I. MICROWAVE SPECTROSCOPY[*]

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A. ULTRASONIC ATTENUATION IN SUPERCONDUCTORS

The work described here was initiated several months ago in the very high frequency range. Equipment was designed and constructed for the generation and reception of ultrasonic pulses at 165 mc. The transmitter consists of the high-frequency and blocking-oscillator portions of the circuit used by Chick, Anderson, and Truell.[1] The transmitting and receiving transducers are identical helical resonators[2] exciting x-cut quartz rods that are inserted in an rf electric field produced in a gap of nearly identical geometry to that of the re-entrant cavity used in the microwave phonon experiments.[3] These helical resonators are shown in Fig. I-1a. The two quartz rods (0.118 inch in diameter, 0.504 inch long) with a metal specimen (usually approximately 0.1 inch thick) sandwiched between their ends are inserted through a small hole in the end of each helical resonator into the electric-field gap. The resonators are then clamped in this coaxial configuration, mounted on stainless-steel coaxial lines, and immersed in a standard double-dewar helium cryostat. Signal power from the receiving transducer is mixed with local oscillator power in a strip-line hybrid coupler; the dual output is then passed through a balanced detector to a 30-mc, 120-db, if strip.

This apparatus has been used to study ultrasonic attenuation in specimens of indium and mercury, both in the superconducting and normal states. The metals were obtained from standard chemical reagent stock (In, 99.99 per cent pure; Hg, 99.999 per cent pure). No attempt was made to obtain single crystals with known orientation. In the case of indium, the quartz transducer rods were clamped in a vee-block and the indium was allowed to solidify in a Teflon mold.[4] The mercury specimen was contained in a small, cylindrical aluminum capsule; the quartz transducer rods had been fitted into aluminum bushings that were inserted into the ends of the capsule and cemented into place. (See Fig. I-1b.)
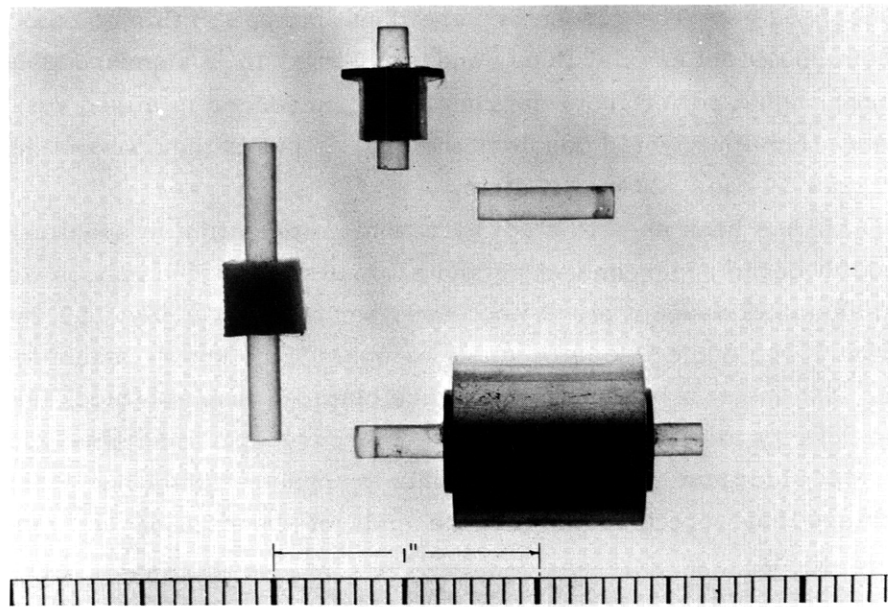
1. Experiment

In a typical experiment the entire transducer assembly containing the metal specimen is cooled to some fixed temperature well below the superconducting transition

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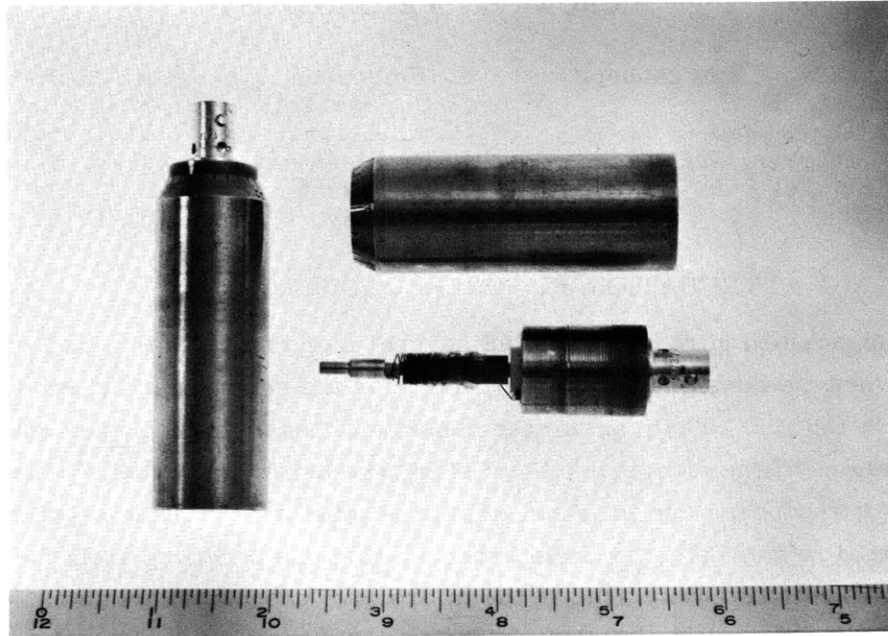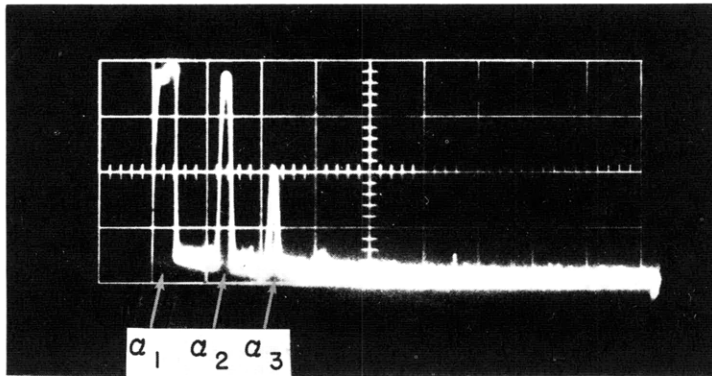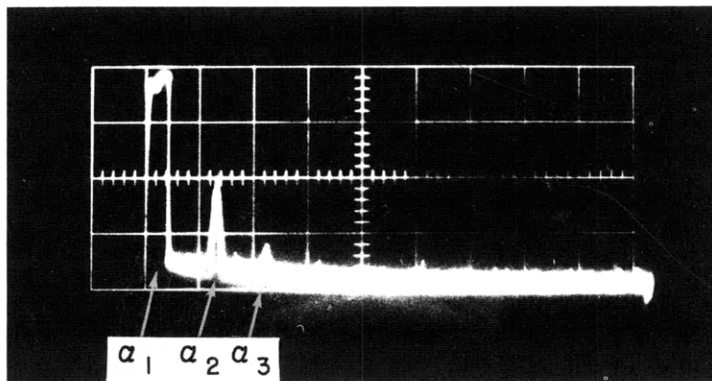
Fig. I-1. (a) Helical resonators used for the transmitting and receiving transducers.
(b) Mercury capsule and the indium mold. The quartz transducer rods are shown protruding from each.

(a)

(b)

Fig. I-2. (a) Acoustic pulses transmitted through superconducting
mercury. a_1 is rf leakage; a_2, an acoustic pulse that
has passed through both quartz rods and the mercury
specimen; a_3, superposition of two acoustic pulses,
one having made a double reflection in the first rod,
and the other, a double reflection in the second rod.
(b) Acoustic pulses transmitted through normal mercury.
This is exactly the same situation as in (a), except
that a magnetic field $H \gg H_c(T)$ has been switched on.

temperature. The transmitter initiates, in the transmitting transducer, a pulse of rf power of approximately 25 watts, peak, and 1 μsec in duration. A small fraction of this power ($\approx 10^{-3}$) is converted into a pulse of acoustic energy of longitudinal polarization by the piezoelectric effect. This pulse passes along the first quartz rod, through the specimen, and into the second quartz rod. At the receiving transducer the same fraction of the acoustic power is reconverted into an rf signal, and the rest is reflected. Figure I-2 shows the resulting oscilloscope traces. The first pulse is rf leakage. The second is an acoustic pulse that has passed through both quartz rods and the metal specimen. The third pulse is actually a superposition of two acoustic pulses: one has made a double reflection in the first rod; the other, a double reflection in the second rod. The superposition is caused by the fact that the two quartz transducer rods are equal in length (within ±.001 in.).

An electromagnet surrounds the portion of the cryostat containing the metal specimen, which is capable of providing a variable magnetic field normal to the ultrasonic wave vector and the cylindrical axis of the metal specimen. If a magnetic field greater than the critical field of the superconductor is switched on, the metal returns to the normal state and the attenuation increases abruptly. Figure I-2b shows the same pulse-echo pattern as that shown in Fig. I-2a, except that the magnetic field $H \gg H_c(T)$ has been turned on. This change in the ultrasonic attenuation is due entirely to the effect of the conduction electrons in the metal.

The relative attenuation was measured as a function of applied magnetic field for both metals, and is plotted in Fig. I-3. Observe that it is a continuous function, a result that is implicit in the aluminum data of David, Van der Laan, and Poulis.[5] Data on the total change in attenuation between the normal and the superconducting states should probably be taken from the first pulse only. The reason for this is that the acoustic energy that arrives at the receiving transducer at the time indicated by the second pulse, since it is a superposition of two acoustic pulses that may well have travelled over slightly different paths, has an unknown phase factor determined by the details of the paths.

A specimen of mercury, approximately 0.25-in. thick, was prepared in order to obtain reasonably reliable velocity data. The velocity was measured at room temperature at 165 mc and found to be 1.47 ± .03 km/sec, which is in agreement with the measurements of Ringo, Fitzgerald, and Hurdle.[6] A specimen of this length, however, is more vulnerable to thermal contraction effects. Thus far, it has not been possible to maintain satisfactory acoustic contact between such a long specimen and the quartz transducer rods as the temperature is lowered.
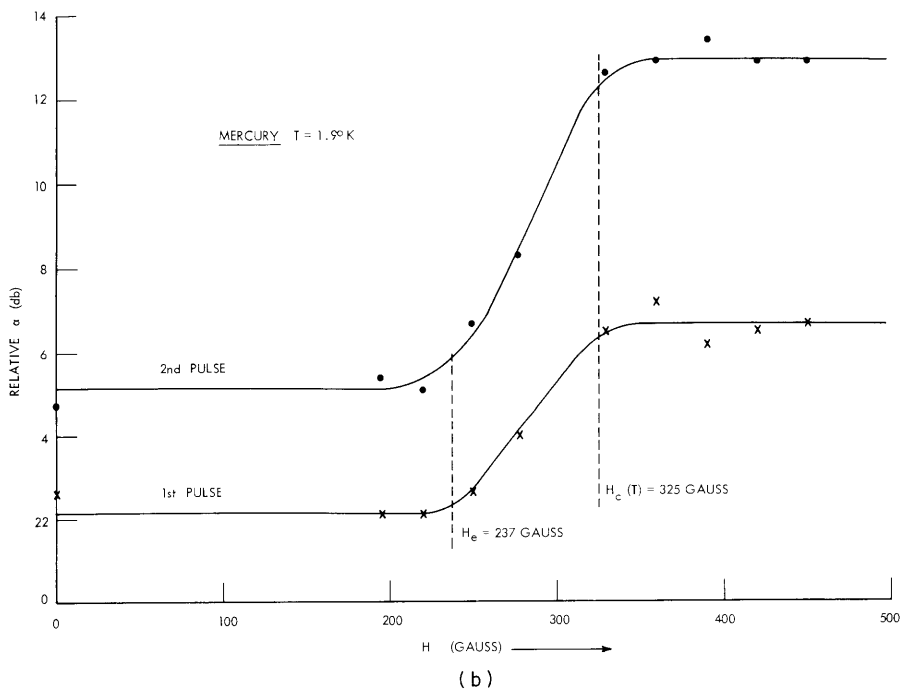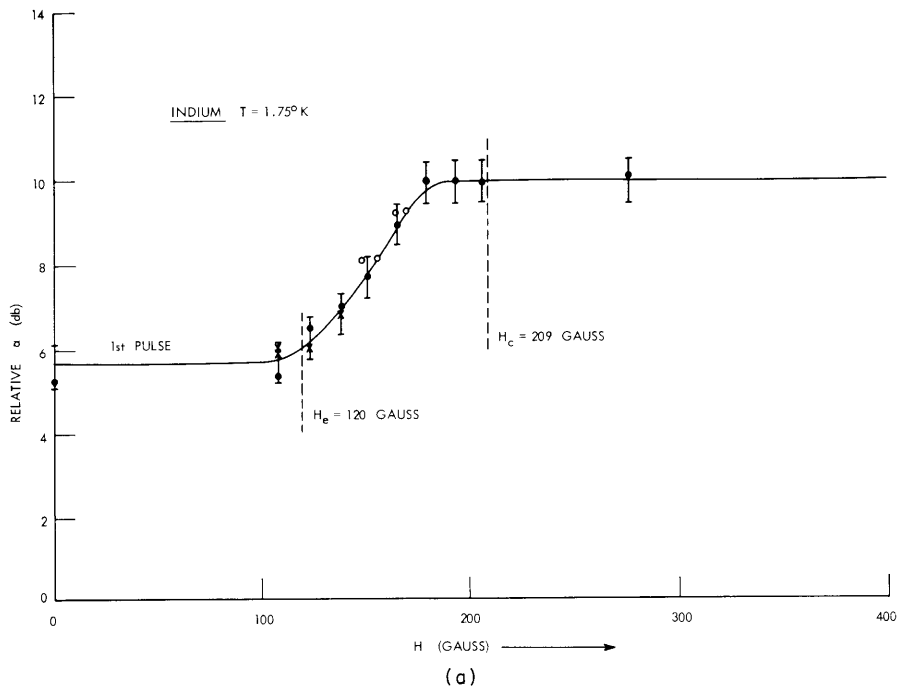
Fig. I-3. (a) Relative attenuation of a 165-mc ultrasonic pulse in
indium as a function of magnetic field.
(b) Relative attenuation of 165-mc ultrasonic pulses in
mercury as a function of magnetic field.

2. Discussion

An important mechanism for ultrasonic attenuation in metals at sufficiently low temperatures that the electronic mean-free path becomes of the same order of magnitude as the acoustic wavelength (usually below 10°K) is the electron-phonon scattering. Thus, if we have P_o acoustic power incident on a metal specimen of length L, the power P leaving the specimen will be

$$P = P_o \exp[-(\alpha_{elec}+\alpha_o)L].$$ (1)

Here, α_{elec} is that portion of the attenuation coefficient caused by scattering from the conduction electrons; α_o is that portion of the attenuation coefficient caused by scattering from thermal phonons, lattice defects, grain boundaries, and so forth. The coefficient α_o is temperature-independent in the range of temperatures which we are considering, and therefore will not be discussed further. Using the free-electron model, Pippard has calculated expressions for the electronic contribution to the attenuation coefficient for arbitrary mean-free path.[7] This approach has been well justified by the experimental results of Morse at frequencies between 9 mc and 56 mc.[8] Pippard's result for longitudinal polarization is

$$\alpha_n = \alpha' \cdot \frac{6}{\pi} \left(\frac{1}{ql}\right)\left[\frac{1}{3} \frac{(ql)^2 \tan^{-1}(ql)}{ql - \tan^{-1}(ql)} - 1\right],$$ (2)

where

$\alpha_n \equiv$ the electronic contribution to the attenuation coefficient for the metal in the normal state

$q \equiv$ the acoustic wave vector

$l \equiv$ the electronic mean-free path.

The coefficient α', which is the limiting form of α_n for long electronic mean-free path, $ql \gg 1$, is given by

$$\alpha' = \frac{\pi N m v_o \omega}{6\rho_o u_\ell^2},$$ (3)

where

$N \equiv$ the number of free electrons per unit volume

$m \equiv$ the electronic mass

$v_o \equiv$ velocity of the free electrons at the Fermi surface

$\omega \equiv$ the ultrasonic frequency

$\rho_o \equiv$ the density of the metal specimen

$u_\ell \equiv$ the longitudinal acoustic velocity in the metal specimen.

Reasonable estimates based upon the free-electron model, under the assumption of

one free electron per atom, yield the following values for a' at 165 mc:

Indium $a' = 4.2 \text{ cm}^{-1}$

Mercury $a' = 8.6 \text{ cm}^{-1}$.

The Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity has been applied to the calculation of the ultrasonic attenuation in metals in the superconducting state.[9] Their result is

$$\frac{a_s}{a_n} = 2f[\epsilon(T)],\tag{4}$$

where

$a_s \equiv$ the electronic contribution to the attenuation coefficient for the metal in the superconducting state

$f[\epsilon] \equiv$ the Fermi function of the superconducting energy gap

$\epsilon(T) \equiv$ the temperature-dependent superconducting energy gap.

Let us represent the change, in decibels, of the ultrasonic attenuation coefficient between the normal and the superconducting states at any given temperature by $D(T)$. Then

$$a_n = \frac{D(T)}{10 \log_{10}(e) \, L \tanh\left[\dfrac{\epsilon(T)}{2kT}\right]}\tag{5}$$

where

$L \equiv$ the length of the specimen in cm

$k \equiv$ Boltzmann's constant

$T \equiv$ the temperature in °K.

By using the theoretical curve for $\epsilon(T)$ given in BCS,[9] it is possible to determine a_n from our experiments on indium and mercury.

Specimen	Purity (per cent)	D(T) (db)	T (°K)	L (cm)	$(a_n)_{exp}$ (cm^{-1})
In	99.99	3.7 (±.5)	1.75	.172	5.0
Hg	99.999	5 (±1.0)	1.9	.252	4.6

These a_n, determined experimentally, are of the same order of magnitude as the limiting values a' calculated, by using Eq. 3, for the metals on the basis of the free-electron model. We conclude that for 165 mc the purities of the metals are sufficient to provide electronic mean-free paths at least of the same order of magnitude as the acoustic wavelength. That is, $ql \geqslant 1$.

As we have already pointed out, the relative attenuation, plotted as a function of magnetic field, does not yield a discontinuity at H_c, but rather begins a slow rise at H_e, well below H_c. This might be due to the presence of metallic impurities. Superconducting alloys are sometimes characterized by such a slow transition as a function of magnetic field, but this generally occurs at fields greater than H_c for the corresponding pure elements. A more cogent explanation is that the metal enters the intermediate state at H_e, and then proceeds to the normal state in a continuous manner, the transition reaching completion at $H_c(T)$. This intermediate state is a property of macroscopic superconductors, and depends upon the geometry of the specimen. The same behavior has been observed in the resistivity of macroscopic superconductors as a function of magnetic field.[10] The temperature dependence of H_c is given by

$$H_c(T) = H_c(0) \left[1 - \left(\frac{T}{T_c} \right)^2 \right]. \tag{6}$$

This formula is an approximation, but it is sufficiently accurate for our use. Here, $H_c(0)$ is the critical magnetic field at absolute zero, and T_c is the critical temperature at zero magnetic field. Both are properties of the particular superconductor under investigation. We now evaluate $H_c(T)$ for each specimen using their critical constants and the temperature, which was determined from the vapor pressure of the liquid helium.

Specimen	$H_c(0)$ (gauss)	T_c (°K)	T (°K)	$H_c(T)$ (gauss)	H_e (gauss)	n_{exp}
In	283	3.41	1.75	209	120	.43
Hg	411	4.15	1.9	325	237	.27

H_e was determined from the graphs of the experimental data (Fig. I-3).

The macroscopic theory of superconductivity predicts that the metal will enter the intermediate state at a field H_e that is such that

$$\frac{H_e}{H_c} = 1 - n. \tag{7}$$

This formula holds for macroscopic bodies whose dimensions are large compared with a characteristic length $\Delta \approx 10^{-5}$ cm. The product $4\pi n$ is the demagnetizing coefficient of the specimen which takes into account the effect of its magnetization on the external magnetic field while in the superconducting state. Obviously, n depends upon the shape of the specimen. We list a few theoretical n values for simple cases:

	$\underline{n}$
Sphere	1/3
Long cylinder in transverse field	1/2
Long cylinder in parallel field	0
Large disc with its plane normal to field	1

The shapes of our specimens were not controlled accurately, but can be described approximately. The indium was formed into a disc, 0.30 cm in diameter and 0.172 cm thick. At one edge of the disc, however, there was a sprue almost as large as the disc. The applied magnetic field was oriented parallel to the plane of the disc. The mercury was contained in a cylinder, 0.635 cm in diameter and 0.635 cm long. The quartz rods penetrated 0.192 cm into the specimen at each end. The applied magnetic field was perpendicular to the axis of the cylinder. The experimental values for n show, indeed, that the shape of the specimen is important in determining the onset of the intermediate state.

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<div align="right">J. M. Andrews, Jr.</div>

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