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Ultrasonic studies of condensed matter in the 1950s and beyond*

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1. Introduction

Unlike the long history, spanning millenia, and the wide range of uses of sound propagation in the audible domain of frequencies, studies of "ultrasonic" (especially in the megahertz region and beyond) elastic waves in condensed matter are a twentieth century phenomenon; they have gained particular prominence in the last five decades. The reason for this is, of course, the lack of means in earlier times to excite, in a controlled fashion, mechanical vibrations in the high-frequency region. The advent of devices suitable for this purpose, such as, for example, piezoelectric transducers, made these studies possible.

This brief account of the use of ultrasonic waves in studies of condensed matter concentrates largely on activities at Brown University in the 1950s and somewhat beyond, but research work in other institutions is also recorded. The choice of Brown University and of the period are, of course, arbitrary, yet it is hoped that a fair perspective of the history and growth in the field will emerge from the examples discussed. Needless to say, the coverage is incomplete, even with regard to the work done at Brown University; readers interested in more details, or a broader view of the field, are encouraged to consult the vast literature on ultrasonics, only a very small sample of which, further limited to the period covered by this account, is given in the references.

The research activities at Brown University based on ultrasonics evolved from an earlier tradition in acoustics, that originated in the first half of the century under the guidance of Bruce Lindsay. Three lines of work followed, starting in the late 1940s; these are summarized below.

In 1948, the late Rohn Truell organized the Metals Research Laboratory. Its original purpose was to develop and use ultrasonic methods in the study of solids in general, and metals in particular. Some of its early successes included the refinement of pulse-echo techniques and the development of instrumentation for use in ultrasonic investigations of solids and liquids, extensive studies of crystal defect behavior, the development of the Granato–Lücke theory of dislocation damping and calculations by Truell and coworkers on the elastic scattering of ultrasonic waves by defects in solids.

Among the early faculty members whose membership in this laboratory was relatively brief, but who left a very strong legacy, was Kurt Lücke. In 1959, Charles Elbaum joined the laboratory and these activities were gradually extended to higher and higher frequency ultrasonic waves and to their interactions with other excitations in solids, including thermal phonons, electrons, etc. As a natural evolution of the above, the research eventually encompassed thermal and electrical transport studies. Alongside these developments, the emphasis extended from the study of metals to that of semiconductors and dielectrics. In subsequent years, liquid helium and solid helium assumed an increasingly prominent role in the laboratory's activities. In the same period, Akira Hikata was very actively involved in and contributed extensively to many aspects of the research discussed.

Throughout, a very prominent role was played by Bruce B. Chick and, in the early years, by George Anderson, who were responsible for the development and construction of very sophisticated electronic instrumentation that constituted a cornerstone of the laboratory's experimental research. This instrumentation was periodically refined and updated, thus remaining in the forefront of the field.

In addition to those mentioned above, many other people were, of course, involved in these activities and contributed enormously to the overall research effort. These included numerous graduate students, post-doctoral associates and visitors from various institutions in the USA and other countries. While it would be difficult to present a complete list, we must mention at least Andrew Granato, Norman Einspruch and Roland Dobbs, and ask forgiveness of all others whose work is acknowledged collectively as a group.

Another related activity at Brown University, started in the late 1940s, was that of Robert T. Beyer and coworkers. This group was involved primarily in studies

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of liquids using megahertz frequency ultrasonics, and made numerous contributions to the field, until Beyer's retirement in the early 1980s.

In the mid-1950s, Robert W. Morse initiated an extensive program of study at Brown University in the area of magnetoacoustic properties of metals. From this program emerged numerous determinations of the Fermi surface of various metals, as well as the first experimental verification of the ultrasonic attenuation dependence on temperature in the superconducting state, predicted by the theory of Bardeen, Cooper and Schrieffer (BCS).

In the following, a few highlights among the activities mentioned above will be discussed. Given the nature and purpose of this summary, only a descriptive outline is offered, with no attempt at quantitative rigor or comprehensive coverage. More detailed treatments of these topics may be found in ref. 1.

2. Attenuation and velocity determinations

Measurements of attenuation and velocity changes of ultrasonic waves in the megahertz frequency domain are usually carried out using two approaches: the pulse-echo method and the continuous wave (CW) method. The former is more common, because it is applicable to a much wider range of experimental circumstances. While the latter approach provides higher resolution, it is very limited in the permissible amplitude, because of total energy dissipation and heating of the sample. For this reason, it is practically never used at low temperatures (liquid helium), and even at higher temperatures it is not suitable for studying amplitude-dependent (non-linear) phenomena. Nonetheless, the CW method played an important role in some applications, such as nuclear acoustic resonance, in which its high resolution was essential to the success of the studies.

In the pulse-echo method, a short (relative to the transit time of the wave pulse through the sample) duration pulse is introduced into a solid or liquid, propagating in a direction normal to two flat and parallel confining surfaces. The decreasing amplitude of the resulting successive reflections of the pulse from the two confining planes provides a measure of the attenuation of the waves in terms of either the distance or time of travel. A succession of pulses, separated in time by a period longer than the time required by the preceding pulse to decay to undetectable amplitude, is generated by an appropriate transducer (for example, a piezoelectric device) coupled mechanically to the sample under study. The transducer is excited by an electrical signal from a pulsed transmitter whose carrier frequency corresponds to that of the fundamental frequency of the transducer or one of its odd harmonics. A single transducer is often used as both the source of the input pulse and the receiver of the successive echoes that result from the single pulse. A second transducer on the opposite confining face of the sample is sometimes used as a separate receiver, depending on the circumstances and objectives of the experiment [2, 3]. The same type of arrangement is generally used to measure the velocity of the waves propagating in the medium under study, by determining the transit time of the pulse traveling between the confining planes of the sample, whose separation is known. For both attenuation and especially velocity determinations, various refinements and signal processing methods are used to enhance the resolution and precision of the measurements. It should also be noted that, in many circumstances, the quantities of primary interest are the changes in attenuation and velocity as a function of another parameter, such as, for example, temperature, magnetic field, mechanical stress, etc. [4–7]. Furthermore, absolute values of attenuation and velocity are more difficult to obtain with an accuracy comparable with that for the changes. This is due largely to the energy losses from the ultrasonic wave not related to the properties of the medium under study. Such losses come, for example, from various geometrical features (such as beam divergence) and from the conversion of some of the mechanical energy back into electrical energy in the transducer as part of the signal detection process [8, 9]. Finally, the importance of concurrent measurements of attenuation and velocity changes (which are proportional to the imaginary and real parts of the complex propagation constant respectively) is emphasized. Indeed, such concurrent measurements provide much more complete information on the physical properties studied than either one separately. (This is so, notwithstanding the fact that, in principle, either one can be obtained from the other via a Kramers–Kroning transformation, provided that they are known over a wide frequency range.)

3. Scattering

Elastic scattering of stress waves in solids, at any frequency, is brought about by differences in elastic properties from point to point. However, the strength of scattering depends on the relation between the wavelength λ and the size of the scatterer a. This dependence is usually expressed in terms of the product ka where $k = 2\pi/\lambda$ is the wavenumber, as will be further mentioned below.

The attenuation of plane waves due to elastic scattering is usually expressed in terms of the scattering cross-section γ, defined as the ratio of the total energy scattered per unit time to the energy per unit area per unit time in the incident wave front normal to the direction of propagation. If the scattering cross-section for a single scatterer is known, and if the individual scatterers are identical and may be regarded as independent of each other, then the amplitude attenuation (α) can be expressed through the loss in intensity dI at a point x

$$dI = -n_0 \gamma I dx = -2\alpha I dx \quad (1)$$

and

$$\alpha = \tfrac{1}{2} n_0 \gamma \quad (2)$$

where n_0 is the density of scatterers. When the scatterers are not independent, multiple scattering occurs and obtaining α is much more complex, even with regard to the statement of the problem.

In the context of the *ka* product, megahertz and higher frequency wave scattering become particularly important for small scatterers. Indeed, two different scattering regimes occur for $ka \ll 1$ and for $ka \gg 1$. In the Rayleigh limit, $ka \ll 1$, *i.e.* for small scatterers, the scattering cross-section, and thus the attenuation of the incident wave, is proportional to ν^4, where ν is the frequency [10, 11].

4. Dislocations

Attenuation of stress waves and wave velocity changes in solids due to dislocations were first proposed by Read [12]. The wide variety of forms in which dislocation damping occurs was subsequently rationalized in terms of the vibrating string model. This model provides a very elegant means of interpreting the vast majority of experimental observations, especially when augmented by some discretizing features. The basic model evolved from early calculations of Mott [13] and of Koehler [14], with subsequent contributions by Friedel [15], Weertman [16] and other. Finally, in 1956 (at Brown University), Granato and Lücke (G–L) connected the various elements previously discussed by others into a coherent, qualitative theory of dislocation motion, based on the vibrating string model [17]. This theory included both amplitude-independent and amplitude-dependent losses, cast in a format amenable to direct comparisons with experimental results. Various elaborations and refinements of this theory have subsequently appeared from Granato, Lücke and coworkers, as well as many other researchers. Concurrently with these theoretical contributions, numerous experimental studies were conducted and discussed in terms of the G–L theory [18, 19].

The essential features of the G–L theory are obtained by starting with the equation of motion of a damped, vibrating string pinned at both ends. Clever solutions are then obtained for the attenuation (or damping) and velocity change (or elastic modulus defect) in familiar forms of the responses of a damped harmonic oscillator. These solutions are expressed in terms of the dislocation loop lengths (or their distribution), the damping parameter, the dislocation density and characteristics of the solid. While not all these parameters are directly measurable, various ratios and combinations are accessible to experiment and others can be independently determined through the application of external fields, particularly static or dynamic bias stresses [20–27].

The G–L theory has been remarkably successful in accounting for a very wide range of dislocation-related phenomena. It has withstood the test of time essentially in its original form, with only a few refinements and elaborations, mostly in the area of thermal effects, which are not addressed explicitly in the initial treatment.

One of the elaborations involved the introduction of a discretized form of the dislocation displacements consisting of kink motion, rather than the continuum-like "stretching" of an elastic string [26]. This approach applies to dislocation motion at low temperatures, and has been used to account for experimental observations of ultrasonic attenuation and velocity changes in that regime.

5. Harmonic generation due to dislocations

Anharmonicity of a medium may be viewed as a departure from linear dependence between applied stress and resulting strain, *i.e.* a departure from Hooke's law. In all condensed matter, this non-linearity originates from the anharmonic terms in the interatomic potential. In addition, in solids containing dislocations capable of glide displacements in response to applied stresses, non-linear stress–strain behavior can also occur due to these displacements.

In the preceding section, the work described assumed that the stress–strain relation is linear, except in the amplitude-dependent regime involving dislocation unpinning from weak pinning points. However, even in the absence of unpinning the stress–strain or force–displacement relation is inherently non-linear. This feature is quantitatively negligible at low amplitudes, but becomes significant as the amplitude increases. One important consequence of this behavior is that a sinusoidal driving force associated with a propagating ultrasonic wave at a frequency ω will produce displacements at ω, as well as its higher harmonics. Depending on the shape of the potential in which the dislocation vibrates, even or odd harmonics (or both) will be generated.

Detailed studies of harmonic generation as a function of various parameters were carried out in the megahertz region [28–32]. These studies contributed both to the general scope of the non-linear response of systems containing dislocations, as well as to clarifying aspects of the intrinsic properties of dislocations. It may be worth noting that, although the harmonic generation processes just described are a general phenomenon applicable at all frequencies, their experimental observations are, in practice, restricted to a narrow range of frequencies in the low megahertz region. This is because the amplitude of the harmonics relative to that of the fundamental wave increases with increasing frequency of the fundamental, but so also does the attenuation. This may be illustrated by the following expressions for the amplitude (A_2) of the second harmonic and the amplitude (A_3) of the third harmonic at a distance x from the source (at $x=0$), where a fundamental wave of amplitude A_{10} and frequency ω is introduced

$$A_2 = \frac{\omega^2}{k} P A_{10}{}^2 \frac{\exp(-2\alpha_1 x) - \exp(-\alpha_2 x)}{\alpha_2 - \alpha_1} \tag{3}$$

$$A_3 = \frac{\omega^2}{k} Q A_{10}{}^3 \frac{\exp(-3\alpha_1 x) - \exp(-\alpha_3 x)}{\alpha_3 - 3\alpha_1} \tag{4}$$

Here P and Q are combinations of material properties and factors pertaining to the characteristics of the dislocation network and α_1, α_2 and α_3 are the attenuation coefficients of the fundamental wave of frequency ω, the second harmonic (2ω) and the third harmonic (3ω) respectively. In view of the fact that, over a wide range of conditions, α_1, α_2 and α_3 are proportional to the square of ω, 2ω and 3ω respectively, it is readily seen from the above equations that, as x increases from $x=0$, A_2 and A_3 first increase, go through a maximum and then decay rapidly due to the exponential factors. Thus there exists a small frequency interval over which harmonics are fairly readily accessible to observation for sample lengths of common laboratory practice.

6. Interactions with conduction electrons in metals

Attenuation of ultrasonic waves through interactions with conduction electrons becomes appreciable at temperatures below about 10 K. At higher temperatures, other mechanisms are usually dominant.

Interactions between electrons and thermal phonons are, of course, an essential feature of electron transport. The differences between the two cases arise from the different frequency ranges involved, and the basically "monochromatic" character of ultrasonic waves, in contrast with the wide (Planck) distribution of thermal phonons.

After the initial experimental observations of these interactions by Bömmel [33] and MacKinnon [34], several workers calculated the resulting attenuation using semi-classical arguments and the free-electron approximation [25–37].

The results obtained by these workers were essentially the same. These results are illustrated in the following expression for the attenuation (α) of longitudinal waves, as derived by Morse [35] for the condition $ql_e < 1$, where q is the wavenumber and l_e is the electron mean free path.

$$\alpha = \frac{2}{15} \frac{N m v_F}{\rho_0 v_l} q^2 l_e \tag{5}$$

Here N is the electron number density, m is the electron mass, v_F is the Fermi velocity, ρ_0 is the mass density of the material and v_l is the longitudinal wave velocity. As can be seen, in this regime, α varies linearly with l_e and thus has the same temperature dependence as l_e, the other parameters being essentially temperature independent in the region $T < 10$ K.

Pippard [38], again using the free-electron model and semi-classical arguments, extended this approach to arbitrary values of ql_e. He further clarified the problem by pointing out that the displacements of the lattice ions by a stress wave induce an internal electrical field. This field is electrostatic for longitudinal waves and electromagnetic for transverse waves.

Pippard's argument for calculating the ultrasonic attenuation may be summarized as follows. A lattice wave traveling through a metal gives rise to variations of electric forces on the electrons. The positively charged lattice ions will undergo periodic displacements with velocity v, but the electron density may not remain constant. For example, in the compressed region of a longitudinal wave, the increased positive charge density will attract electrons, the opposite happening for the expanded region. If the electron and ion densities do not follow each other exactly, space charges develop, resulting in a periodic electric field in the direction of wave propagation. In the case of the transverse waves there are no density changes, and hence no electric fields resulting from space charges. If, however, the lattice and electronic currents do not cancel each other, periodic magnetic fields are generated; these give rise, in turn, to electric fields by induction. There will also be relaxation effects due to collisions of electrons with thermal phonons, defects, etc., which will tend to restore equilibrium with the surroundings. Pippard, in effect, calculated the net distortion of the Fermi surface due to the combined influence of the internal electric fields and collisions.

In the case $ql_e > 1$, Pippard's calculations give an attenuation coefficient (α) that varies linearly with frequency and is independent of ql_e

$$\alpha(ql_e>1)=\frac{\pi N m v_F}{12\rho_0 v_e^2}\,\omega \tag{6}$$

Pippard's treatment yields for the overall dependence of the attenuation of longitudinal waves on ql_e

$$\frac{\alpha}{\alpha(ql_e>1)}=\frac{6}{\pi}\left[\frac{ql_e A}{3(1-A)}-\frac{1}{ql_e}\right] \tag{7}$$

where $A=(\arctan ql_e)/(ql_e)$.

A more elaborate and more general treatment of the attenuation and dispersion of ultrasonic waves was given by Steinberg [39], using Boltzmann's transport equation and the free-electron model of a metal. He also assumed the knowledge of an effective relaxation time for restoring the thermal equilibrium distribution of electrons.

The expressions for attenuation obtained by Steinberg [39] are similar in form to those discussed above and have the same dependence on frequency and on the electron mean free path. However, there are differences in the numerical coefficients. The experimental results generally agree well with the theoretical predictions in terms of their temperature dependence (through the electron mean free path) and frequency dependence (for the different regimes, $ql_e<1$ and $ql_e>1$). Numerical values of the electronic contribution to the attenuation are, however, difficult to obtain with high accuracy. Two difficulties contribute to this uncertainty. The first is that the theoretical treatments discussed are based on the free-electron approximation, while most experiments are carried out on metals that depart in varying degrees from this approximation. The second is due to the attenuation arising from causes other than interactions with conduction electrons. At very low temperatures, this attenuation is usually temperature independent, but not independent of frequency; it may constitute a significant fraction of the total attenuation measured, particularly at high frequencies. It may be difficult, therefore, to separate the various effects and to obtain accurate values of the numerical coefficients for electron attenuation.

Finally, it should be pointed out that many quantum mechanical calculations of the interaction between ultrasonic waves and conduction electrons in metals have been carried out. A simple approximate treatment of the subject by Morse [40] provides an overview of the physical issues and of the results obtained. It is noteworthy that when applied to free electrons, these results are identical with those derived by semi-classical methods (eqn. (6)).

7. Influence of a magnetic field; the magnetoacoustic effect

The dependence of ultrasonic attenuation due to conduction electrons on a magnetic field can be described in terms of the change in the electron mean free path. Qualitatively, it may be assumed that a magnetic field, by causing the electrons to follow curved paths, reduces the distance between collisions in the direction of the ultrasonic wave propagation. This results in a reduction in attenuation, which depends on the distance in the direction of propagation traveled by an electron between collisions (relative to the wavelength). The following experimental observations support this viewpoint. When $ql_e>1$, the attenuation (α) varies linearly with frequency (ω) for zero magnetic field. The application of an increasing magnetic field gradually increases the frequency dependence of α, and eventually α becomes proportional to ω^2 for sufficiently high magnetic fields. The latter occurs when the field is high enough for the electron orbit diameter to become smaller than the ultrasonic wavelength. Since in the absence of a magnetic field the attenuation varies as ω^2 for $ql_e<1$, the effect of applying a strong magnetic field is interpreted in terms of a shortening of the mean free path of the electrons.

A more quantitative treatment of this effect, based on the free-electron approximation, was given by Steinberg [41]. He showed that, for transverse waves, with the magnetic field perpendicular to both the polarization and propagation directions, the field dependence of the attenuation for $ql_e<1$ should be of the form

$$\frac{\alpha(H)}{\alpha(0)}=\frac{1}{(1+2\omega_c\tau)^2} \tag{8}$$

where $\omega_c=(eH)/(mc)$ is the cyclotron frequency. For a magnetic field parallel to the direction of polarization and perpendicular to the propagation direction, this relation becomes

$$\frac{\alpha(H)}{\alpha(0)}=\frac{1}{1+(\omega_c\tau)^2} \tag{9}$$

These equations predict that, for large fields, the attenuation should vary as H^{-2}, which agrees quite well with experimental results [40] when $ql_e<1$. Moreover, although eqn. (8) was derived for transverse waves, it was found experimentally to apply also for longitudinal waves when the field is perpendicular to the wave propagation direction.

8. Applications to Fermi surface studies

Far more interesting than the case of $ql_e<1$, from the point of view of the electron theory of metals, is the ultrasonic attenuation in the range $ql_e > 1$, where an oscillatory variation of α with field strength is observed. These oscillations were interpreted as a resonance-like effect arising when the electron mean free path is sufficiently long so that the electron orbit size matches the ultrasonic wavelength. Pippard [42] then proposed that this phenomenon could be used to investigate the shape of the Fermi surface. When the electron mean free path is greater than the ultrasonic wavelength, the electrons pass through many phases of the wave without being scattered. In view of the large difference between the Fermi velocity and the ultrasonic velocity, the ultrasonic waves appear stationary to the electrons. A magnetic field value can thus be found such that the orbit diameter of the electron matches with a multiple of the ultrasonic wavelength. As the magnetic field changes, the orbit diameter will change until it again matches a multiple of wavelength, and periodic oscillations as a function of field strength H, with a period inversely proportional to H, will result. Furthermore, the orbit diameter is proportional to the electron momentum in a direction perpendicular both to the magnetic field and to the ultrasonic wave. Thus the periodicity of the attenuation for various field directions with respect to the axes of a single crystal can be used to determine the shape of the Fermi surface. The earliest experiments using these ideas were carried out on metal single crystals by Morse *et al.* [43–45] and by Reneker [46].

Shortly after Pippard's original proposal, three fairly distinct types of oscillations were recognized. These are geometric resonances, quantum oscillations (also known as ultrasonic de Haas–van Alphen oscillations) and acoustic cyclotron resonances. All three are periodic in the reciprocal of the applied magnetic field, with periods that are related to the extremal linear dimensions, extremal cross-sectional areas and curvatures of the Fermi surface respectively.

9. Applications to superconductivity

Experiments by Bömmel [33] on lead and by MacKinnon [34] on tin first established that the ultrasonic attenuation decreases rapidly, as a function of decreasing temperature, below the superconducting transition of a metal. Unambiguous proof that conduction electrons in the normal state cause attenuation was provided in this connection; when, below the transition temperature, a magnetic field of sufficient strength to destroy superconductivity is applied to the metal, the attenuation returns to the higher value observed above the transition temperature. As a follow-up to these observations, shortly after the advent of the BCS theory of superconductivity [47] came one of the earliest experimental tests of one of its predictions, namely the temperature dependence of the ultrasonic attenuation below the transition temperature. This prediction

$$\frac{\alpha_s}{\alpha_n} = \frac{2}{\exp(E_g(T)/k_B T) + 1} \tag{10}$$

where α_s and α_n are the ultrasonic attenuations in the superconducting and normal states respectively (at the same temperature), E_g is the temperature-dependent superconducting energy gap and k_B is Boltzmann's constant, was well verified experimentally [40].

In the limit of very low temperatures (*i.e.* where E_g becomes essentially constant and $E_g \gg k_B T$), eqn. (10) reduces to an exponential dependence of α_s/α_n on $(-1/T)$. Measurements of α_s/α_n thus provide a means to determine E_g, and when carried out on single crystals as a function of orientation, the anisotropy of E_g can also be found. Such studies were carried out by Morse *et al.* [40, 48, 49], providing some of the earliest values of E_g and especially of its anisotropy.

10. Concluding remarks

This brief and, admittedly, very incomplete account of high-frequency ultrasonic studies in the 1950s provides, it is hoped, a glimpse of some of the important and exciting activities of that period. It is interesting to note that many of the approaches, both experimental and theoretical, developed then continue to be widely used in basic research as well as in numerous applications that have evolved in the intervening years. The significance and vitality of high-frequency ultrasonics are well illustrated by the large number of scientific and technical publications that continue to appear on this subject.

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