses--
in the case of liquids we are concerned with thickness of the order of 10 to 100 microns. For a high absorption liquid measured in the kilomegacycle frequency range, a path length of ten microns is sufficient distance to move the probe to obtain an accurate measure of the absorption coefficient. In the case of ultrasonic microscopic examination of a solid, one would choose a thickness which is determined by the size of the structure elements to be resolved. The thickness of the specimen should not be much greater than the diameter of the structural elements that are to be detected within it. Otherwise, the adjacent probe would not detect the result of absorption in a single structural element but would respond to a transmitted signal complicated by contributions from a number of elements and would thus not resolve structure at the level of interest.

Harold A. Thomas: What sort of time constants are characteristic of the detection equipment as limiting the ultimate capability?

William J. Fry: The thermocouple junctions themselves have time constants considerably shorter than the electronic equipment which is presently employed. For example, junctions 10 microns in diameter have time constants between 1/10 and 1/100 of a second. However, I should point out that the temperature change indicated by the thermocouple results from more than one mechanism, and these are characterized in general by different time constants. For example, in addition to heating caused by absorption in the body of the medium, viscous force action results because the wire does not move with a displacement amplitude equal to that of the imbedding field, and heat is thus produced in the immediate neighborhood of the wire boundary. The alternating heat transfer at the wire interface, which results because a sound wave is accompanied by temperature cycling at the frequency of the field, also causes some acoustic energy to be transformed into heat at the thermocouple junction. However, for nonmetallic liquids the viscous mechanism resulting from relative motion between the thermocouple leads and the medium itself is much more important. For an operating frequency of 1 Mc and for a pulse length of one second, the viscous and body absorption mechanisms make approximately equal contributions to the observed temperature increments for the oils discussed here.

The response of the electronic equipment that we currently use drops 3 db in 0.1 second. One might consider employing wider band equipment and shorter pulse lengths.
rather than simply using the electronic equipment as an integrating device. One amplifier that we have employed with these probes is a transistor type in so-called "hushed transistor" operation. It provides a 60-kc band width (DC to 60 kc) at a noise level of about half a microvolt. I should also like to emphasize that the values on resolution that I quoted in the lecture were consistent with the present capability of these electronic instruments.
FIGURE 1. SCHEMATIC REPRESENTATION OF ULTRASONIC MICROSCOPE SHOWING TRANSDUCER

The transducer is excited to produce pulses of ultrasound in the coupling medium, the specimen under examination which is movable in the coupling liquid, and the thermocouple probe array whose junctions are immediately adjacent to the specimen. The figure illustrates motion in only one of the two mutually perpendicular directions in the plane of the specimen.
FIGURE 2. SCHEMATIC DIAGRAM OF MINIATURE THERMOCOUPLE FOR DETECTING UHF ACOUSTIC WAVES IN LIQUIDS
FIGURE 3. CYLINDRICALLY SYMMETRICAL STRUCTURE ELEMENT IN SPECIMEN

FIGURE 4. BLOCK DIAGRAM OF INSTRUMENTATION FOR GENERATING AND DETECTING UHF SOUND IN LIQUIDS
FIGURE 5. ACOUSTIC AMPLITUDE ABSORPTION COEFFICIENT OF A SILICONE FLUID VS FREQUENCY. THE DASHED LINE IS THE STOKES LOW-FREQUENCY ABSORPTION. THE DATA ILLUSTRATES A SINGLE RELAXATION PROCESS CENTERED AT 40 MC
FIGURE 6. ACOUSTIC AMPLITUDE ABSORPTION COEFFICIENT OF CASTOR OIL VS FREQUENCY. THE DASHED LINE IS THE STOKES LOW-FREQUENCY ABSORPTION.