Ultrasonic Characterization of High $T_c$ and Other Unconventional Superconductors

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Ultrasonic techniques were used to characterize the properties of high $T_c$ and other unconventional superconductors. A pontoon technique was developed to investigate thin films with surface acoustic waves SAW. A cw sampling technique was modified in order to sensitively study vortex motion in small superconducting single crystals of YBCO. The penetration field of melt textured YBCO was observed ultrasonically and analysis of the data yields the superconducting energy gap in its domain boundaries. Phase diagrams of superconducting UPT$_3$ were determined ultrasonically. The effect of a coherence phase transition on ultrasonic attenuation in CeCu$_6$ was measured. A theoretical model was developed to quantitatively explain the decrease in insertion loss at the superconducting transition of a SAW delay line made with YBCO interdigital electrodes. Two sets of attenuation and velocity signatures were observed in a large single crystal La$_{0.88}$Sr$_{0.15}$CuO$_4$ which correspond to the superconducting transition and to a Neel transition in the superconducting state. SAW measurements using the pontoon technique on single crystals and thin films of YBCO demonstrate the importance of pinning sites in determining the nature of vortex transitions. A two dimensional theoretical model was developed for determining the acoustic radiation impedance of resonating rectangular parallelepipeds. The effect of a surrounding medium on the resonance spectra of a vibrating solid sphere was derived theoretically, and the theoretical results compare favorably with experimental data.

Subject Terms:
High $T_c$ superconductors, YBCO, UPT$_3$, CeCu$_6$, La$_{0.88}$Sr$_{0.15}$CuO$_4$, ultrasonic techniques, surface acoustic Waves, SAW, acoustic radiation impedance, resonating solid sphere, cw sampling technique, vortex melting, vortex depinning

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I. REPORT DOCUMENTATION ABSTRACT

Ultrasonic techniques were used to characterize the properties of high Tc and other unconventional superconductors. A pontoon technique was developed to investigate thin films with surface acoustic waves SAW. A cw sampling technique was modified in order to sensitively study vortex motion in small superconducting single crystals of YBCO.

The penetration field of melt textured YBCO was observed ultrasonically and analysis of the data yields the superconducting energy gap in its domain boundaries.

Phase diagrams of superconducting UPt3 were determined ultrasonically. The effect of a coherence phase transition on ultrasonic attenuation in CeCu6 was measured.

A theoretical model was developed to quantitatively explain the decrease in insertion loss at the superconducting transition of a SAW delay line made with YBCO interdigital electrodes.

Two sets of attenuation and velocity signatures were observed in a large single crystal La1.85Sr0.15CuO4 which correspond to the superconducting transition and to a Neel transition in the superconducting state. SAW measurements using the pontoon technique on single crystals and thin films of YBCO demonstrate the importance of pinning sites in determining the nature of vortex transitions.

A two dimensional theoretical model was developed for determining the acoustic radiation impedance of resonating rectangular parallelepipeds.

The effect of a surrounding medium on the resonance spectra of a vibrating solid sphere was derived theoretically, and the theoretical results compare favorably with experimental data.
II. DESCRIPTION OF PROJECT

The objectives of this research project are to characterize the properties of high $T_c$ superconductors and other unconventional superconductors using ultrasonic techniques in order to provide insights into the mechanisms that are responsible for the unusual superconducting properties of high $T_c$ superconductors; and, in a grant expansion, to determine the effect of acoustic radiation impedance on the resonant frequencies of resonant ultrasound modes in order to evaluate third order elastic constants from gas pressure measurements.

III. SCIENTIFIC APPROACH TAKEN

Bulk acoustic wave attenuation and velocity measurements were performed on melt textured samples of $Y_1\text{Ba}_2\text{Cu}_3\text{O}_7$ and on single crystals of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$, $\text{UPt}_3$, $\text{URu}_2\text{Si}_2$, and $\text{CeCu}_6$.

Surface acoustic wave SAW measurements were performed on thin films of $Y_1\text{Ba}_2\text{Cu}_3\text{O}_7$. In some measurements, the interdigital SAW transducers were made of $Y_1\text{Ba}_2\text{Cu}_3\text{O}_7$. A pontoon technique was developed to launch surface acoustic waves across a single crystal platelet of $Y_1\text{Ba}_2\text{Cu}_3\text{O}_7$ and across thin films of $Y_1\text{Ba}_2\text{Cu}_3\text{O}_7$ deposited on non-piezoelectric substrates and on piezoelectric substrates which are too small to accommodate interdigital SAW electrodes.

A novel very sensitive resonance technique, a modified cw sampling technique, was developed for investigating small platelets single crystal samples of high $T_c$ superconductors.

The acoustic radiation impedance of resonating silica rectangular parallelepipeds and spheres was calculated and compared with the results obtained by the UCLA group for the quality factors and frequency shifts of different resonant modes of such samples.
IV. ACCOMPLISHMENTS

A. High $T_c$ Superconductors

1. Bulk Waves

   a. Melt textured $Y_1Ba_2Cu_3O_7$

   Changes in both attenuation and velocity have been observed at the penetration field $H'_c$ of a melt textured sample of $Y_1Ba_2Cu_3O_7$. This is the first time that such effects have been observed. The observation of such effects was made possible by the sensitivity achieved in our phase sensitive pulsed ultrasonic spectrometer which were better than .01 dB in attenuation and better than 1 ppm in velocity. Since the value of $H'_c$ at a particular temperature was independent of magnetic field orientation with respect to the c axis, it was concluded that $H'_c$ should be associated with the penetration field of the domain boundaries in the melt textured sample, which has the c-axis of 95% of its domains aligned within 5° of each other. As should be expected, $H'_c$ has a parabolic temperature dependence. If it is assumed that the density of flux lines penetrating at $H'_c$ is proportional to $H'_c$, then it is found that the change in velocity observed at $H'_c$ is inversely proportional to the separation between flux lines; and, therefore, proportional to the repulsive force between flux lines. Since this model appears to be self consistent, it was applied to the changes in attenuation. It was found that the change in attenuation per unit flux line observed at $H'_c$ has a maximum value around $t = T/T_c = 0.83$, where $T$ is the temperature, $T_c$ the superconducting transition temperature and $t$ is the reduced temperature. This immediately suggests that a relaxation mechanism is responsible for the attenuation; and, therefore, a relaxation time may be extracted from the data. The activation energy obtained from the low temperature part of the data is equal to $1.76 \pm 0.2 \text{kT}_c$. This value of $\Delta$ is surprisingly close to the BCS superconducting energy gap. Therefore, it is proposed that the relaxation time is associated with
the motion of flux lines induced by the sound waves. If one assumes that a flux line consists of two parts, a normal magnetic flux line core and the part of the vortex line composed of the superconducting quasi-particle screening currents, then it is possible that a sound wave will move one part with respect to the other. Thus, when the magnetic flux line moves, it leaves behind a part of the normal core which has to become superconducting and encompasses superconducting quasi-particles which have to become normal. The activation energy for this process would just be the superconducting energy gap. If it is further assumed that the relaxation time is controlled by electron-phonon interaction, then it is possible to evaluate the temperature dependence of the superconducting energy gap from the temperature dependence of the inverse of the relaxation time, as shown in Fig. 1, and to determine a value of $10^{11}$ secs for the relaxation time in the normal state. The temperature dependence of the energy gap follows the BCS relation. These results are surprising, since they appear to indicate that a BCS like model could account for the superconductivity in the high $T_c$ superconductors.

b. Sinter Forged $Y_{1}Ba_{2}Cu_{3}O_{7}$

The 180 K relaxation maxima that we observed in $Y_{1}Ba_{2}Cu_{3}O_{7}$ compounds and its isomorphs, with longitudinal waves propagating along the c-axis, appear to be produced by the motion of the CuO planes with respect to each other. It was determined that the excitations producing these relaxation maxima have energies in the range of $1100 \pm 200$ K. These are the energies that most theoretical models require for the excitations that produce Cooper pair coupling and which may be responsible for the high $T_c$ of these superconductors. Furthermore, recent theoretical models suggest that the interactions responsible for the superconductivity in these systems are interplanar, originating in the reservoir of electrons between the CuO planes. Thus, it may be possible that the excitations producing the 180 K relaxation maxima are also the ones
Figure 1(a): Temperature dependence of the ratio of the relaxation time due to electron phonon interaction in the normal state $\tau_{\text{ep}}$ to the relaxation time in the superconducting state $\tau_s$. The solid line is a plot of $\tau_{\text{ep}}/\tau_s = 2/(e^\Delta/kT+1)$ which is obtained by assuming that the relaxation time is inversely proportional to the attenuation coefficient due to electron phonon interaction.

(b) Temperature dependence of the superconducting energy gap $\Delta$ obtained from the above equation and the data in 1(a). The solid line is the BCS temperature dependent energy gap with $\Delta(0) = 1.76 \, kT_c$. 
which are involved in the interaction which produces the high $T_c$'s.

c. Single Crystal $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$

Ultrasonic attenuation and sound velocity were measured in a single crystal of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ with a superconducting transition temperature of 37K that was obtained from Professor K. Kitazawa, University of Tokyo. Longitudinal sound waves were sent along the c-axis of the crystal at frequencies ranging from 25 MHZ up to 305 MHZ. As a function of temperature, a small peak was observed at around 27 K and a large peak at around 37 K. These two peaks, especially the small one, became more obvious at the higher frequencies. Simultaneously the velocity exhibited a drop of about 100 ppm below the superconducting transition and a dip at around 27 K. From the shape of the velocity changes, it is possible to deduce that the 37K transition involves a quadratic coupling between the order parameter and the strain, which would be expected for a superconducting transition. The 27 K transition involves both quadratic and linear coupling. The behavior of the peak in attenuation at 27 K is consistent with a relaxation mechanism associated with antiferromagnetic fluctuations which occur above an antiferromagnetic transition. Measurements in a magnetic field of 1.1 Tesla appear to indicate that there is a magnetoelectric interaction threshold at about 200 MHZ wherein the interaction abruptly increases above this frequency.

2. Surface Acoustic Waves and SAW Pontoon Technique on $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$

The pontoon technique, wherein a single crystal platelet is placed as a bridge between two $\text{LiNbO}_3$ substrates overlaid with SAW interdigital electrodes, was used to measure the attenuation of 168 MHZ SAW in an untwinned single crystal of YBCO with a superconducting transition temperature $T_c = 88$ K. A peak in attenuation was observed at $T_c$ which shifted to lower temperatures and became more pronounced in a magnetic field. This peak in attenuation could
be associated with a melting transition in the flux line lattice in the vortex state of the superconductor.

The pontoon technique was also used to investigate, with 250 MHZ SAW, YBCO films deposited on LiNbO$_3$ substrates, which were too small to have interdigital electrodes deposited directly on them. Again, a strongly field dependent attenuation peak was observed below Tc.

Since the film had a large density of vortex pinning sites, the behavior of the observed peak in attenuation for different applied constant magnetic fields, as a function of temperature, was consistent with a depinning transition, since the magnitudes of the peak depended on the square of the applied magnetic field. This dependence is expected from thermally activated flux flow models, where the attenuation can be shown to be proportional to the shear modulus of the vortex system which is proportional to the square of the flux density.

The field dependence of the magnitude of the peaks observed in the untwinned single crystal was larger than quadratic which may imply that there is increased defect motion in the vortex system which could be associated with a melting transition. Thus, these two sets of measurements demonstrate the role played by pinning sites in determining whether a vortex system will undergo a depinning transition, a melting transition, or both, depending on the density of pinning sites.

Surface acoustic wave SAW measurements were made on a granular thin film of YBCO. The film is modeled as a two-dimensional network of Josephson junctions. A percolation model developed in our group to describe the SAW attenuation was modified to take into account the resistivity of the grains in the normal state, and a reasonable fit to the data was obtained.

Both the pontoon technique and conventional 164 MHZ SAW delay lines were used to study YBCO films around 200 K. Abrupt changes in SAW attenuation and velocity are observed
at around 200 K on cooling and at around 230 K on warming. In order to further investigate these phenomena, a capacitive technique was developed, wherein interdigital capacitors deposited on a glass substrate are brought into close proximity of the films, and the electromagnetic reflection coefficient at 260 MHZ and 400 MHZ of the capacitors is measured as a function of temperature. Changes in the dielectric constant of the YBCO films are observed at 200 K and 230 K. These effects may be produced by a first order phase transition to a piezoelectric or ferroelectric crystal structure.

3. Modified cw Sampling Technique on Single Crystal Y\textsubscript{1}Ba\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7}

A new technique was developed for studying vortex motion in the superconducting state of an untwinned single crystal of Y\textsubscript{1}Ba\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7} whose dimensions are 1 mm by 1 mm by 50 μm. The crystal is brought into close proximity of a 5 MHZ x-cut quartz transducer. It is pressed by two 25 μm strands of varnish to the transducer, which is held by copper wires. This ensemble has a resonant frequency of 3 MHZ. The quality factor Q of the system is then measured as a function of temperature and magnetic field in the vicinity of the superconducting transition temperature. Quality factors as high as 10\textsuperscript{4} may be achieved in this system; and according to our theoretical model, the change in Q produced by a change in attenuation in the superconducting sample is proportional to Q. This is how the sensitivity of the measurements is enhanced. A sensitivity of 1 x 10\textsuperscript{4} dB/cm can be achieved for the system which translates to a sensitivity of 5 x 10\textsuperscript{3} dB/cm for the superconducting sample. The measurements were done with the magnetic field parallel to the ab plane of the crystal. The motion of the transducer was parallel to the c-axis. This resulted in a Lorentz force parallel to the ab plane so that the vortex motion was also along the ab plane. As a function of magnetic field at a constant temperature below T\textsubscript{c}, the attenuation was proportional to the magnetic field below 0.2 T, which indicates that the interaction
is proportional to the vortex density and therefore the interaction is, essentially, with individual, weakly interacting vortices, or a soft vortex system. At fields above 0.28 T the attenuation is proportional to approximately the cube of the applied field. This is probably due to the fact that with increasing vortex density the interaction between vortices becomes stronger leading to a quadratic increase in the viscosity of the system. Thus, at around 0.2 to 0.28 T there is a transition from a soft vortex system to a rigid, strongly interacting vortex system. Measurements very close to $T_c$, appear to indicate that slightly below the upper critical field the vortex lattice undergoes another transition from the rigid vortex system to the soft vortex system, as predicted by a recent theoretical model in a paper we published in Philosophical Magazine.

B. Heavy Fermion Systems

1. UPt$_3$

Phase diagrams in the superconducting state of UPt$_3$ were determined in our laboratory by using measurements of longitudinal waves. These are the first such phase diagrams obtained by a single measuring technique on the same sample. This is important because the theoretical models developed to account for the unconventional superconductivity in UPt$_3$ make different predictions about the existence of tetracritical points in the phase diagrams; and, therefore, measurements have to be consistent with each other within a few millikelvin. This requirement is difficult enough to achieve in a single apparatus with the same sample; it would be almost impossible on different cryostats with different samples when one considers that the transition widths of the best samples are about 10 or 15 millikelvin and the transition temperatures can vary by as much as 30 millikelvin.

The first set of ultrasonic measurements exhibited conclusively a tetracritical point when the magnetic field was applied perpendicular to the c axis. The data allow for the existence a
tetrical point when the magnetic field is applied parallel to the c axis. However, recent data obtained with high resolution pulsed ultrasonic velocity measurements with sensitivities greater than 200 parts per billion show that when the magnetic field is applied at 45° to the c-axis, the tetracritical points appear to separate into two tricritical points. At present none of the theoretical models proposed appear to be able to account for this development. However, theorists are modifying their models in order to include these new results.

Since the method of analyzing the data to obtain these phase diagrams is somewhat novel, a brief description follows.

In order to systematically determine the critical fields at which the transitions occur, the first order derivative of the velocity versus field was taken and is displayed as a function of field for several constant temperatures in Fig. 2. The two peaks in the derivative corresponding to the two transitions for the field sweep at 341 mK were fitted by two independent Gaussians; this set the height and width of the Gaussians, since the peaks are far apart. Then, the centers of the two Gaussians were shifted to best fit the data in the field sweeps at other temperatures. This was done while assuming that the height and width of the Gaussians were constant. The critical fields were positioned at the values of the centers of the two Gaussians. The resulting phase diagram from these and other measurements at constant field as a function of temperature is shown in Fig. 3.

Attenuation measurements in zero magnetic field as a function of temperature in the superconducting state were compared to the BCS relation for the ratio of the attenuation in the superconducting state $\alpha_s$ to the normal state $\alpha_n$,

$$\frac{\alpha_s}{\alpha_n} = \frac{2}{e^{h/kT_1} + 1} = 2f(\Delta)$$

(1)
Figure 2: The first derivative of the velocity versus the field. Two well-defined peaks correspond to the two transitions at $H_{c2}(T)$ and $H_{FL}(T)$. The two peaks for the field sweep at 341 mK were fitted to two independent Gaussians. Assuming the width and the height of the two Gaussians to be constant (which is clearly a good approximation as seen by the fit prior to the overlap of the two peaks), the center positions of the Gaussians were then varied to best fit the data for field sweeps at other temperatures. The two critical fields $H_{c2}(T)$ and $H_{FL}(T)$ are associated with the center positions of the two corresponding Gaussians.
Figure 3: The superconducting phase diagram of UPt$_3$ for $H \wedge c = 45^\circ$ determined from velocity measurements, plotted in the region where multicritical points are located. It is unambiguously determined that the $H_{FL}$ line terminates on the $H_{c2}$ line at a point where a kink is observed in the $H_{c2}$ line. The $H(T_c)$ line appears to end on the $H_{FL}$ line at a slightly different location, resulting in two tricritical points, rather than one tetracritical point which is found in the phase diagram for $H \perp c$. The inset shows the full phase diagram for $H \wedge c = 45^\circ$. 
where $k$ is Boltzman's constant, $T$ is the temperature and $\Delta$ is the superconducting energy gap, which may be a function of $\theta$, the angle with respect to the c-axis. If it is assumed that in the hydrodynamic limit, the energy dissipation for electrons traveling along a particular direction, $\theta$, is given by $f(\Delta(\theta))$ then the attenuation may be obtained by integrating the properly weighted $f(\Delta(\theta))$ over all momentum space. If this is done, we find that the best agreement between our data and this model is obtained for an anisotropic energy gap, wherein the gap vanishes along an equatorial circumference of the Fermi surface, which is described as the polar state. These measurements, together with the superconducting phase diagrams previously described, confirm the unconventional nature of the superconductivity of UPt$_3$.

Even in the normal state, this heavy Fermion superconductor exhibits anomalous behavior. Longitudinal waves along the b axis show a peak in attenuation at about 10.5 K which does not move in temperature as the frequency is increased from 225 MHZ up to 850 MHZ. The value of the attenuation at the peaks is proportional to the frequency squared. The peak in attenuation appears to move to a lower temperature and its amplitude increases when magnetic fields up to 8 Tesla are applied. In order to understand interaction that produced this peak, further measurements were performed in high magnetic fields up to 23 T at 192 MHZ, from 2 K up to 35 K. The attenuation peak that had been observed at 10.5 K at zero field was found to shift to lower temperatures with increasing field up to about 17 T. Thus, this attenuation behavior is qualitatively consistent with an interaction with a narrow density of states peak, slightly above the Fermi surface, which is associated with the Kondo Effect. However, as the magnetic field is increased above 17 T up to 23 T the energy level of the localized magnetic moments crosses the Fermi surface producing a metamagnetic transition which results in the observation, for the first time, of a very large attenuation peak, associated with the crossing of the levels, which
overshadows the peak associated with the Kondo Effect.

2. \( \text{URu}_2\text{Si}_2 \)

Neutron diffraction measurements on \( \text{URu}_2\text{Si}_2 \) reveal antiferromagnetic ordering along the c-axis with a Neel temperature \( T_N = 17.5 \text{ K} \) and an ordered moment of 0.03 \( \mu_B \) along the c-axis. We have measured the temperature dependence of the longitudinal velocity of waves traveling along the a and c-axis and of the shear velocity of waves traveling along the a-axis and polarized along the c-axis. The behavior of the velocity appears to be very similar for the three modes. The velocity increases monotonically as the temperature is lowered. Cusps in the velocity are observed for the three modes at \( T_N \). The velocity appears to increase more rapidly below \( T_N \). There is a larger change below \( T_N \) for longitudinal waves propagating along the a-axis than for those along the c-axis.

The attenuation of longitudinal waves along the a-axis exhibits a peak at the superconducting transition which is suppressed by a magnetic field. The attenuation decreases below its normal state value at temperatures below the peak. A velocity change of about 100 ppm is observed below \( T_c = 1.4 \text{ K} \) for longitudinal waves traveling along the a-axis; but, no velocity change, to less than 2 ppm, is observed along the c-axis.

3. \( \text{CeCu}_6 \)

Although neither a superconducting transition nor a magnetic transition has been confirmed in \( \text{CeCu}_6 \) down to temperatures below 10 millikelvin, this heavy Fermion compound exhibits interesting ultrasonic properties.

Below 1 K, \( \text{CeCu}_6 \) undergoes a coherence transition, which may be viewed as electrons coherently scattering from the dense Kondo lattice. The electrical conductivity increases as \( T^{-1} \) below 1 K, and as \( T^2 \) below 200 mK. We have performed attenuation measurements and electrical
conductivity measurements on two samples of CeCu$_6$ obtained from the same boule. We found that the temperature dependence of the electrical conductivity goes as $T^{-1}$ and $T^{-2}$ as described above. However, over the same temperature range, the attenuation increases as $T^{-1/2}$ and $T^{-3/2}$. For conventional electron phonon interaction it is expected that the attenuation should be proportional to the electron mean free path, and therefore to the electrical conductivity. Thus, we have found a discrepancy of $T^{1/2}$ between the two sets of measurements. It may be possible that the coherent scattering of electrons below the Kondo temperature adds a decoupling factor between the electrons and the lattice which depends on $T^{1/2}$ and this may account for the $T^{1/2}$ difference between the electric conductivity and the attenuation measurements.

Above the Kondo temperature of CeCu$_6$, an ultrasonic attenuation peak is observed which moves up in temperature when the frequency is increased from 80 to 800 MHZ and whose amplitude at the peak is proportional to the frequency. Thus, this peak may be associated with a relaxation model wherein the attenuation is proportional to

$$\alpha = \frac{\omega^2 \tau}{1 + \omega^2 \tau^2} \quad (2)$$

where $\omega$ is the angular frequency and $\tau$ is the relaxation time. Since $\omega \tau = 1$ at the maximum, it is possible to evaluate the proportionality constant from the attenuation data and consequently the temperature dependence of the relaxation time. It was found that $\tau(T) = (T - T_o)^{1.75}$ where $T_o = 2$ K, which is at the lower range of the values reported for the Kondo temperature of CeCu$_6$.

C. Acoustic Radiation Impedance

1. Fused Silica Rectangular Parallelepiped

   By analyzing the Resonant Ultrasound Spectroscopy RUS data obtained by the UCLA group on a fused silica rectangular parallelepiped sample, it was found that the quality factor Q
of the resonant curves is inversely proportional to the pressure of the gas surrounding the sample, and decreases when the molecular mass of the gas is increased. These results are obtained after a constant background loss due to mechanical supports and electrical coupling is subtracted from the total loss, remembering that $Q$ is inversely proportional to the attenuation coefficient $a$. We have proposed a two dimensional radiation impedance model that fits some of these data either qualitatively or quantitatively.

The UCLA group is performing these RUS measurements in order to determine the third order elastic constants of minerals from the change in the resonant frequency of these samples produced by changing the pressure of the ambient gas. It is important to ascertain if the pressure dependence of the mass loading produced by the surrounding gas will change the resonant frequency by an amount which is comparable to that produced by the third order elastic constants. Since the frequency change can have at least two possible sources, it was felt that the $Q$ values were more accessible to direct comparison with a theoretical model. Therefore, the two dimensional radiation impedance model was developed to account for the pressure effects on $Q_r$, which is the part of $Q$ affected by radiation into the surrounding gas, and then to estimate the effect of mass loading on the resonant frequency of the sample.

In this model, the six surfaces of the resonating sample are approximated by rectangular pistons moving in an infinite baffle. The displacement of these surfaces is not uniform, but may be approximated by sine or cosine functions whose wavelength is equal to twice the dimensions of the rectangular surfaces. The surfaces will radiate sound into the surrounding gas at the resonant frequency of the sample. Since the velocity of sound in the gas is approximately an order of magnitude smaller than in the solid sample, the wavelength of the sound in the gas is approximately an order of magnitude smaller than the dimensions of the faces. Therefore, it is
reasonable to assume that the directivity pattern of the sound emanating from the surface is mainly in the forward direction; and, as a first approximation, it may be assume that the six surfaces may be treated independently.

Calculation of the radiation impedance in Cartesian coordinates involves a four fold integral, which Professors Sorbello and Beck, in our Physics Department, reduced to a two fold integral by a change of variables. And, by adding and subtracting a specially designed integral, they were able to eliminate the divergence that occurs at the origin of this integral. The final results for the resistive and reactive parts of the radiation impedance are obtained by careful numerical integration.

The theoretical results obtained properly predict the linear dependence with pressure of the inverse of the quality factor, $1/Q_r$. And, also, $1/Q_r$ increases when the molecular mass of the gas is increased, as found experimentally. However, quantitative agreement is found only for the torsional mode, and this only when He is the surrounding gas. And, the square root mass dependence for $1/Q_r$ that is predicted theoretically is only found experimentally for the compressional modes. It was also found that the effect of mass loading is about an order of magnitude smaller than the experimentally found shifts in resonance frequency produced by the increase in pressure. Therefore, according to this simplified model, third order elastic constants may be safely determined by the RUS pressure technique used by the UCLA group. A preliminary report of these results was presented at the Acoustical Society Meeting; and a full paper was submitted to JASA.

However, since the agreement between our model and the experimentally obtained $Q_r$ values is only qualitative in some instances, it is not yet definitively proven that mass loading is truly negligible.
An obvious modification to our two dimensional model would be to perform the calculation for a three dimensional sample.

The three dimensional calculation appears to be straightforward for a spherical sample, and the results will be discussed in the following section. Professor Beck has formulated a Green's function approach for calculating, in three dimensions, the acoustic radiation impedance of a rectangular parallelepiped. He is in the process of inverting the resulting complex matrix in order to obtain values of the acoustic radiation resistance and reactance as a function of the product of the wavevector in the gas, $k$, times the appropriate dimensions of the sample.

2. Fused Silica Sphere

Three resonance modes obtained by the UCLA group on a fused silica sphere were sufficiently separated from adjoining modes to warrant analysis. These modes correspond to the two lowest breathing modes, the spheroidal modes, $S_{00}$ and $S_{11}$, and the fundamental torsional mode, $T_{02}$. The fundamental mode $S_{00}$ consisted of a well behaved single peak. The inverse of the quality factor $Q_R$, obtained as described in the previous section, was proportional to the applied pressure of the gas and was also proportional to the square root of the molecular mass of the gas $M^{1/2}$. The frequency shift was linearly dependent on the applied pressure. The frequency shifts and $Q_R$ are shown in Fig. 4. The $S_{11}$ mode consisted of three peaks which behaved essentially the same as the $S_{00}$ mode, except that the quality factor was about four times higher.

The torsional mode $T_{02}$ exhibited four peaks, one very large, one barely distinguishable from the background and two quite distinct. The one with the largest amplitude consisted of two degenerate peaks, possibly the $m = \pm 1$ modes which have large displacements at the poles, and would be strongly coupled to the driving and receiving shear transducers. This accounts for their larger amplitudes, their broken degeneracy and the difficulty in obtaining reliable data from them.
Fig. 4

Frequency shifts $\Delta f$ and the inverse of the quality factor associated with radiation $Q_r$ plotted versus applied pressure for the spheroidal $S_{00}$ mode of a fused silica sphere. The isotropic pressure is produced by helium, nitrogen or argon gas. The theoretical lines in (b) are plotted without any adjustable parameters. The almost horizontal line in (a) identified as "Mass Loading $\Delta f$" is the theoretical result for $\Delta f$, which appears to be negligible compared to the experimentally observed frequency shifts.
The $T_{02}$ peaks and the degenerate modes are described in Fig. 5. Since these modes do not possess any radial motion, the only coupling to the surrounding medium would be through the viscosity of the medium which would lead to a square root relationship with pressure, as will be discussed later. After careful optimization of the sensitivity of their RUS apparatus, the UCLA group was able to obtain high enough Q's to extract this square root dependence as shown in Fig. 6b.

Richard Sorbello and Don Beck formulated the acoustic radiation impedance problem in spherical coordinates, and then Sorbello derived the real and imaginary components of the acoustic radiation impedance for the three relevant modes. In the process, Sorbello derived a universal relationship, a la Kramers Kronig, for the ratio of Q and the relative frequency-shift.

For the breathing modes, in the limit where the wavelength in the gas is much smaller than the dimensions of the sphere, Sorbello found that the attenuation $\alpha$, which is proportional to $Q_R^{-1}$, is proportional to $pc$ divided by the real part of the logarithmic derivative of the appropriate free space Hankel function evaluated at the surface of the sphere. Here $c$ is the velocity of sound in the gas, which is independent of pressure and is inversely proportional to the square root of the molecular mass $M$. The density $\rho$ depends linearly on pressure and $M$. Therefore the product is linearly dependent on pressure and $M^{1/2}$. This is what was found experimentally for the $S_{00}$ and $S_{11}$ modes. Sorbello also evaluated the factor which determines the slope of the lines drawn in Fig. 4b. It is apparent that the theoretical lines fit the data well.

An additional factor of $c$ is obtained for frequency shifts, so that these are proportional to $pc^2$ and therefore independent of the molecular mass of the pressurizing gas. The theoretical results and experimental values are shown in Fig. 4a. As may be seen in this figure, the frequency shift produced by acoustic radiation loading is negligible. Therefore, the frequency
Degenerate modes: \( n = 0, \ l = 2, \ m = 0, \pm 1, \pm 2 \)

\[ m = 0: \]
\[ u_\theta = 0 \]
\[ u_\varphi = A \sin 2\theta \cos \omega t \]

\[ m = \pm 1: \]
\[ u_\theta = A \cos \theta \cos (\omega t \pm \phi) \]
\[ u_\varphi = A \cos 2\theta \cos (\omega t \pm \phi) \]

\[ m = \pm 2: \]
\[ u_\theta = A \sin \theta \cos (\omega t \pm 2\phi) \]
\[ u_\varphi = \frac{1}{2} A \sin 2\theta \cos (\omega t \pm 2\phi) \]

Fig. 5
The torsional \( T_{02} \) modes of a fused silica sphere are plotted versus frequency. The relationships for the surface velocity of each of the degenerate \( T_{02} \) modes are shown on the bottom left hand side and the motion corresponding to the \( m = 0 \) mode is depicted on the bottom right hand side.
shifts observed experimentally must be due to the third order elastic constants, and some other effect which depends on the mass of the gas. The UCLA group has redesigned their RUS sample holder in order to try to eliminate this perplexing mass dependance.

The analysis of the torsional data was reassuring. As shown in Fig. 6, the frequency shifts observed for the three gases overlapped each other fairly closely. It is obvious that argon, which has a molecular mass of 40 as compared to 28 for nitrogen has a larger effect on the frequency than nitrogen.

Since the torsional mode has no radial motion, a model was set up for viscous losses, which resulted in an acoustic radiation impedance which is proportional to the viscous length. The resulting frequency shifts and energy losses are proportional to the square root of pressure. The theoretical results, are plotted in Fig. 6. Again, the mass loading effect on the frequency shift is negligible compared to the experimentally observed data. It does appear that a square root dependence is consistent with the quality factor data. Finally, it should be emphasized that the theoretical viscous loss model yields a reasonable fit to the quality factor data.
Fig. 6  (a) Frequency shifts $\Delta f$ and (b) the inverse of the radiation quality factor $Q_R$ plotted versus applied pressure. The mass loading effect on $\Delta f$ is negligibly small. The solid lines drawn in b are a plot of the theoretical square root dependence of $1/Q_R$ on the applied pressure.
OFFICE OF NAVAL RESEARCH
PUBLICATIONS/PATENTS/PRESENTATIONS/HONORS REPORT

01 Jan. 91 through 30 June 97

Contract/Grant Number: N00014-91-J-1211

Principal Investigator: Moises Levy

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a. Number of papers submitted to refereed journals but not yet published: 5
b. Number of papers published in refereed journals (ATTACH LIST): 21
c. Number of books or chapters submitted but not yet published: 3
d. Number of books or chapters published (ATTACH LIST): 12
e. Number of printed technical reports & non-refereed papers (ATTACH LIST): 12
f. Number of patents filed: 1
g. Number of patents granted (ATTACH LIST): 0
h. Number of invited presentations at workshops or professional society meetings: 13
i. Number of contributed presentations at workshops or professional society meetings: 39
j. Honors/awards/prizes for contract/grant employees, such as scientific society and faculty awards/offices (ATTACH LIST): 6
k. Number of graduate students supported at least 25% this year this contract/grant: 6
l. Number of post docs supported at least 25% this year this contract/grant: 2

How many of each are females or minorities? These six numbers are for ONR's EEO/Minority Reports. Minorities include Blacks, Aleuts, Amindians, etc., and those of Hispanic or Asian extraction/nationality. The Asians are singled out to facilitate meeting reporting semantics re “underrepresented”.

Graduate student FEMALE: 1 Post doc FEMALE: 
Graduate student MINORITY: 
Graduate student ASIAN E/N: 3 Post doc MINORITY: 
Graduate student ASIAN E/N: 1
Ultrasonic Characterization of High T_c and Other Unconventional Superconductors
Moises Levy

A. Papers Submitted to Refereed Journals But Not Yet Published.


B. Papers Published in Refereed Journals


C. Books or Chapters Submitted but not yet published


D. Books or Chapters Published


E. Printed Technical Reports and Non-Refereed Papers


F. Number of Patents Filed

Frequency Tunable SAW Filter and Dispersion Lines Using High T$_c$ Superconducting Films", H. P. Baum, B. K. Sarma and Moises Levy. (Final patent documents prepared by M. Levy and Irv Ross, an attorney and director of Technology Transfer at the Graduate School, University of Wisconsin-Milwaukee, and submitted to U. S. Patent Office by the UWM Graduate School, 1991).

G. Number of Patents Granted

None

H. Number of Invited Presentations at Workshops or Professional Society Meetings


I. Number of presentations at Workshops or Professional Society Meetings


32. "Preliminary Studies of Resonant Ultrasound Spectroscopy of Silicon and \(Y_{1.5}Ba_{2}Cu_3O_7\) Single Crystals", Hong Zhang, Mark J. McKenna, Carsten Hucho, Bimal K. Sarma and Moises Levy, Second Annual Meeting of the Consortium on Resonant Ultrasound Spectroscopy, held August 24 and 25, 1995 in Santa Fe, NM.


J. Honors/Awards/Patents/Presentations

1. Distinguished Lecturer, IEEE Ultrasonics, Ferroelectrics and Frequency Control Society, July 1991 to June 1992

2. Fellow IEEE Ultrasonics, Ferroelectrics and Frequency Control Society, 1995

3. Fellow Acoustical Society of America, 1996


5. Chairman IEEE Applied Superconductivity Committee, 1995 - present

6. Chairman 1st Resonant Ultrasonic Spectroscopy Workshop - 1994
K. Graduate Students Supported at Least 25% for at Least 1 year

Joseph Herro
Ron Gaffney
Hong Zhang

Shi Wan Lin
Jeff Feller
Debashis Dasgupta

L. Post Docs Supported at Least 25% for at Least 1 year.

Zheng Xiao Li

Mark McKenna
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