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Scanning Acoustic Microscopy. Physical Principles and Methods. Current Development

1.1

Basics of Acoustic Wave Propagation in Condensed Media

An acoustic wave is a physical phenomenon responsible for the transfer of dilatational and shear strains [11–16]. Generally, acoustic waves of three sorts can propagate along a certain direction, each of them spreading at its own velocity and imparting oscillatory motion in particles of the medium in its own direction – wave polarization. Only two sorts of waves propagate in an isotropic solid; namely, longitudinal and transverse ones. In a longitudinal wave, particles are displaced in the direction of wave motion while the overall strain transferred is a combination of the dilatational and shear strains. Transverse waves transfer shear strain alone, particles of the medium oscillate in an arbitrary direction in a plane normal to the propagation direction.

Distinguishing between longitudinal and transverse waves in a crystalline solid makes no sense because all the three acoustic waves spreading in a given direction transfer both the dilatational and shear strains [12, 16–21]. A wave in which longitudinal particle displacements dominate is called quasi-longitudinal; normally, it propagates at the highest velocity. The other two waves are called fast and slow transverse waves, in accordance with the value of their propagation velocity. A crystalline medium, even of high symmetry, exhibits a significant anisotropy of its acoustic properties, that is, its velocity heavily depends on the propagation direction with respect to the crystallographic axes. The phase and group acoustic wave velocities can appreciably differ both in their values and directions [12, 18, 21].

Longitudinal waves alone can propagate in liquids where no shear stresses exist. However, many liquids exhibit significant shear stresses at hypersonic frequencies, thereby approaching solids in their mechanical properties. Thus, high-frequency shear waves can propagate in them. This phenomenon was discovered in studying the Mandelstam–Brillouin scattering on the so-called anisotropy fluctuations in liquids which are, in essence, shear waves [16, 22, 23].

The propagation velocity and absorption coefficient of acoustic waves are the basic characteristics of their acoustic properties. According to the customary notions of fluid mechanics, the speed of sound is a parameter of the medium which is

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independent of frequency [13–16, 20]. Absorption of acoustic waves in a medium is governed by dissipative processes, namely, by viscosity and heat conduction, coefficients of which are assumed to be constants of the material. The absorption coefficient is proportional to viscosity and increases proportionally to the sound frequency ν: $\alpha \propto \nu^2$ [13–16, 20]. Within this assumption, dispersion of the acoustic velocity shows up at very high frequencies at which the notion of continuity of the material becomes unjustified and the discrete nature of its structure manifests itself [24].

However, many materials exhibit frequency dispersion of their speed of sound and deviation from the quadratic frequency dependence of their acoustic absorption coefficient caused by the frequency dependence of the effective material viscosity [12–17, 20, 25–30]. The frequency dependence of the speed of sound and viscosity stems from the occurrence of two sorts of processes in the material induced by the acoustic wave; namely, resonant excitation of the internal degrees of freedom in the material and acoustic relaxation [13–16, 25, 27, 28].

Resonant excitation processes are inherent in heterogeneous media (oscillations of individual crystallites in polycrystals [31] or of air microbubbles in liquids), although they can be observed in homogeneous materials as well, e.g., when displacements are induced in crystals [16, 32].

Acoustic relaxation is a process of local equilibration of the material perturbed by an acoustic wave [13–16, 25, 27, 28]. If the relaxation time of a certain parameter τ is much greater than the wave period: $\nu\tau \gg 1$, its variation in an acoustic wave can be neglected, that is, the appropriate degrees of freedom are "frozen" in the wave.

If, however, the period of acoustic oscillations is commensurate with the relaxation time or is greater than τ, that is, $\nu\tau \le 1$, the parameter at issue varies at an acoustic frequency due to instantaneous attainment of its equilibrium value. Inasmuch as relaxation processes are irreversible, some portion of the acoustic energy goes to heat. Hence, an additional dissipation mechanism arises in the material when the sound frequency decreases and the fluid viscosity increases. Concurrently, the elasticity modulus reduces because the relaxation processes proceed in such a way that the elastic stresses induced by material deformation diminish. As a result, dispersion of the acoustic velocity arises near frequency $\nu \sim \nu_\mathrm{r} = 1/\tau_\mathrm{r}$ called the relaxation frequency: as the frequency increases so does the speed of sound from its low-frequency value c_0 at $\nu\tau \ll 1$ to the high-frequency value c_∞ at $\nu\tau \gg 1$.

The frequency dependence of acoustic absorption in the relaxation domain deviates from the quadratic law because the material viscosity reduces from its low-frequency value η_0 to its high-frequency one η_∞ as the frequency increases. The above-mentioned shear stress arising in a liquid at high frequencies [25, 27] is an example of acoustic relaxation. Displacement of one fluid layer with respect to another brings about elastic shear stresses that lead within time $\Delta t \ge \tau_\mathrm{r}$ to irreversible motion of the layers, that is, to their flow. Naturally, at frequencies $\nu\tau_\mathrm{r} \gg 1$, the time allotted is too short for the displacement to occur and the liquid behaves as an elastic solid.

Apart from the shear relaxation, there are a number of other mechanisms of acoustic relaxation in liquids, e.g., those associated with liquid restructuring, energy redistribution among the internal degrees of freedom, and some other processes. The relaxation frequencies lie in the mega- and gigahertz ranges; the dispersion of speed of sound in these ranges can attain a few tens of percent. It is worth noting that water, which is the liquid most frequently used in acoustic microscopy, exhibits no dispersion of its speed of sound.

The basic process responsible for dissipation in solids is relaxation in the system of thermal phonons, the equilibrium state of which is perturbed by an acoustic wave (Akhiezer's mechanism) [26, 30]. Inasmuch as the relaxation time of a phonon system is short: $\tau_r \approx 10^{-11}$ s, the aforesaid mechanism fails to give rise to a frequency dependence of the speed of sound and viscosity up to a few tens of a gigahertz. Dispersion in this frequency range is governed by the fluid discreteness and absorption of sound is controlled by the direct three-wave interaction of the acoustic wave with thermal phonons (Landau–Rumer mechanism) [26, 30].

Apart from the phonon viscosity, some other mechanisms responsible for the absorption and dispersion of the acoustic velocity exist in crystals. In many crystals, interaction of an acoustic wave with crystalline structure defects (with dislocations in the first place) becomes significant at frequencies in the 10^6–10^{11} range [30, 32, 33].

Interaction with dislocations can be of a resonant nature, that is, it is most efficient when the frequency of the acoustic wave is close to the fundamental frequency of dislocation loops [32]. Interaction of oscillating dislocations with thermal phonons brings about dislocation absorption of acoustic waves. Naturally, the absorption and dispersion associated with dislocations depend on the degree of crystal perfection.

Semiconductors have a peculiar mechanism of absorption and dispersion of their speed of sound associated with the interaction of conduction electrons with an acoustic wave [16, 34–37]. This interaction is inherent in all sorts of semiconductor crystals, but is most prominent in piezosemiconductors in which the acoustic wave induces a variable electric field redistributing the free charge carriers in the material bulk due to the piezoelectric effect. As a result, there arises an induced wave of the electron density that spreads together with the acoustic wave.

Dissipation of the energy of directed electron motion results in effective ultrasound absorption. Interaction of an acoustic wave with free charge carriers is of a relaxation nature, that is, inhomogeneous electron distribution forms within some representative relaxation time period τ_e. At high frequencies, when $\nu\tau_e \gg 1$, the electron wave has no time to arise so that the presence of electrons virtually does not affect the behavior of the propagating acoustic wave. In many semiconductors (CdS, CdSe, ZnO, Zn, Te, and others) electron absorption dominates within a wide frequency range up to few gigahertz.

Dispersion of the speed of sound and absorption are also caused by interaction of ultrasound with other crystal subsystems, namely, spin waves, domain walls in ferromagnetics and ferroelectrics [15, 16, 19, 21, 24, 30, 38–40].

Apart from bulk acoustic waves, surface acoustic waves can also propagate in bounded solids; the amplitude of these latter waves diminishes exponentially as they move away from the boundary [12, 16, 20, 41–48]. The energy transferred by surface waves is concentrated in a surface layer whose thickness is of the order of the wavelength. The velocity of a surface wave must be less than that of bulk waves in bounded materials; otherwise, the surface wave would be re-emitted inwards into the solid in the form of bulk waves.

Depending on the nature of contacting materials, various types of surface wave can propagate along the interface between them [12, 41, 47, 48]. Rayleigh waves spreading along a solid–vacuum or solid–rarified medium interface [12, 41, 42, 45–48] are particularly important for reflection acoustic microscopy. The motion of the material particles in a Rayleigh wave is a superposition of two oscillations at the surface aligned with the wave propagation direction and perpendicular to it. The propagation velocity of Rayleigh waves is close to the speed of transverse acoustic waves, but is always somewhat lower than this.

A Rayleigh-type wave can also propagate along a solid–liquid or solid–solid interface [41, 46–49]. Propagation of Rayleigh waves along the boundary between solid half-space and a solid or liquid layer is of importance for acoustic microscopy practice.

The use of Stoneley–Scholte surface waves arising at the solid–liquid interface is a promising approach to developing a system for surface wave acoustic microscopy because absorption of these waves is sensitive to the mechanical structure of liquid or quasi-liquid (that is, a fluid with a small shear stress) fluid. In compliance with the structure of Stoneley–Scholte waves, their absorption is governed by energy dissipation in the crystal and liquid. Contribution of the internal friction effects in crystals is insignificant; therefore, the wave absorption is controlled solely by the viscoelastic properties of a liquid. A theoretical analysis demonstrates [50] that the absorption behavior depends on the ratio between the speed of sound in the liquid and the velocity of the transverse acoustic waves in the crystal.

Dissipation of the surface wave energy can be controlled both by absorption of the longitudinal component of the surface acoustic wave in liquid and by a viscous shear wave arising in the liquid near the interface.

Surface waves of another type are the Love waves that propagate along the boundary of the solid half-space on which the solid layer lies [41, 44, 47]. The material particles oscillate in the wave normal to the wave propagation direction within the layer plane; therefore, the wave should be qualified as transverse. The energy transferred by the wave is concentrated both in the layer and in the half-space zone adjacent to the interface. The velocity of Love waves depends on frequency and varies in the range between the velocities of transverse acoustic waves in the layer and half-space.

The existence of specific acoustic surface waves on some planes of piezoelectric crystals, called the Guliayev–Bluestein waves, is a distinguishing feature of these crystals. These waves are Love waves spreading over a free surface [49].

At this point, we terminate our familiarization with the properties of acoustic waves in condensed media and resort to consideration of the physical principles of acoustic microscopy.

1.2
Physical Principles of Scanning Acoustic Microscopy

The lens system (Figure 1.1) is the heart of a scanning acoustic microscope (SAM). An ultrasonic wave is generated in the acoustic lens system by a transducer mounted at one of its ends. The wave spreads through an acoustic duct possessing a large acoustic impedance and is then focused with the aid of a spherical recess (lens) at the other duct end in an immersion material (liquid as a rule) filling the space between the lenses and objects examined. The immersion liquid provides a large refractive index for the acoustic lens and good acoustic contact between the duct and object. The focused beam interacts with the object, being partially reflected and scattered by the object and partially transmitted through it. If the reflected wave is detected, the microscope operates in the reflection mode. When the transmitted acoustic flux is recorded with the aid of the second lens, we have a transmission acoustic microscope.

It should be mentioned that a great number of modifications has been suggested within the basic principle underlying SAM that extends opportunities of the method. Thus, detection of acoustic radiation scattered by the object was arranged by rotating the lens through various angles with respect to the axis of the emitting lens [51]. This operating regime is essentially similar to the dark field regime of optical microscope operation and allows the effective resolution depth to be varied. A transmission regime with a single lens was realized in which a detector based on the acoustic-electric effect quadratic in the signal amplitude is used [52].

Additional information is obtained with the use of various nonlinear regimes in which the signal from the focal zone is recorded at harmonics of the input sig-

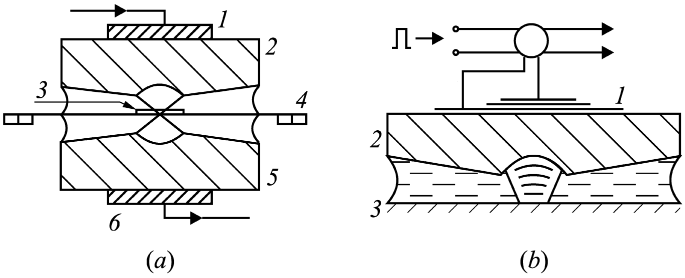

Figure 1.1 The lens system of a scanning acoustic microscope. (*a*) Transmission microscope: *1* – piezotransducer, *2* – emitting lens, *3* – object, *4* – scanner, *5* – receiving lens, and *6* – receiving transducer. (*b*) Reflection microscope: *1* – piezotransducer, *2* – lens, and *3* – object.

nal [53] or at combined frequencies [54] rather than at the frequency of the input signal itself. In this case, both linear and nonlinear object properties contribute to the measurement results. An interference technique in the transmission microscope operation mode was suggested in [55] to measure local speeds of sound. The $V(z)$-characteristics method described in detail below is at present the most advanced technique for measuring local values of the velocity of Rayleigh waves spreading over the surface of samples studied. Of certain practical interest is also the possibility of using it in SAM surface acoustic waves; nowadays, some ideas have already been realized in acoustic microscopy.

At present, some promising approaches in enhancing the contrast and improving the quality of the image and SAM resolution have been proposed. One of these approaches is the use of appropriate immersion liquids, in particular, those possessing the lowest values of the speed of sound and transmission coefficient.

A set of cryogenic acoustic microscopes employed to study material surfaces was developed in Stanford University (USA). One of the instruments using liquid helium at $T = 0.1$ K [56] and operating at 8 GHz provided a 250–300-angstrom resolution, which is so far the highest level attained.

Finally, acoustic microscopy opportunities can be significantly extended by invoking the contemporary methods for computer image processing. This would obviously improve the image quality [57], enhance resolution, and offer the possibility of processing and documenting the data, and analyzing dynamic processes, etc. [58].

Resolution of the instrument and depth of radiation penetration are the most important characteristics of the method. They depend on the ultrasound frequency, parameters of the lens system, immersion material, and properties of the object. As the resolution increases, the depth of ultrasound penetration in the object reduces. Therefore, the ultrasound frequency is to be selected after taking into account the properties of the object and the problem to be solved and reasonably compromising between the penetration depth and resolution.

It is ultrasound radiation which permits one to obtain new information about the mechanical properties of a micro-object that cannot be provided by other microscopic methods.

Currently, we can formulate the following basic points of acoustic microscopy development:

- The development of physical grounds and the search for new foundations of acoustic microscopy.
- The study of the physical basics of acoustic imaging.
- The development of automated methods for recovering and analyzing the acoustic structure of objects.
- Investigations into the principles of acoustic microtomography.
- The development of acoustic microscopy techniques as applied to contemporary scientific and technical problems.
- Engineering developments.
- The design and manufacture of industrial samples of new acoustic microscopes.

1.3
Acoustic Imaging Principles and Quantitative Methods of Acoustic Microscopy

Obviously, of the above investigation lines, the first two are the most interesting and important; of particular importance is understanding the physical principles of acoustic imaging, in the first place the problems of image contrast and artifacts. Acoustic images cannot be interpreted without clear understanding of the physical mechanisms of their formation and the nature of acoustic contrast. Knowledge of these mechanisms allows one to perform quantitative measurements and to quantitatively characterize the materials studied. Measuring the output signal of an acoustic microscope, its amplitude and phase, and comparing them with the amplitude and phase of the reference signal in liquid, one obtains information about the speed of sound, acoustic impedance, attenuation, and geometric characteristics of the sample (thickness, curvature, and slope angle of the surface).

We start with considering how the output signal of an acoustic lens forms in a general case. The signal is detected with a piezoelectric transducer whose response is linear. In order for electric signal to be generated by the transducer, the incident wave front must be parallel to its surface, that is, the acoustic radiation refracted at the lens surface must impinge on the transducer surface normally to it. In other words, all the beams must pass through the lens focus. Furthermore, they all must arrive at the transducer in the same phase. Otherwise, signals from various beams would interfere and the resulting signal would be attenuated. Consider first how the signal forms in a reflection microscope (Figure 1.2). If the object boundary is at the lens focus (Figure 1.2*a*), the output signal is controlled by the integral (over all incidence angles from $\theta = 0$ to $\theta = \theta_m$, where θ_m is half the aperture angle) refraction index. If the lens is moved away from the object (Figure 1.2*b*), the cone

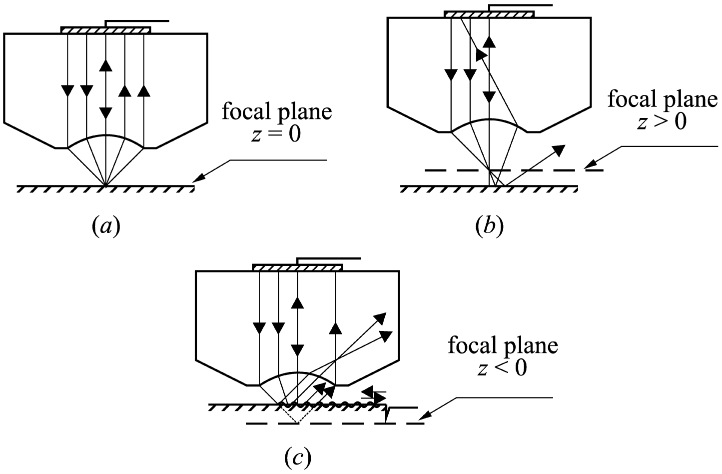

Figure 1.2 Schematic of the output signal of a reflection microscope: (*a*) $z = 0$, (*b*) $z > 0$, and (*c*) $z < 0$; z is the deviation from the focal plane of the lens.

of beams received by the transducer narrows rapidly and the level of the signal from the transducer drops drastically.

The $V(z)$ curve depicting the output signal amplitude as a function of distance from the focal plane of the lens shows a rapid decrease in the signal amplitude with small shallow oscillations as z increases in the $z > 0$ range. The oscillations are caused by a difference between the phases of differently directed beams. A dissimilar behavior of the $V(z)$ dependence can be observed at negative z values when the lens approaches the object (Figure 1.2*c*).

In solid objects, a Rayleigh wave propagates over the sample surface. If its velocity exceeds the sound speed in the immersion liquid, a surface wave would arise which is re-emitted back into the liquid. Waves of this type are called leaky Rayleigh waves. They arise at any position of the lens with respect to the object. However, they are detected solely at $z < 0$. Figure 1.2(*c*) demonstrates how it occurs. The output signal shows up as a superposition of the signal generated by the mirror-reflected paraxial beam and the signal generated by the leaky surface wave. The phase difference between these signals depends on the distance z; as a result of their interference, the $V(z)$ dependence exhibits a regular set of maximums and minimums [59, 60]. The distance between neighboring maximums and minimums is unambiguously related to the velocity of the Rayleigh wave spreading over the sample surface (Figure 1.3). The onset of a leaky Rayleigh wave is extremely important in acoustic imaging in the reflection mode. In particular, it brings about interference fringes near sharp inhomogeneities, at curved surfaces, and so on. It also gives rise to the effect of acoustic contrast inversion caused by small lens displacements. Figure 1.2(*c*) demonstrates how the interference fringes arise near a surface defect. Reflection from inhomogeneities produces not only the forward wave but the backward leaky Rayleigh wave as well, the latter wave can also be detected by the transducer. Inasmuch as its phase depends on the position of the lens axis with respect to the inhomogeneity, lens scanning produces interference fringes that follow the inhomogeneity contour.

The use of linear lenses that excite Rayleigh waves spreading in one direction turned out to be extremely informative in reflection microscopy. Rotating the lens with respect to the sample and thereby varying the propagation direction, one can measure local anisotropic properties of the material surface studied. This method was suggested and successfully developed in the works of Japanese scientists J. Kushibiki and N. Chubachi. At present, local velocities of Rayleigh surface waves are measured by this method with an accuracy of 10^{-4} [61].

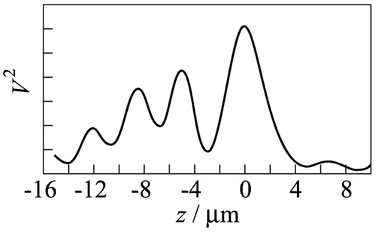

Figure 1.3 Representative pattern of the amplitude of output microscope signal V vs distance z between the object and lens focal plane.

What produces the image in an acoustic microscope operating in the transmission regime? We start with a discussion of the refraction phenomena. Let us consider transmission of a focused beam through a thin plate. The beam passing through a plate is refracted twice. As a result, its direction after it exits from the plate is parallel to that of the incident beam; however, the exit beam is shifted with respect to the incident one. The shift value depends on the incidence angle.

The rays, after passing a plate, form a diverging beam when the speed of sound in the plate exceeds the speed of sound in the immersion liquid; rays which are close to the lens axis make a paraxial focus while the rays incident at greater angles on the plate surface are focused at different points of the acoustic axis. As a result, the cone of rays received by the transducer narrows after passing the lens and the output signal diminishes. Variations of the signal due to the refraction effects depend on the ratio of the speeds of sound in the sample and liquid. Similarly, the cone of rays recorded by the receiving lens is narrowed due to phase aberrations. Phase aberrations stem from the fact that rays incident at various angles upon the object travel in it through various distances. Therefore, they have a different shape and can mutually attenuate the signals they induce in the output piezoelectric transducer. The values of phase aberrations are also controlled by the difference between the speeds of sound in the liquid and sample. Therefore, phase aberrations cause additional contrast of acoustic images.

To measure the local mechanical properties of samples quantitatively, we have developed a method of $A(z)$-characteristics [62, 63]. An $A(z)$ curve is the dependence of the output signal A of the receiving lens on the distance z between the lenses. A model $A(z)$ curve is displayed in Figure 1.4. We place an object between the lenses. As follows from the aforesaid, the focal point of the received acoustic beam is shifted due to refraction in the plate. Accordingly, the maximum of the

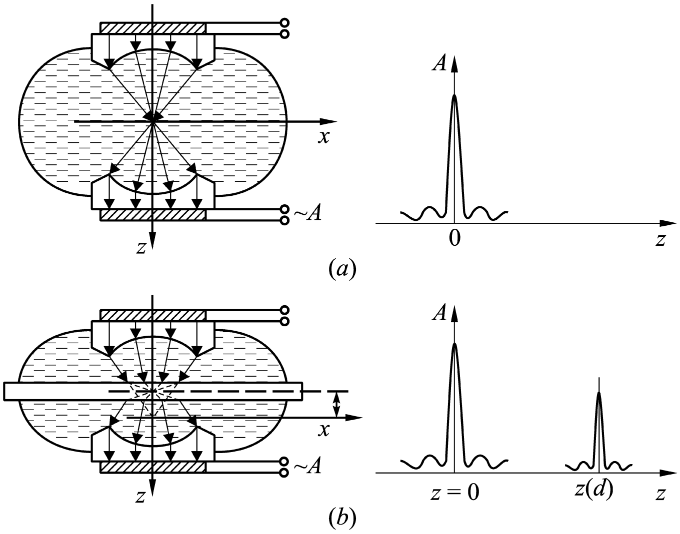

Figure 1.4 Illustration of the method of $A(z)$-curves: (*a*) without object and (*b*) with object.

$A(z)$ curve is displaced toward smaller z, if the speed of sound in the plate exceeds the speed of sound in the immersion liquid, and toward greater z, if the ratio of the acoustic velocities reverses.

The shift of the $A(z)$ maximum is proportional to the difference between the speeds of sound in the immersion liquid c_0 and in the object c, and to the sample thickness $\Delta Z = d(c/c_0 - 1)$. A local value of the speed of sound in a zone 5 or 10 µm in size can be assessed by measuring ΔZ. The ratio between the maximum amplitudes yields the coefficient of sound transmission through the plate. Knowing the object density and the value of the velocity c, the local ultrasound attenuation coefficient can be evaluated using the known impedance and transmission coefficient. The method of $A(z)$ characteristics was employed to measure the ultrasound velocity in various polymer films [63].

It should be mentioned that refraction in solids gives rise to shear waves. Being refracted at the bottom plate surface, these shear waves generate an additional converging or diverging acoustic beam with a different focus position [64]. The $A(z)$ dependence in this case has two maximums. The main maximum is shifted more and pertains to the longitudinal waves traversing the plate. The additional maximum arises due to the transverse waves. It is shifted to a lesser extent because its shift depends on the velocity of transverse waves in the material. Measuring the two shifts by the method of $A(z)$ characteristics, one can assess local distributions of the velocities of both longitudinal and transverse acoustic waves in the material [65]. Bifurcation of the maximums of the $A(z)$ curve was observed in studies of a dry gelatin film 7 microns thick at the same parameters of the setup (450-megahertz frequency, lens aperture of 30°) using mercury as an immersion liquid. Measurement of the two maximums permitted the longitudinal and shear wave velocities in the sample to be calculated and the vanishing of the shear modulus to be traced in samples swollen with water [66].

Another interesting example is the generation of a Lamb leaky wave with a set of modes in an individual layer or multi-layer system with dissimilar impedance and low absorption. This wave can significantly contribute to the received signal of the microscope operating in the reflection or transmission mode.

All the above examples can be related to the field of purely physical investigations; therefore, it is expedient to dwell, at least briefly, on the practical value of particular applications.

1.4
Methodological Limitations of Acoustic Microscopy

The acoustic microscopy technique is fairly sensitive to the presence of various inhomogeneities in a sample and to breaks of material continuity because mismatch of acoustic impedances at the boundaries brings about intense reflections. At present, acoustic microscopy allows one to reveal the following defects: failure of adhesion, exfoliation, microcracks, alien inclusions, deviations from the preset layer thickness in multi-layer systems and coatings, technological deviations in sizes, orientation and distribution of grains.

Based on the aforesaid, we believe that the following points are important in developing the methods of reflection and transmission acoustic microscopy and are promising in studies of surface and subsurface structures of diverse materials:

- The topography of surfaces, including height of steps, width of cracks and the pattern of mechanical stress fields around them, the curvature radii of bulges or concavities, wedge angles, etc.
- The morphology of smooth surfaces with inhomogeneous distribution of acoustic properties, including characterization of individual components of granulated and laminated structures, acoustic images of internal planes, structures, grains, and analysis of thin-filmed heterogeneous objects.
- The measurement of local values of the propagation velocity and attenuation of Rayleigh waves in materials employing acoustic microscopy techniques in which spherical and cylindrical lenses are used.
- The study of distributions of local anisotropic elastic properties in crystals and other materials.
- Quantitative measurements of the mechanical properties by acoustic microscopy techniques, including local measurements of piezoelectric, photoelectric, and high-temperature superconducting properties of films.
- Studies of dynamic phenomena associated with reconstruction of the material properties induced by varying physical factors (temperature, ultraviolet (UV), infrared (IR), and superhigh frequency impacts) and also by mechanical, chemical, and pharmacological impacts.