Ultra resolution in acoustic imaging of bulk microstructure in solids

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Abstract

Impulse acoustic visualization of bulk microstructure in solid specimens is performed in the dark-field regime. Dark-field regime images display small details of microstructure and their distribution over the specimen bulk - spatial resolution of the method is derived by efficiency of ultrasonic scattering at small obstacles and sensitivity of an ultrasonic radiation-reception system, but not by the probe radiation wavelength (ultramicroscopic technique). The technique is attractive when efforts are concentrated at revealing presence and location of small bulk microstructure elements (microdefects, particles distribution in composite media and so on). Detailed knowledge of their shape could not be reached with this regime. In the paper theoretical analysis of interaction of probe ultrasonic radiation with diverse types of small inclusions, both acoustically soft and hard, has been done. It was shown ultrasound interaction with soft obstacles is much more effective than it is for hard inclusions. It is the main source of the contrast in acoustic images of the bulk microstructure for diverse types of solids. Performed theoretical assessments have been employed for interpreting acoustic images of bulk microstructure in the graphite-epoxy nanocomposites.

Keywords: acoustic imaging; bulk microstructure; acoustic ultra microscopy

1. Introduction

Possibility to observe details appreciably smaller than the wavelength of probe radiation is the ultimate goal of the most of visualization techniques. There are two main ideas for realization of this intention. In scanning near-field microscopy application of small-sized diaphragms and tight arrangement of the radiation source and receiver to the specimen face makes it possible to resolve details in the specimen surface that are essentially smaller than the probe radiation wavelength (Courjon, 2003). The other approach is ultramicroscopical regime of imaging that is

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employed for visualizing fine microstructure inside the specimen bulk. The technique is applied to dispersion of small particles distributed in the object interior. Interaction of the probe radiation with particles gives rise to secondary radiation scattered by the particles. Particles are perceived by the receiver as point sources; they are displayed in images as small bright spots against the dark background corresponding to parts of the specimen volume that are free of scatterers. Spots that depict small-sized scatterers do not depend on shape and sizes of particles – they are described by the point spread function of the vision system and coincide with the focal spot of the focusing system (Airy disk). Images performed in ultramicroscopical regime give information on presence of a dispersive phase and respectively, brightness of corresponding imaging spots. The efficiency falls with decreasing characteristic particle size $a$; sensitivity of the receiver sets a limit size of particles that could be seen in corresponding images.

In light microscopy the ultramicroscopical regime corresponds to dark-field microscopy. Present paper is aimed at establishing the basic principles of fine bulk microstructure imaging in impulse acoustic microscopy. General theory of focusing acoustic beam interaction with spherical elastic objects of arbitrary sizes has been developed in the frame of Fourier-optics earlier in (Zinin, 1997). Our paper is focused at analysis of small-particle visualization features that were not considered before.

2. Theoretical analysis and experimental results

Application of ultra-short probe pulses of focused ultrasound and time selection of echo signals coming from structural elements situated at different depth underlie ultrasonic bulk visualization technique – impulse acoustic microscopy (Gilmore (1999); Zakutailov (2010)). Secondary radiation coming back from the specimen forms an echo pattern at each point of the scanning area. Echo patterns are seen as sets of individual echo pulses separated by time intervals equal to double time spent by the probe pulse to run the difference in depth positions of two corresponding structural elements; the pulse reflected from the specimen face is used as the reference signal. Data on the recorded signal at each point of the scanning area save in the computer memory together with coordinates of the observation point and, then, are employed to produce acoustic images – C-scans. A special electronic instrument – electronic gate; is used to organize the imaging process. It is a pair of switches that sets depth and thickness of an imaging layer inside of the specimen bulk. A value of the echo signal within the electronic gate is displayed as a pixel at a corresponding point of a raster grey-scale image; brightness of the pixel is determined by the signal value. Scattered radiation provides imaging small-scale inclusions, sizes of which could be appreciably smaller the diffraction limit. Principles of ultramicroscopical imaging in acoustic microscopy are presented in Fig.1. Low-aperture probe beams and ultrashort pulses of focused ultrasound are employed for bulk visualization. Wave field within the focal zone of the beam could be treated as a bounded plane wave: $p_0 \cdot e^{j(kz - \omega t)}$, where $\omega = \alpha/c$, $c$ is sonic velocity in immersion (Born, 1968). The zone is a long cylinder with diameter $d_F = 0.61 \lambda/\theta_b$ and length $L_F = 4 \lambda/\theta_m^2$ ($\lambda$ – ultrasonic wavelength, $\theta_b$ – half-aperture angle of the beam). For $\theta_m = 11^\circ$. At $f = 100$ MHz: $d_F = 50 \mu$m; $L_F = 1.5$ mm. The incident beam partly reflects from the interface, partly penetrates into the specimen bulk as bounded plane waves. The reflected wave: $p_0 \cdot R \cdot e^{j(kz - \omega t)}$, goes out the focal area and is received by the focused system as a divergent beam coming from its focus. $R$ is the reflection coefficient at the interface $R = (\rho_{cL} - \rho c)/\rho_{cL} + \rho c)$. $\rho$ and $\rho c$ - density and sonic velocity, $\rho_{cL} = \rho c$ and $L$ are density and longitudinal wave velocity in the specimen.

The refracted beam enters in a specimen as a bounded plane wave: $p_0 \cdot T_1 \cdot e^{j(kz - \omega t)}$, $k_L = \alpha a_{cL}$ is wave number of longitudinal elastic wave and $T_1 = 2\rho_{cL}/(\rho_{cL} + \rho c)$ is the coefficient of transmission from the immersion into the specimen. Interaction of the transmitted radiation with a small inclusion of size $a$ could be considered as scattering of the bounded plane wave a small particle. Scattered radiation arises only when the inclusion is within of the transmitted beam aperture determined by the focal spot on the immersion-specimen interface. Small-sized scatterers are observed as bright spots of the same size $d_F$ independently on their real size $a$ (Fig.1). Sizes of particles affect only brightness of the spots – it decreases with lowering particle sizes. Dependence on particle sizes and shape appears in scattered radiation only when scatterer sizes are compared with the focal spot diameter.

The probe impulse possesses a finite time extent $\Delta t$ (usually $1.5 \pm 2$ oscillations at the nominal operation frequency $\alpha$). The same time extent is characteristic for the echo pulse reflected from the specimen face. Because of presence of the high-level echo signal reflected from the specimen face the scattered radiation could be resolved by the receiver only from scatterers underlying the specimen face at distances $l$ bigger than thickness $l_0 = (1.5 + 2)(a_{cL}/c)\lambda$ of a layer corresponding to the probe pulse time extent $\Delta t$. 
When the scatterer is inside the entrance aperture of the transmitted beam, it gives rise scattered spherical wave:

\[ p_0 \cdot T_1 \cdot A \cdot e^{ik_1 t} \cdot [e^{ik_1 r \cdot \text{Im}} / r], \quad (1) \]

where \( r \) is distance from the scatterer to an observation point, \( p_0 T_1 \) – amplitude of the incident beam, \( A \) – amplitude of scattering. If scatterer position \( l \) is outside the nearest subsurface layer with thickness \( l_0 \) all scattered rays arrive at points of the focal spot with close phases. The back scattered wave within the focal spot could be treated as a plane wave; it goes out the specimen as a bounded plane wave:

\[ p_0 \cdot T_1 \cdot T_2 \cdot (A/l) \cdot e^{ik_1 t} \cdot e^{-ik_2 z \cdot \text{Im}}, \quad (2) \]

where \( T_2 = 2 \rho c/(\rho c_L + \rho c) \) - coefficient of transmission from the specimen bulk into immersion.

Output signal \( V \) of the focusing system:

\[ V = V_B + V_{sc}, \quad V_B = B \cdot p_0 \cdot R \cdot e^{-\text{Im} t} \quad \text{and} \quad V_{sc} = B \cdot p_0 \cdot T_1 \cdot T_2 \cdot (A/l) \cdot e^{-i(\omega t - 2z/c_1)}. \quad (3) \]

To receive scattered echo signals their values should lie within dynamic range \( S \) of the ultrasonic focusing receiver. The dynamic range is defined as the ratio of the maximal output signal \( V_{max} \) to the noise signal \( V_{\text{noise}} \): \( S = V_{max} / V_{\text{noise}}. \) The value of \( S \) is found from the echo patterns for acoustic microscopes being in use it close to 40 dB.

A value of the scattered echo signal \( V_{sc} \) is determined by the back-scattering amplitude. Its finding is a routine procedure of the scattering theory. All wave fields are represented as sets of spherical harmonics \( P_m(\cos \theta) \) in the spherical coordinate system with the center at the point scatterer and the polar axis directed along the plane wave incidence direction. Expansion of the incident plane wave \( p_m \) with terms of spherical harmonics is well known:

\[ p_m (\vec{r}, t) = p_{00} \cdot \sum_{m=0}^{\infty} \sum_{l=0}^{\infty} (2m+1) \cdot i^m \cdot j_m (k_1 r) \cdot P_m (\cos \theta) \cdot e^{i\omega t r}. \quad (4) \]

The scattered \( p_m \) in the surrounding medium outside the obstacle and standing wave field \( p_b \) inside its volume are expressed through amplitudes \( A_m \) and \( B_m \) of the spherical harmonics \( P_m(\cos \theta) \):

\[ p_{sc} (\vec{r}, t) = p_{00} \cdot \sum_{m=0}^{\infty} A_m \cdot h_m^{(1)} (k_1 r) \cdot P_m (\cos \theta) \cdot e^{-i\omega t r}, \quad (6) \]

\[ p_b (\vec{r}, t) = p_{00} \cdot \sum_{m=0}^{\infty} B_m \cdot j_m (k_1 r) \cdot P_m (\cos \theta) \cdot e^{-i\omega t r}. \quad (7) \]

Here \( p_{00} \) is amplitude of the plane wave incident onto the obstacle; \( \rho_1 \) and \( c_L \) are density and sonic velocity in the specimen, \( \rho_b \) and \( c_b \) are density and longitudinal wave velocity in the obstacle; \( k_L = \omega c_L \) and \( k_b = \omega c_b. \) Amplitudes \( A_m \) and \( B_m \) are derived from the boundary conditions – continuity of pressure and normal components of the
oscillation velocity at the “surrounding medium – obstacle” interface.

For small scatterers contributions of spherical harmonics of the lowest orders — \( n = 0 \) (spherically symmetric mode) and \( n = 1 \) (dipole mode) are important only:

\[
p_sc(\bar{r}, t) = -p_{\infty} \cdot \left\{ i \cdot A_0 + A_1 \cdot \cos \theta \right\} \cdot \left( e^{i k_L \cdot r \cdot \cos \theta} / k_L r \right).
\]  

(8)

Two limit cases of scattering – at hard and soft particles; are of special interest. In the case of very stiff scatterers in a solid matrix: \( \rho_b c_b / \rho_1 c_L >> 1 \); amplitudes of the both modes are of the same order. They are proportional to the obstacle characteristic sizes \( a \) and squared small parameter \( ka << 1 \):

\[
p_sc(\bar{r}, t) = -(1/3) p_{\infty} \cdot \left( e^{i k_L \cdot r \cdot \cos \theta} / r \right) \cdot \left\{ 1 - (3/2) \cdot \cos \theta \right\} \cdot \left( k_L \cdot a \right)^2.
\]  

(9)

Back-scattering amplitude for stiff scatterers is \( A_{\text{stiff}} = -(5/6) a \cdot (k_L \cdot a)^2 \). It gives the total cross-section: \( \sigma = 4\pi a^2 \cdot (k_L \cdot a)^4 \) well-known in optics for Rayleigh scattering.

The formula of the back-scattering amplitude allows getting expression for the scattered echo pulse in the case of stiff inclusions:

\[
V_{sc} = -B \cdot p_0 \cdot T_1 \cdot T_2 \cdot \left( e^{-i\omega t} / c_L \right) \cdot \left[ (5/6) \cdot a \cdot (k_L \cdot a)^2 \right].
\]  

(10)

Amplitude of the echo pulse is proportional to the obstacle size \( a \) and the small parameter \( (ka)^2 << 1 \).

The inverse relationship between acoustic impedances: \( \rho_b c_b / \rho_1 c_L << 1 \), is characteristic for soft inclusions. The best examples of this kind of inclusions are presented by gas bubbles and small particles with large air content submerged in a solid matrix. For such obstacles the amplitude \( A_0 \) of the spherically symmetric mode is proportional to the obstacle size \( a \) only, it does not include the small parameter \( (ka)^2 << 1 \). It is, respectively, much bigger than the dipole mode amplitude \( A_1 \sim a \cdot (ka)^2 \). The scattered field takes form:

\[
p_sc(\bar{r}, t) = -p_{\infty} \cdot \left( e^{i k_L \cdot r \cdot \cos \theta} / r \right)\cdot \left( k_L \cdot a \right)^2.
\]  

(11)

The back-scattering amplitude: \( A_{\text{soft}} = -a \), gives the total cross-section \( \sigma = 4\pi a^2 \cdot (k_L \cdot a)^4 \) equal to the geometrical cross-section of a scatterser for soft air-filled small particles are significantly larger then for stiff particles. Accordingly, the amplitude of echo signal caused by scattering of probe radiation at air-filled small particle:

\[
V_{sc} = -B \cdot p_0 \cdot T_1 \cdot T_2 \cdot \left( e^{-i\omega t} / c_L \right) \cdot a,
\]  

(12)

does not contain the small-valued factor \( (ka)^2 << 1 \), and it is much bigger than an echo pulse caused by scattering at a hard particles of the same sizes.

To be seen in acoustic images small scattersers should provide the output signal \( V_{sc} \) and back-scattering amplitude \( A \) that satisfy the requirement: \( V_{sc} > V_{\text{noise}} \) or \( V_{sc} / V_B > V_{\text{noise}} / V_B = 1 / S \). Different amplitudes of back scattering at hard and soft obstacles result in different values of minimal sizes of inclusions to be seen in acoustical images. The limit size of soft obstacles:

\[
a_{\text{soft}} = \frac{1}{S} \cdot \frac{R}{T_1 \cdot T_2} \cdot l,
\]  

(13)

is linearly depends on the depth location of the particle when the particles are located outside the layer \( l \) that is perceived by the ultrasonic receiver as the specimen front border. The same limit size of stiff scattersers:

\[
a_{\text{stiff}} = 0.31 \cdot \sqrt{\frac{1}{S} \cdot \frac{R}{T_1 \cdot T_2} \cdot \left( \frac{c_L}{c} \right)^2 \cdot \sqrt{l \cdot \lambda^2}}
\]  

(14)

is characterized by softer dependence on the depth position.
Assessments give essentially higher efficiency of probe ultrasonic scattering at soft inclusions. For nanocarbon-epoxy nanocomposites \((\rho_1 \approx 1.08 \text{ g/cm}^3; c_L \approx 2.9 \text{ km/s}, \rho/c_L \approx 1/3, R \approx 4/3, T_1 \approx 2/3, T_2 \approx 3/8)\) the minimal size of an soft obstacle visible at depth of \(4\lambda\) is \(\approx 1.5 \cdot 10^{-2} \lambda\) (\(\approx 0.2\div0.3 \mu\text{m} \text{ at } 100 \text{ MHz})\). The minimal size of a hard obstacle visible in carbon nanocomposites at the same depth is by an order bigger: \(\approx 12.5 \cdot 10^{-2} \lambda\) (\(\approx 2 \mu\text{m} \text{ at } 100 \text{ MHz})\). In fig.2 acoustic image of air-filled conglomerates of carbon nanoparticles inside the epoxy matrix is shown. Size of nanoparticles was \((7\div10) \text{ nm} \times (0.5\div5) \mu\text{m}\).

![Acoustic image](image)

Fig.2 Acoustic image of air-filled nanoparticles conglomerate distribution inside the graphite nanoplatelet-epoxy composite (1.56 mm thick) at the depth of 0.7 mm. Scanning area 10×11 mm.

3. Conclusions

Radiation scattered by small-sized inclusions with characteristic sizes appreciably smaller than the probe ultrasound wavelength gives meaning contribution into imaging of bulk microstructure in composite specimens. Ultra resolution is realized in the ultramicroscopical regime of imaging. Ultramicroscopical ultrasonic visualization makes it possible to reveal presence of micron- and submicron-sized structural elements and to observe their distribution over the specimen volume. Acoustic images could not give any information on particle sizes and shape. Scattering at soft inclusions – air-filled agglomerates and particles, gas bubbles, cavities and so on; are essentially more efficient than ordinary Rayleigh scattering at small stiff particles.

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