

Advances in applications of time-domain Brillouin scattering for nanoscale imaging

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Citation: [Applied Physics Reviews](#) **5**, 031101 (2018); doi: 10.1063/1.5017241

View online: <https://doi.org/10.1063/1.5017241>

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APPLIED PHYSICS REVIEWS—FOCUSED REVIEW

Advances in applications of time-domain Brillouin scattering for nanoscale imaging

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(Received 26 November 2017; accepted 21 May 2018; published online 12 July 2018)

Time-domain Brillouin scattering is an all-optical experimental technique based on ultrafast lasers applied for generation and detection of coherent acoustic pulses on time durations of picoseconds and length scales of nanometers. In transparent materials, scattering of the probe laser beam by the coherent phonons permits imaging of sample inhomogeneity. The transient optical reflectivity of the sample recorded by the probe beam as the acoustic nanopulse propagates in space contains information on the acoustical, optical, and acousto-optical parameters of the material under study. The experimental method is based on a heterodyning where weak light pulses scattered by the coherent acoustic phonons interfere at the photodetector with probe light pulses of significantly higher amplitude reflected from various interfaces of the sample. The time-domain Brillouin scattering imaging is based on Brillouin scattering and has the potential to provide all the information that researchers in materials science, physics, chemistry, biology, etc., get with classic frequency-domain Brillouin scattering of light. It can be viewed as a replacement for Brillouin scattering and Brillouin microscopy in all investigations where nanoscale spatial resolution is either required or advantageous. Here, we review applications of time-domain Brillouin scattering for imaging of nanoporous films, ion-implanted semiconductors and dielectrics, texture in polycrystalline materials and inside vegetable and animal cells, and for monitoring the transformation of nanosound caused by nonlinearity and focusing. We also discuss the perspectives and the challenges for the future. *Published by AIP Publishing.* <https://doi.org/10.1063/1.5017241>

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E. Subcellular imaging inside plant and animal cells	11	Imaging materials and probing their properties (morphology, electrical, and magnetic properties, for example) at the nanoscale with near-field methods such as Scanning Force Microscopy, ¹ including all its extensions (Atomic Force Microscopy—AFM, ^{2,3} magnetic Force Microscopy—MFM, ⁴ piezoresponse Force Microscopy—PFM, ⁵ Scanning Tunneling Microscopy—STM, ⁶ Near-field Scanning Optical Microscope—NSOM, ⁷ and AFM-based nanotribology tools ⁸) has been significantly improved since the 1980s and represents now an array of powerful diagnostics in materials science, in nanotechnology, and in biology, as well. ⁹ When the properties or characteristics of interest are not limited to the surface or/and not accessible by the near field methods,	

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i.e., it means when the region to be probed is beneath the surface typically (10 nm–1 μ m), different strategies are required to get the information, most of them being based on the use of a penetrating beam or wave to probe the materials. Electron beams can be used to realize subsurface image of materials by cross-sectional analysis with atomic resolution through the well-known Transmission Electron Microscopy—TEM or with high resolution transmission electron microscopy—HRTEM but these techniques require, however, individualized sample preparation and materials that do not undergo damage under electron beams irradiation.¹⁰ X-Ray Reflectivity (XRR) is also widely used for the studies of thin and ultra-thin films, and with specific algorithms, permits reconstruction of the structure profile of these films.¹¹ Optical super resolution methods have been recently developed for biological materials imaging including photo-activated localization microscopy (PALM) and stochastic optical reconstruction microscopy (STORM).^{12,13} In general, optical confocal spectrometry such as used in Brillouin and Raman spectroscopies permits subsurface investigation through depth dependent lattice dynamics. But even with latest confocal optical geometry, the Brillouin and Raman spectroscopy based imaging of materials remains limited to micrometer scale.^{14,15} Besides electromagnetic waves and electrons or particles beams, the use of penetrating acoustic waves is also very widely used for imaging in both materials science and medicine.^{16–20} The so-called “echography” is based on the analysis of reflection and transmission of ultrasound with various extensions (which is out of the scope of this review). Echography is also well-known in structural health monitoring for investigation at a macroscopic scale and is based on imaging with traditional bias-controlled piezoelectric transducers that are used to emit and receive the sound waves. Standard medical ultrasound imaging gives an in-depth resolution not better than tens of micrometers, because of MHz limited instrumental bandwidth and of the viscous damping of sound at the frequencies higher than a few MHz, thus precluding nanoscale imaging. Nanoscale imaging can be achieved using particular methods of picosecond ultrasonics.

Picosecond ultrasonics, pioneered in the 1980s,^{21–25} is a technique that uses ultra-short pump laser pulses to generate coherent acoustic pulses (CAPs) through the optoacoustic effect and uses time-delayed, ultra-short probe laser pulses to detect the CAPs (through the acousto-optic effect) as the CAPs propagate through a transparent medium or reach surfaces or interfaces of an opaque medium [see Fig. 1(a)]. A thin film pump-light-absorbing optoacoustic generator for launching the CAPs into transparent samples can be located on the interface of a film and a substrate^{26–28} as in Fig. 1(a). The thin film generator (semitransparent for probe laser pulses) can be also deposited on the front surface of a transparent film or bulk material.^{23–25,29–31} Even a single atom layer light absorber³² can be used to generate the CAPs. There is no need for additional optoacoustic generators in the case of a pump light absorbing substrate, film, or bulk material.^{26,33–35} The mechanism of the optoacoustic transformation is mainly thermo-elastic, i.e., deformation of the material by stresses induced by laser-induced heating.^{22,36} The other mechanisms, such as electron-hole-phonon

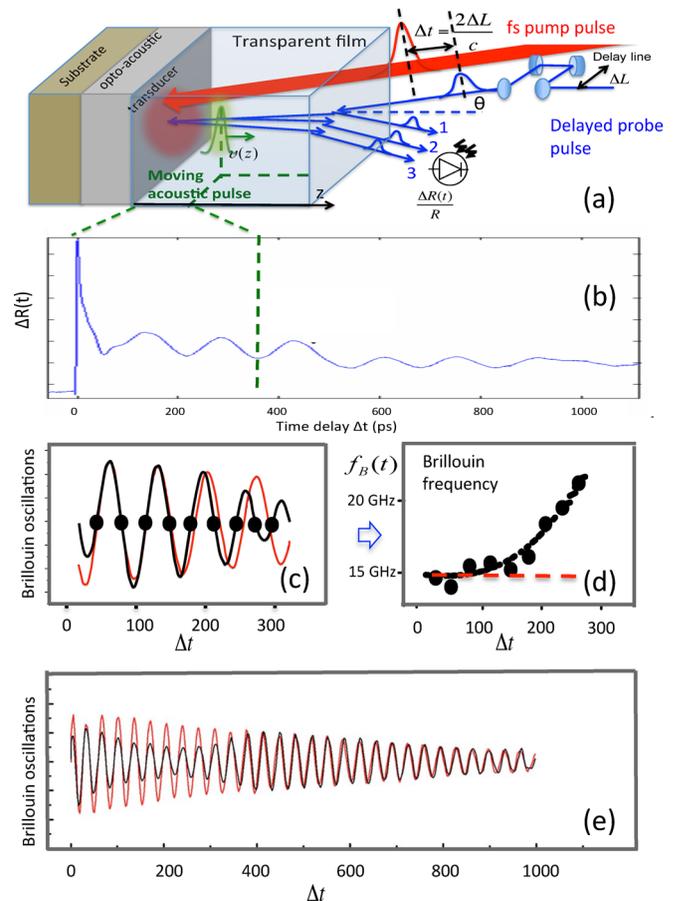


FIG. 1. Principles of opto-acousto-optic depth profiling by picosecond acoustic interferometry (time-domain Brillouin scattering). (a) A thin film pump-light-absorbing optoacoustic generator for launching the coherent acoustic pulses into transparent samples is located on the interface of a film and a substrate. The transient optical reflectivity signal (b) contains a sinusoidal oscillating component, the Brillouin oscillation. The Brillouin frequency in an inhomogeneous film can change with time.²⁸ In the left part of (c), a comparison of the experimental Brillouin oscillation, in black, with model constant-frequency Brillouin oscillation, in red, reveals an increase in the experimental Brillouin frequency, also evidenced by the Fourier transform in a moving time window in the right part of the figure. Reproduced with permission from A. M. Lomonosov *et al.*, ACS Nano **6**, 1410 (2012). Copyright 2012 American Chemical Society. In (d), the Brillouin oscillation amplitude in an inhomogeneous medium, in black, exhibits variations in addition to those caused by the probe photons and the CAPs absorption in homogeneous media, shown in red,⁴² while the Brillouin frequency is fairly constant. Reproduced with permission from Appl. Phys. Lett. **94**, 111910 (2009). Copyright 2009 AIP Publishing LLC.

deformation potential, the inverse piezoelectric effect, electrostriction, etc., can be efficient under particular circumstances.^{22,36–38} The duration of the CAP depends, in general, on the duration of the pump laser pulses, the lifetime of the photo-induced mechanical stresses, and the time of sound propagation across the localization depth of the photo-induced mechanical stresses, which, in turn, depends on the pump light absorption depth.^{22,36,37}

In the case where sound generation takes place in an opaque (metallic) thin film attached to the sample, as shown in Fig. 1(a), the thickness and absorption of the thin film determine the acoustic pulse length. Through optimization of the optical and material factors involved in producing the acoustic wave, it is possible to generate CAPs of picoseconds

duration with spatial lengths of nanometers making them suitable for nanoscale imaging.

In application to transparent materials, the technique of picosecond ultrasonics was originally referred to as picosecond acoustic interferometry (PAI).^{23–25} In optically transparent materials (see Fig. 1), the probe laser pulse preferentially interacts with those GHz-frequency phonons in the CAP spectrum that satisfy the momentum conservation law for photon-phonon, photo-elastic interaction, that is, they satisfy the Brillouin scattering (BS) condition.^{22,39,40} The experimental method is based on heterodyning where weak light pulses scattered by the coherent acoustic phonons (beam 2 in Fig. 1(a), for example) interfere at the photodetector with probe light pulses of significantly higher amplitude reflected from various interfaces of the sample [beams 1 and 3 in Fig. 1(a), for example]. The recorded transient optical reflectivity signal is proportional (to first order) to the product of the electric fields of the two light fields. Thus, heterodyning of a weak field against a strong one is achieved. The measured signal varies with time because the relative phase of the light scattered by the CAP and reflected by immobile interfaces continuously changes with time due to the variation in the spatial position of the CAP. If the CAP propagates at a constant velocity in a spatially homogeneous medium, the phase difference between the interfering light fields changes linearly in time causing sinusoidal variations in the signal amplitude at a frequency precisely equal to the Brillouin frequency (BF)^{22–25} [see Fig. 1(b) and Sec. II]. The acoustically induced oscillating contribution to the reflectivity signal [see Figs. 1(c) and 1(e)] is commonly known as the Brillouin oscillation (BO). In view of the above-mentioned intrinsic relations of the PAI to the Brillouin scattering, the technique is currently more and more often called the time-domain Brillouin scattering (TDBS). In the collinear scattering geometry, the BF depends linearly on the product of the sound velocity and the optical refractive index of the sample at the probe wavelength. The potential of the PAI for depth-profiling of the materials has been explicitly pointed out in Ref. 24, viz., “Velocity variations as a function of distance from the surface can be determined from a measurement of how the period of oscillation varies with time delay of the probe pulse.” Surprisingly though, the first experimental application of the PAI for depth-profiling was published almost 20 years later.⁴¹ At the same time, it was shown

experimentally⁴² that the information on material spatial inhomogeneity can be revealed by monitoring the evolution in time not of the frequency of the BO, but rather of its amplitude. Indeed, the amplitude of the acoustically induced transient reflectivity signal is proportional to the strength of the acousto-optic interaction, that is, to the photo-elastic constant characterizing variation of the optical properties of the probed media with the acoustic strain.^{39,40} An inhomogeneity in the material photo-elasticity could be the dominant mechanism providing the contrast for depth-profiling by the TDBS.^{28,42} Following the first experiments on depth-profiling of ion-implanted crystals⁴² and nanoporous thin films,⁴¹ progress was reported in both of these applications^{28,43–46} and extended to imaging of radiation damage.^{43–45} More recently, the TDBS was applied to two and three-dimensional imaging of textured polycrystalline materials at high pressures^{47,48} and inside both plant and animal cells.^{49–51} Very recently, the TDBS was used to reveal the gradient of the temperature distribution in liquid.⁵² The imaging was based on analysis of the temporal evolution of either the BF^{47,50,52} [Fig. 1(c)] or the BO amplitude^{43–46} [Fig. 1(e)], or of both.^{28,48,49} An axial resolution of about 40 nm was achieved in low-k nanoporous sub- μm thick films^{28,41} and of about 30 nm in ion-irradiated semiconductors.^{42,43}

Among other imaging techniques based on the combination of coherent acoustical and optical waves, it is worth mentioning for comparison the optoacoustic (photoacoustic) microscopy^{53–60} applied for imaging at mm/ μm spatial scales. In optoacoustic imaging, coherent acoustic waves, thermo-elastically generated by the optically induced heating of spatially inhomogeneous media, read the information on the inhomogeneity and deliver it to the detection modality. The contrast is primarily due to spatially inhomogeneous optical absorption. Photoacoustic microscopy (PAM) and optical-resolution photoacoustic microscopy (OR-PAM) have seen their widest application in biology and medicine.^{55–60} The physical principles of TDBS imaging and the origin of the contrast in TDBS imaging are drastically different from those in optoacoustic (photoacoustic) imaging. In TDBS imaging, coherent ultrashort acoustic pulses travel in the inhomogeneous medium, while probe light pulses read information on the material parameters at the current position of the coherent acoustic pulse and deliver it to the detection modality. The contrast in TDBS imaging is usually a result of the spatial inhomogeneity of optical properties (refractive index), acoustical properties (sound velocity and acoustic impedance), and/or opto-acoustic properties (photoelastic tensor).

Speaking generally, TDBS imaging is based on Brillouin scattering and has the potential to provide all the information that researchers in materials science, physics, chemistry, biology, etc., get with classic frequency-domain Brillouin scattering (FDBS) of light. However, TDBS incorporates Brillouin scattering not by the thermal phonons that exist at all points of the sample, but rather by the coherent phonons of much higher amplitudes intentionally launched into and highly localized in the sample. Thus, in addition to its reliability and speed for three dimensional imaging of transient processes at nanoscale, TDBS can be viewed as a replacement for Brillouin scattering and Brillouin

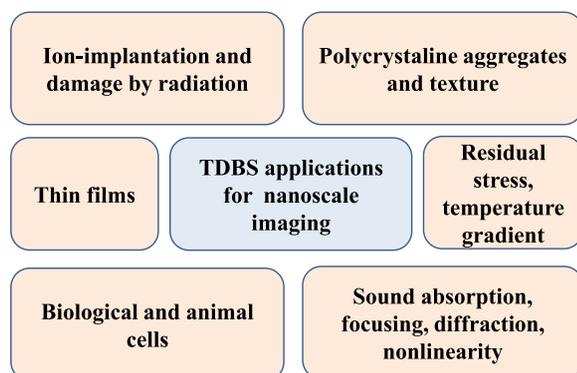


FIG. 2. Applications of time-domain Brillouin scattering for imaging.

microscopy^{14,15} in all investigations where nanoscale spatial resolution in materials characterization is either required or advantageous (Fig. 2).

Below we review some research results constituting the background for the progress achieved in TDBS imaging and the results of several imaging experiments. We also discuss the applications of the TDBS for monitoring of nonlinear acoustic processes^{61,62} and hypersound nanofocusing⁶³ as well as perspectives and the challenges for the future.

II. THEORETICAL BACKGROUNDS FOR IMAGING VIA TIME-DOMAIN BRILLOUIN SCATTERING

In time-domain Brillouin scattering (TDBS), probe laser pulses are scattered by laser-generated picosecond coherent acoustic pulses (CAPs) due to the acousto-optic effect.^{22,39,40} By monitoring the reflected light, information on the time development of the CAPs is obtained as they travel through the experimental sample.²² In most of the experiments, the transient relative changes $dR(t)/R$ in the intensity of the reflected probe light are detected. The reflectivity R and electromagnetic field reflectivity r are related by $dR(t)/R = 2\text{Re}[dr(t)/r]$. Among possible contributions to this signal, there is one generated by the acousto-optic interaction, which is proportional to the photo-elastic (acousto-optic) coefficient and the overlap integral of the acoustic strain field with the optical field sensitivity function.²² The integral is over the complete volume probed by the light, while the sensitivity function depends on the specific geometry of the light distribution in the sample in the absence of the CAPs.^{22,64,65} The integral gives the solution for electromagnetic wave propagation equation in an optically inhomogeneous medium under the single-scattering approximation,^{66–68} which is valid because the perturbation of optical properties by the CAP is weak. In particular, when light normally incident on the sample surface probes the propagation of the acoustic pulse in an optically and acousto-optically homogeneous half space (Fig. 3), the photo-elastic contribution is given by

$$\frac{dr}{r} = -iC \int_0^{\infty} \eta_{zz}(t, z) e^{i2kz} dz, \quad (1)$$

where C is the known complex constant proportional to the acousto-optic (photo-elastic) interaction,^{22,64,65} $\eta_{zz}(t, z)$ is the strain field of the longitudinal CAP propagating along the z axis, and k is the complex wave number of the probe light in the medium. Thus, $dr(t)/r$ provides information on the

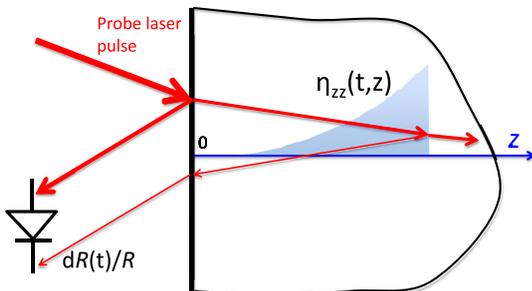


FIG. 3. Time-domain Brillouin scattering in a half space.⁶⁵ The laser beams are shown at an angle for clarity. Reproduced with permission from J. Appl. Phys. **110**, 124908 (2011). Copyright 2011 AIP Publishing LLC.

evolution in time of the Laplace component of the acoustic field with the complex-valued Laplace parameter $p = -i2k$.⁶⁴

Other interpretations of Eq. (1) can be given, which could be more insightful for the physical understanding of specific experimental realizations. For the TDBS imaging, it is essential that the strain field $\eta_{zz}(t, z)$ in Eq. (1) at each time instant is strongly localized in space in the form of a nanoscale CAP. For example, for a CAP propagating in the positive z direction starting at $t=0$ from $z=0$ (Fig. 3), slow evolution of its strain field in a medium with inhomogeneity of the acoustic parameters only can be written in the form

$$\eta_{zz}(t, z) \equiv \eta_{zz}(t, \xi = z - z_a(t)) = \int_{-\infty}^{+\infty} \tilde{\eta}_{zz}(t, q) e^{-iq\xi} dq / (2\pi). \quad (2)$$

Here, $z_a(t) \equiv \int_0^t v_s(t') dt'$ describes the variation of the CAP position in space, v_s is the velocity of the CAP, which depends on the co-ordinate and, thus, is different in the different time instants of CAP propagation, while the first argument of the strain function describes possible slow evolution of the CAP's amplitude and profile in the time domain or of the components of the CAP spectrum $\tilde{\eta}_{zz}(t, q)$ in the reciprocal q space. Slow evolution of the CAP takes place if the width/length of the CAP spatial localization region, $l_a(t)$, is significantly shorter than a characteristic spatial scale for the variation of the acoustical parameters of the medium and the characteristic lengths of dispersion, diffraction, and non-linearity.^{36,37,67–69} However, here for the simplicity, we completely neglect possible transformation of the CAP, which could be caused by dispersion, diffraction, and nonlinear phenomena, in order to highlight the manifestations of the medium inhomogeneity only. A detailed derivation of the solution for dr/r , when the medium is also optically inhomogeneous and the propagation of probe laser radiation and a CAP are treated within the framework of geometrical optics and geometrical acoustics,^{67,68} respectively, can be found in Ref. 65. Substitution of the strain field (2) into Eq. (1) leads, after the change in the integration variable, to

$$\begin{aligned} \frac{dr}{r} &= -iC e^{i2kz_a(t)} \int_{-z_a(t)}^{+\infty} \eta_{zz}(t, \xi) e^{i2k\xi} d\xi \\ &\cong -iC e^{i2kz_a(t)} \int_{-l_a(t)/2}^{l_a(t)/2} \eta_{zz}(t, \xi) e^{i2k\xi} d\xi. \end{aligned} \quad (3)$$

The final form of the right-hand-side in Eq. (3) is derived under the assumption that the propagation distance of the CAP exceeds its spatial length, $z_a(t) \geq l_a(t)/2$. The solution in Eq. (3) is very instructive. First, it demonstrates that quasi-periodic oscillations of dr/r in time domain are caused by the time-dependent phase shift, $\varphi_D = 2kz_a(t) = 2k \int_0^t v_s(t') dt'$, of the probe light scattered by the CAP. This is just a classic Doppler phase shift of the probe light pulses when the ultra-fast laser is operating as radar to monitor the motion of the acoustically induced weakly reflecting mirror. The derivative of this phase shift over time provides the Doppler frequency shift of the probe light, $\omega_D = \partial\varphi_D/\partial t = 2kv_s(t)$. It is worth noting that neither φ_D nor ω_D depends on the acoustic pulse

profile (spatial spectrum). Under the assumed conditions, all the points of the CAP strain profile are propagating with the same local velocity, corresponding to the current spatial position of the CAP, while φ_D and ω_D themselves do not contain any indication on the preferential scattering of the probe light by a particular spectral component of the CAP. These theoretical predictions are independent of the $l_a(t)$ magnitude relative to the characteristic scale $\pi/k \equiv \lambda/2$ of the variations of the sensitivity function, $f(z) \propto \exp(i2kz)$, in Eq. (1). However, in the TDBS imaging experiments, the CAPs are dedicatedly generated of the nanoscale length $l_a(t)$ significantly shorter than the wavelength of the probe optical light in order to achieve imaging with sub-optical wavelength resolution. In this case, assuming $l_a(t) \ll \lambda/2$, Eq. (3) can be approximated by

$$\frac{dr}{r} \cong -iC e^{i2kz_a(t)} \int_{-l_a(t)/2}^{l_a(t)/2} \eta_{zz}(t, \xi) (1 + 2ik\xi + \dots) d\xi. \quad (4)$$

The solution in Eq. (4) demonstrates that in leading order the signal is proportional to the strain integrated over the localization region of the acoustic pulse (total “area” $S(t)$ of the strain pulse), i.e., to the displacement of the medium retained in it after the CAP propagation. Second term in Eq. (4) should be taken into account in the case of the symmetric bipolar CAPs of zero surface area, which could be generated by the pump laser pulses near the mechanically free surface of the medium.^{22,36,37} Equation (4) demonstrates that in homogeneous medium ($z_a(t) = v_s t$, $S(t) = S_0 = \text{const}$), the TDBS signal, $dr/r \cong -iCS_0 \exp(i2kv_s t) \propto f(z = v_s t)$, can be interpreted as an imaging of the spatial sensitivity function, while weak inhomogeneities of the medium could be revealed because they are causing deviations from this “background” image.

The fact that the probe light exhibits maximum scattering efficiency by the acoustic phonons with the wavenumber $q = 2k \equiv 2k_p$ is evidenced, when the spectral presentation of the CAP from Eq. (2) is substituted in Eq. (3) and the integration over the spatial coordinate is done

$$\frac{dr}{r} \cong -iC e^{i2kz_a(t)} \frac{1}{\pi} \int_{-\infty}^{\infty} \tilde{\eta}_{zz}(t, q) \frac{\sin[(q - 2k)l_a(t)/2]}{(q - 2k)} dq. \quad (5)$$

For the qualitative interpretation of Eq. (5), a precise definition of the characteristic dimension $l_a(t)$ of the CAP localization is not necessary. The earlier assumed inequality, $l_a(t) \ll \lambda/2 = \pi/k$, is sufficient to estimate that a broad spectrum of the acoustic wavenumbers contributes to scattering of the probe light in Eq. (5). The characteristic width of this spectrum, defined by $|q - 2k| \leq 2\pi/l_a(t) \gg 2k$, significantly exceeds $2k$. Thus, although the TDBS scattering exhibits the maximum sensitivity to the spectral component $\tilde{\eta}_{zz}(t, 2k)$, it is not a narrow band process and most of the CAP spectral components are efficient in light scattering to some extent. This broadening of the spectrum of coherent phonons that could contribute to light scattering with comparable efficiency is due to the smallness of interaction length of probe light with the coherent phonons that are carried by a nanoscale CAP. The relation $q = 2k$ is a well-known result of the momentum conservation in back-scattering of a photon by a phonon.^{40,66} However, the small interaction length

does not provide opportunity for the synchronous interaction of probe light with the $q = 2k$ phonons to “win” significantly relative to light scattering by other phonons.

Because of the relation $q \equiv \omega/v_s$, where ω is the cyclic frequency of the phonon, it is common to say that TDBS predominantly monitors the slow time dynamics of the spectral Fourier component of the CAP at the frequency $\omega = 2kv_s \equiv \omega_B$, known as the Brillouin frequency (BF) or the BF shift of a photon back-scattered by a phonon. The BF shift coincides with the Doppler frequency shift discussed earlier. Because of this, the oscillating TDBS signals, due to the time-dependent phase shift of the scattered light, are commonly called the Brillouin oscillations (BO). The wave number $q = 2k \equiv 2k_p$ and the wavelength $\lambda = 2\pi/q$ are known as the Brillouin wave number, q_B , and the Brillouin wavelength, λ_B , respectively.

In Ref. 69, it is demonstrated that the dominant contribution to the TDBS signal comes from the sharp fronts in the strain profile of the CAPs, as it is symbolically depicted in Fig. 3. This result is just a different formulation of the fact that light is not scattered inside a homogeneously strained medium. Thus, the spatial resolution of TDBS imaging can be better than the total spatial length of a CAP.

The separation in Eq. (5) of the amplitudes and the phases of C and of the integral I over the wavenumbers, i.e., $C = A_C \exp(i\varphi_C)$, $I(t) = A_I(t) \exp[i\varphi_I(t)]$, leads to $dR(t)/R = 2A_C A_I \sin(\omega_B t + \varphi_C + \varphi_I)$. Thus, the $dR(t)/R$ signal oscillates at a frequency given by

$$\omega_{dR/R} = \partial(\omega_B t + \varphi_C + \varphi_I)/\partial t = \omega_B + \partial\varphi_I/\partial t, \quad (6)$$

which, in general, differs from the BF defined above, i.e., $\omega_{dR/R} \neq \omega_B$. Equation (6) demonstrates that, in general, TDBS frequency is governed by the temporal variations of the phase of wide-frequency-band transient reflectivity signal in Eq. (5). For example, when TDBS is applied for the monitoring of the nonlinear CAP propagation, $\omega_{dR/R} \neq \omega_B$ provides information on the velocity of the weak shock front in the profile of the CAP.^{61,69}

In the conclusion of this theoretical section, we would like to attract attention of the readers to the fact that the TDBS imaging is neither optical nor acoustical imaging, but is acousto-optical imaging in a sense, where the signal is due to the existence of an overlap region of the optical probe and acoustical fields, Eq. (1), and TDBS imaging is an imaging of their interaction. This provides an explanation for why the TDBS imaging can achieve the spatial resolution much higher than that limited by the wavelength of the optical probe light or by the wavelength, $\lambda_B \equiv 2\pi v_s/\omega_B$, of the acoustic spectral component at Brillouin frequency, ω_B . The above presented theoretical formulas indicate that, although the probe light is interacting the most efficiently with a particular component of the CAP spectrum, with the wavelength equal to λ_B , this interaction takes place in the limited volume, which in optically transparent media is limited by the length of the CAP, l_a , and can be localized even in narrower region(s) corresponding to the sharp front(s) of the CAP. We denote here the length of the CAP front by l_f . Even l_a , not speaking on l_f , can be much narrower than λ_B . Two regions

in the medium separated by a distance l , which is larger than l_a but much shorter than λ_B , can be theoretically resolved by the TDBS imaging because they are probed not simultaneously, but separately, i.e., one by one, when the CAP is traversing the medium. This ability of the TDBS imaging is due to the application of ultra-short laser pulses. The ultra-short pump laser pulses provide opportunity to generate the CAPs on length scales of nanometers, while the time-delayed ultra-short probe laser pulses interact with the propagating CAPs quasi-instantaneously inside the volume currently occupied by the CAP. As a consequence, there is no interference in the TDBS imaging signal of the contributions coming from the regions separated by $l \geq l_a$. The time-dependent frequency of the Brillouin oscillation in continuously inhomogeneous medium, $\omega_B(t) \equiv \omega_D(t)$, although it contributes to the argument of the oscillating function in Eqs. (2) and (3), is a local parameter of the material at the scale of l_a or shorter scale of l_f , as far as $l_a \leq \lambda_B(t)$. As demonstrated by Eqs. (3) and (4), this parameter is determined independently of the spectrum of the acoustic pulse and is, thus, independent of the scale imposed by λ_B . The acoustic pulse of any profile will cause the same $\omega_B(t) \equiv \omega_D(t)$ when all the points of the profile are propagating at the same velocity. The complementary theoretical argument in favour of the TDBS spatial resolution superior to one limited by λ_B is the sensitivity of the TDBS to a large spectrum of the acoustic waves, including those at frequencies much higher than $\omega_B(t) \equiv \omega_D(t)$, as it is demonstrated by Eq. (5). As a consequence, the interpretation of TDBS as the scattering by the localized at the nanoscale moving strain gradients is closer to the physical reality than the interpretations based on the dominant or even predominant role of the BF phonons. Equation (5) explicitly describes a trade-off between the spatial and wave number resolution in TDBS.

The ultimate/highest spatial resolution in determining the parameter $\omega_B(t)$ is theoretically limited by l_a or l_f and theoretically does not require even a registration of at least a single period or a half of the period of the Brillouin oscillation. How to achieve this highest spatial resolution, if it is required in the experiments, by an appropriate processing of the TDBS signals, is another question. Qualitatively speaking, as the evaluation/fitting of the signal is commonly achieved in some moving time window of the duration τ_w , then the spatial resolution in the imaging of the BF parameter, $\omega_B(t)$, could be limited by the length $v_s \tau_w$ when $v_s \tau_w \geq l_a$ or $l_a \geq v_s \tau_w \geq l_f$. Some restrictions on the choice of the shortest τ_w and some other limitations of the TDBS imaging technique will be discussed later in Sec. V.

III. APPLICATIONS OF TIME-DOMAIN BRILLOUIN SCATTERING FOR NANOSCALE IMAGING OF INHOMOGENEOUS MEDIA

A. Monitoring the transmission of coherent nano-acoustic pulses across spatially localized inhomogeneity

The first experiments indicating potential of the TDBS for depth profiling of continuously spatially inhomogeneous materials were those where the transmission of acoustic

pulses through the interface between two different optically transparent homogeneous media was monitored.^{29–31,70–73} Insightful experiments were performed with SiO₂ films deposited on Si substrates^{29–31,70,74–77} as presented in the scheme given in Fig. 4(a).

The experiments³⁰ demonstrated remarkably large sensitivity of the transient optical reflectivity signals to the optical wavelength of femtosecond laser pulses applied to probe the CAPs in the samples. Figures 4(b) and 4(c) shows that, depending on the probe wavelength, transient reflectivity signals $dR(t)/R$ can reveal either only the return of the weak acoustic pulse reflected at the SiO₂/Si interface to the front surface of the sample [red probe detection of acoustic echo indicated by an arrow in Fig. 4(b)] or a much higher in amplitude BO signal corresponding to monitoring of the CAP first in the SiO₂ and then later in the Si [the case of blue probe in Fig. 4(c)]. It was revealed in Ref. 31 that TDBS signals strongly depend on the probe optical

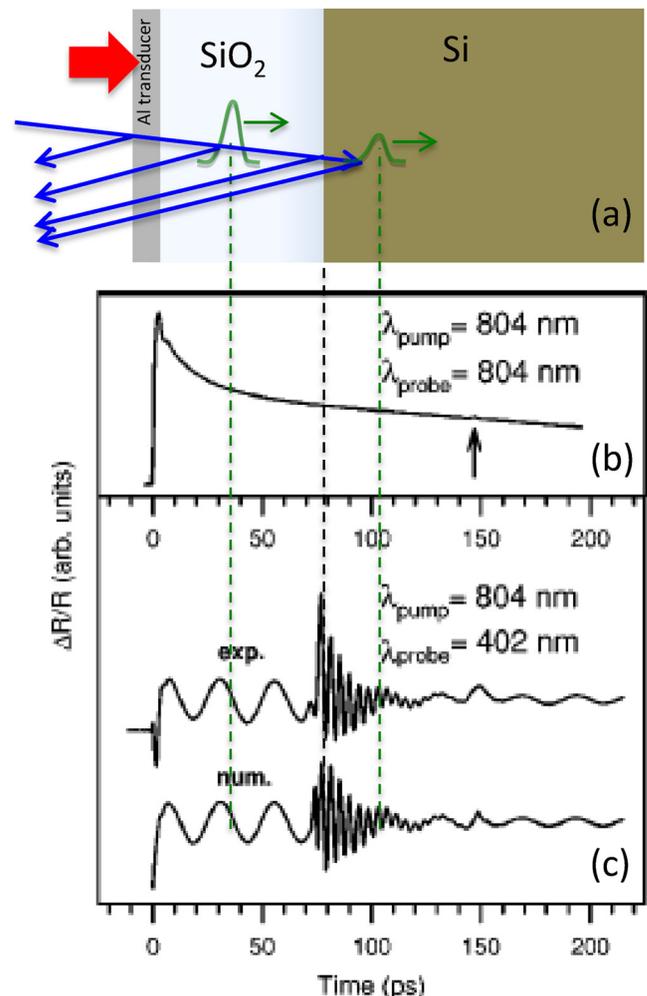


FIG. 4. The detection of a spatially localized inhomogeneity. (a) A metallic, semi-transparent transducer was used to generate the coherent acoustic pulses (adapted from Ref. 30). (b) Reflectivity measurement on an Al/SiO₂/Si sample with pump and probe both centered at 804 nm. (c) Reflectivity measurement when the probe is changed for 402 nm and numerical results which well reproduce all the observed oscillations. The Brillouin oscillation amplitude abruptly increases starting near 75 ps when the coherent acoustic pulse reaches the SiO₂/Si interface. Reproduced with permission from A. Devos *et al.*, Phys. Rev. B **70**, 125208 (2004). Copyright 2004 American Physical Society.

wavelength near the critical points of the silicon energy band structure where there are important variations not only in the optical parameters of this material but also, and most importantly, in the magnitude of photo-elastic parameter to which the constant C in Eq. (1) is proportional. Later, the TDBS experiments were realized with other combinations of transparent materials, such as various transparent layers deposited on silicon,^{31,70,74,78} GaSb-GaAs heterostructures,⁷² ZnO and SiO₂ deposited on GaAs,³⁵ the vacuole of a cell deposited on a thin polyethyleneimine film,⁷⁹ and even microcapsules composed of polymer shell encapsulating a liquid core.^{80,81} The TDBS with a variable probe wavelength were conducted in GaAs.^{72,73} In Ref. 82, the TDBS was applied to follow CAP propagation across a grain boundary. One of the most insightful reports⁷³ indicating the possibilities of TDBS for depth profiling showed how the doping profile in different n -doped GaAs homoepitaxial layered structures [see Fig. 5(a)] could be measured. The authors used an optical pump-probe technique called asynchronous optical sampling (ASOPS) introduced earlier by them in picosecond acoustic studies.^{83,84} This technique permitted detection of changes in optical reflectivity over a 1 ns time delay with a signal-to-

noise ratio of 10^{-7} and 100 fs time resolution in approximately 1 min of acquisition time. It was shown that the doping profile with doping densities of the order of 10^{18} cm^{-3} can be detected due to differences in the optical and acousto-optical parameters of the doped and non-doped regions. The experimentally recorded Brillouin oscillations (BOs) for the samples with the doped GaAs film on top of the substrate [Figs. 5(b) and 5(c)] clearly demonstrate a behavior substantially different from those shown in Figs. 4(b) and 4(c). This difference comes from the fact that for the two experiments the coherent acoustic fields generated by the pump laser pulses differ and have dissimilar spatio-temporal patterns. The shortest acoustic transients are generated in the regions of largest gradients of the photo-induced stresses,^{36,37} which are localized near the mechanically free surface of the sample and at the interface between the layer and substrate.^{73,85} Thus, the coherent acoustic fronts are launched at the surface in the direction of the interface and at the interface in two opposite directions. The BOs in Figs. 5(b) and 5(c) correspond to simultaneous monitoring by the TDBS of three coherent acoustic fronts. However, the BO monitoring still performs adequately for determination of interface localization and for comparing the parameters of interfacing materials. Of parenthesis note is that the research papers cited in this section point out the importance of optical probe color choice in the TDBS depth-profiling experiments.

B. Depth-profiling of ion-implanted semiconductors and dielectrics

The first application of TDBS for depth-profiling of the samples with continuously distributed in space inhomogeneity of acousto-optic parameter was reported in Ref. 42. The experiments were carried out using GaSb/GaAs heterostructures [inset in Fig. 6(a)]. Defect concentrations in GaAs specimens were created through He⁺ ion implantation over a wide range of doses, reliably creating damage profiles, which can be simulated using the transport of ions in matter (TRIM) code.⁸⁶

The experimental results illustrated in Figs. 1(e) and 6(a) demonstrate that the BO in both the undamaged and damaged samples exhibit amplitude attenuation as a result of natural attenuation of the probe light in bulk GaAs, the period, and phase remaining identical for both samples, with a distinct difference seen only in the BO amplitudes, which were found to be as much as 35%. The amplitude modulation in the damaged region can be wholly attributed to changes in the photo-elastic constants of GaAs.⁴² Note that the generalization of the theoretical formulas discussed in Sec. II for the case where the material is inhomogeneous not only acoustically, but also optically and acousto-optically can be found in Ref. 65. In order to develop a tool for quantitative characterization of the defect spatial profiles, the authors calibrated the TDBS measurements. It was shown that the raw data, such as those in Fig. 6(b), can be transformed into quantitative, depth-dependent defect concentration profiles [Fig. 6(c)]. It was also demonstrated that the TDBS may be used to monitor the decrease in defect concentration in the

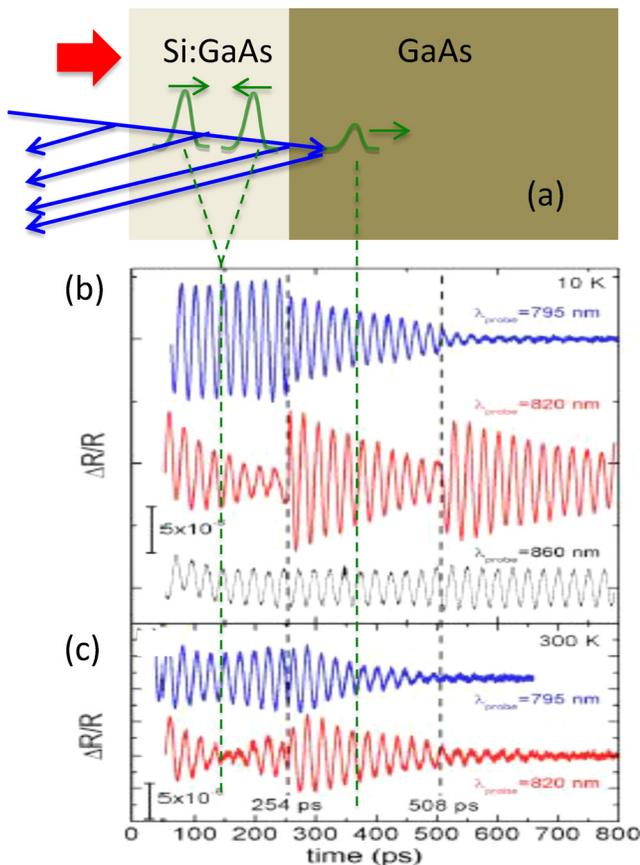


FIG. 5. The detection of a spatially localized inhomogeneity. (a) The sample consists of n -doped GaAs films on non-doped GaAs substrates (adapted from Ref. 73). $\Delta R/R$ results were obtained at 10 K (b) and 300 K (c). The measurements were performed with a pump wavelength of 800 nm and probe wavelengths of 795 and 820 nm. $R(t)$ transients exhibit a distinct phase shift and with most of them showing an amplitude change at 254 ps, the time needed by a coherent acoustic pulse to travel once through the film. Reproduced with permission from J. Appl. Phys. 104, 123509 (2008). Copyright 2008 AIP Publishing LLC.

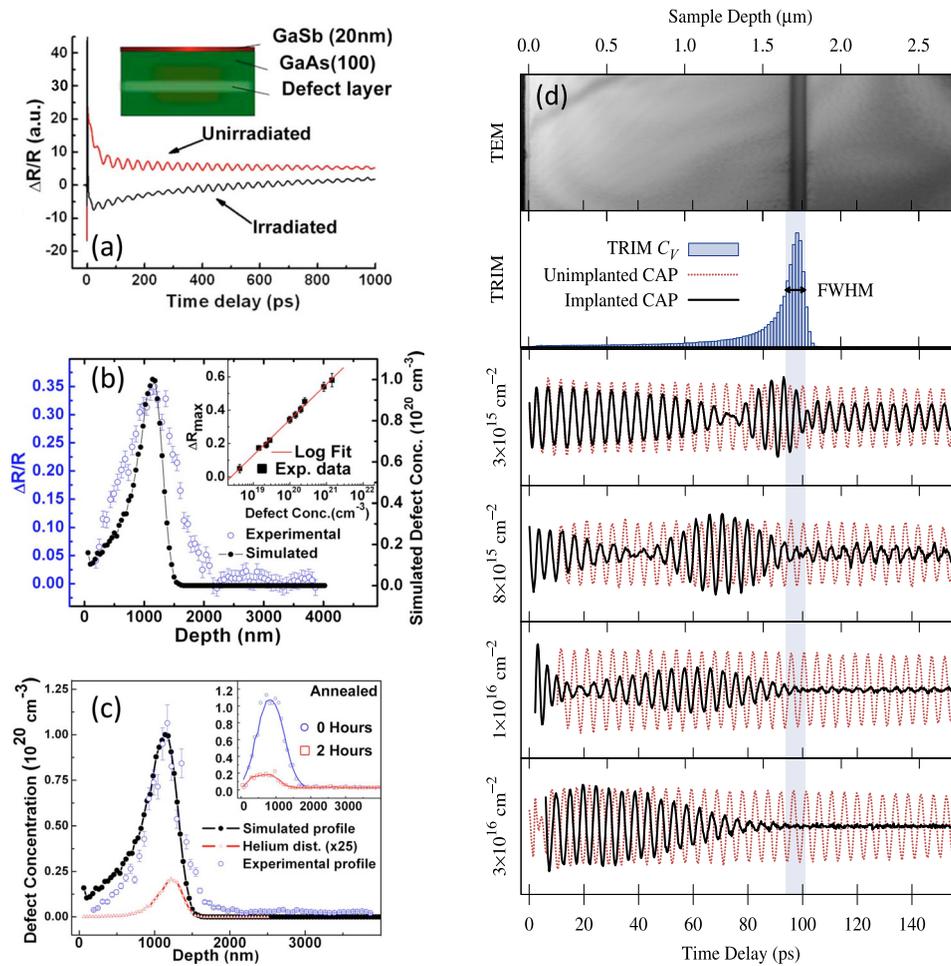


FIG. 6. First experiments on depth-profiling by time-domain Brillouin scattering of a continuously distributed in space acousto-optical inhomogeneity of material. (a) Time-resolved pump-probe optical response of ion-implanted GaAs for undamaged and damaged samples.⁴² The coherent acoustic pulses were generated following the interband absorption of pump laser pulses in GaSb layer. The minimum point in space in the amplitude of the Brillouin oscillation in the damaged sample corresponds quantitatively to the position maximum of the defect concentration evaluated by the TRIM numerical code. (b) Difference in oscillatory amplitudes between irradiated sample and undamaged samples, with comparison to the simulated peak defect concentration (solid black curve). Inset: Experimental dependence of peak amplitude modulation as predicted from TRIM calculations which can be used to transform the raw data in (a) into absolute defect concentration profiles. (c) Experimental defect concentration profile for a sample irradiated at 325 keV at a dosage of 7×10^{13} ions/cm². Observed peak defect concentration agrees well with simulated profiles for total damage (black) and helium ion distribution (red, enhanced 25 \times) from the TRIM code. Inset: Experimentally measured defect profiles before (blue) and after (red) 2 h of thermal annealing at 300 $^{\circ}$ C. Reproduced with permission from Appl. Phys. Lett. **94**, 111910 (2009). Copyright 2009 AIP Publishing LLC. (d) BO in the ion-implanted diamond specimens at multiple fluences (dark lines).⁴⁴ The dotted red line behind each curve is the corresponding response for an unimplanted specimen. Above the oscillating responses are the TEM cross-sections for the specimen dosed at 3×10^{16} cm⁻² [bright field, two-beam diffraction, $g = (220)$] and the damage-induced vacancy distribution as calculated by the TRIM code. The vertical bar indicates the full width at half maximum of the TRIM vacancy profile. The spatial extent of the implantation effects in the diamond specimens on the BO is much broader than seen in the visible structural changes in the lattice shown by TEM. The existence of the node in the amplitude of the BO around which the phase of the oscillations changes by 180 indicates that for threshold values of vacancy concentration the photo-elastic constant of the sample changes its sign passing through its zero value. Reproduced with permission from Appl. Phys. Lett. **101**, 181904 (2012). Copyright 2012 AIP Publishing LLC.

annealing process [inset Fig. 6(c)]. Note that the time delay of the pump/probe reflectivity signal can be correlated with crystal depth by simple multiplication of the delay time by the longitudinal speed of sound, since material is acoustically homogeneous. This simple procedure enables the features in the reflectivity signal to be mapped directly against the damage-induced vacancy concentration profile as calculated by TRIM [see Figs. 6(b) and 6(c)]. As predicted by simulation and verified by ion channeling analysis,⁴³ the reduction of the TDBS signal amplitude was directly related to the structural damage density.

TDBS with an optical probe frequency near the band-edge is also very sensitive to damage-induced modification of the

optical properties of diamond crystals.⁴⁴ The bottom four panels of Fig. 6(d), adapted from Ref. 44, show TDBS signals for diamond specimens irradiated with ion fluences ranging from 3×10^{15} to 3×10^{16} cm⁻². The regions of strongest ion-induced damage manifest themselves by the BO that are opposite in phase relative to those in the undamaged regions. The front boundary of the zone where the damage is easily distinguishable, close to the node in the amplitude of the BO in Fig. 6(d), approaches the sample surface with increasing implantation dose. Recently, TDBS has been applied for the determination of ion-implantation densities in SrTiO₃⁴⁵ and 4H-SiC.⁴⁶

The totality of experimental results and theoretical interpretations^{42–46} convincingly demonstrate that TDBS is a

sensitive, noninvasive, and nondestructive tool for studying the depth-dependence of optical properties induced by doping, ion-implantation, or radiation-induced damage. TDBS provides insight into the influence of defects on optical properties and into the specific relationships between the structural modifications of the crystal lattice and the resulting modulation of local optical properties of relevance to the fabrication of various photonic and optoelectronic devices. As reported in Refs. 42–44, “the technique spans at least four orders of magnitude in defect concentration and has a depth range of the order of tens of micrometers and $\times 10\text{--}30$ nm depth resolution throughout the probed range.” The sensitivity of TDBS makes it a robust defect detection technique with the demonstrated depth limits surpassing those achieved using conventional medium energy ion scattering (MEIS) and ion channeling measurements, which, notably, create damage as a result of analysis.

C. Depth-profiling of nanoporous thin films

The first application of TDBS for the depth-profiling of a continuously distributed elastic inhomogeneity of a sample was reported in Ref. 41. The samples were low dielectric constant, nanoporous organosilicate (SiCOH) thin films [Fig. 7(a)]. Because of their porosity, such materials are mechanically weak. Significant efforts are therefore being directed to improving their mechanical properties by post-deposition treatments. Ultraviolet (UV) radiation-assisted thermal curing, which combines photochemical processing with thermal activation, is perhaps the most promising treatment because it permits efficient removal of porogen, significantly improving mechanical properties with minimum damage to the low- k matrix. However, the intensity distribution of UV light exhibits a gradient inside the low- k film and may create a gradient in its mechanical properties. The first experiment⁴¹ using TDBS for probing a continuously distributed elastic inhomogeneity was conducted using a single-wavelength (≈ 800 nm) and an optical configuration with a single incident probe angle. The elastic inhomogeneity was extracted from the analysis of the variations in the BF by neglecting the inhomogeneity of the optical reflective index. A subwavelength spatial resolution of 40 nm was achieved by direct fitting the BO in the time domain, thus avoiding a windowed Fourier transform of the signal.

Experiments reported later²⁸ on the same samples were conducted in the so-called “red pump–blue probe” (≈ 880 nm– ≈ 440 nm) configuration as presented in Fig. 1(a). It was demonstrated²⁸ that by performing TDBS at a few angles θ of probe light incidence,^{87,88} it is straightforward to determine from the BF temporal variations, $\omega_B(t) = 2\pi\nu_B(t)$, both the profile of the refractive index $n(z)$ and the profile of the sound velocity $v_s(z)$ simultaneously. This opportunity is based in its essence on the dependence of the BF in non-collinear interactions of probe photons with the CAP in the backward BS configuration on the angle between the directions of the propagation of the coherent acoustic waves and the probe laser light, $\nu_B(t) = (1/\pi)v_s(z_a)k[n^2(z_a) - \sin^2(\theta)]^{1/2}$. Note that this theoretical formula, implemented in Ref. 28 for depth-profiling, is also the basis for the interpretation of several other TDBS

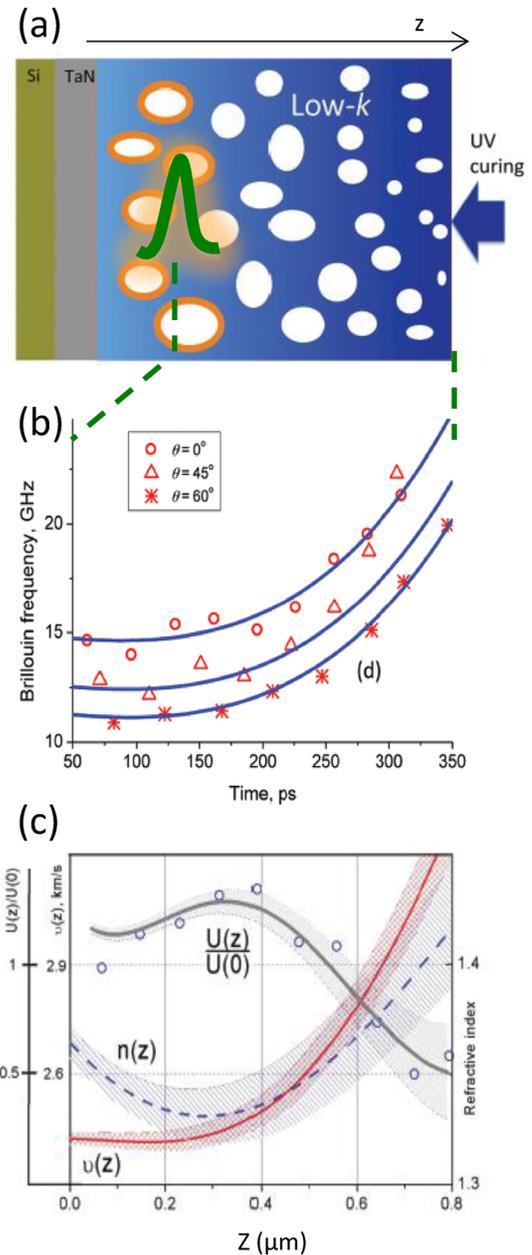


FIG. 7. First experiments on depth-profiling by TDBS of a continuously distributed in space elastic inhomogeneity of material. (a) Representation of the sub-micron inhomogeneities that exist within nanoporous transparent films. The white disks are nanopores. Their dimensions vary from the free surface toward the TaN - low- k interface. The blue matrix is an organo-siloxane matrix. The color gradient indicates that the deeper (lighter) blue is the region of higher (smaller) elastic modulus. Some nanopores still contain polymeric porogen (orange edges of the pores) that was not completely removed during nanomanufacturing. For optical generation of the CAPs, a TaN layer of 30 nm thickness was deposited on a Si substrate before deposition of the low- k material [Fig. 1(a)]. (b) Time-dependent BF of the transient reflectivity signal obtained at different probe beam incidence angles θ .²⁸ (c) Depth profiles of the refractive index, $n(z)$, the sound velocity $v_s(z)$, and the normalized photoelastic coefficient, $P(z)/P(0) \approx U(z)/U(0)$, obtained by the TDBS.²⁸ Reproduced with permission from A. M. Lomonosov *et al.*, ACS Nano 6, 1410 (2012). Copyright 2012 American Chemical Society.

experiments conducted in the homogeneous materials.^{87–91} In the inhomogeneous medium, it is valid within the framework of geometrical optics,^{67,68} while $z_a = z_a(t)$ is the current position of the CAP. As the CAP with initial magnitude $\eta(0)$ and duration τ propagates along the z -axis

from the transducer at $z=0$ [Figs. 1(a) and 7(a)], its strain magnitude and spatial scale vary (within the framework of the geometrical acoustic approximation^{67,68}) as $\eta(t) = \eta(0)[v_s(z_a)/v_s(z=0)]^{-3/2}[\rho(z_a)/\rho(0)]^{-1/2}$ and $v_s(z_a)\tau$, respectively, where $\rho(z)$ is the distribution of density. Moreover, because of the variations of the CAP amplitude, of its spatial extension, and of the photo-elastic constant $P(z_a)$ in depth, the BO amplitude is time-dependent: $A_B(t) \propto P(z_a)[n^2(z_a) - \sin^2(\theta)]^{-1/2}[v_s(z_a)\rho(z_a)]^{-1/2}$. Figure

7(b) gives $\nu_B(\theta, t)$ evaluated²⁸ at three different angles ($\theta = 0^\circ$, 45° , and 60°). The dependencies $v_s(t)$ and $n(t)$ extracted from $\nu_B(\theta, t)$ were transformed using the space-time correspondence, $z = z_a(t) = \int_0^t v_s(t')dt'$, into depth distributions of $v_s(z)$ and $n(z)$ [Fig. 7(c)] and also were used together with the time-dependent amplitudes $A_B(t)$ to examine variations in space of the parameter $U(z) \propto P(z)\rho(z)^{-1/2}$. Experimentally determined, non-monotonic variations of the optical refractive index n along the depth coordinate z of the nanoporous low- k film

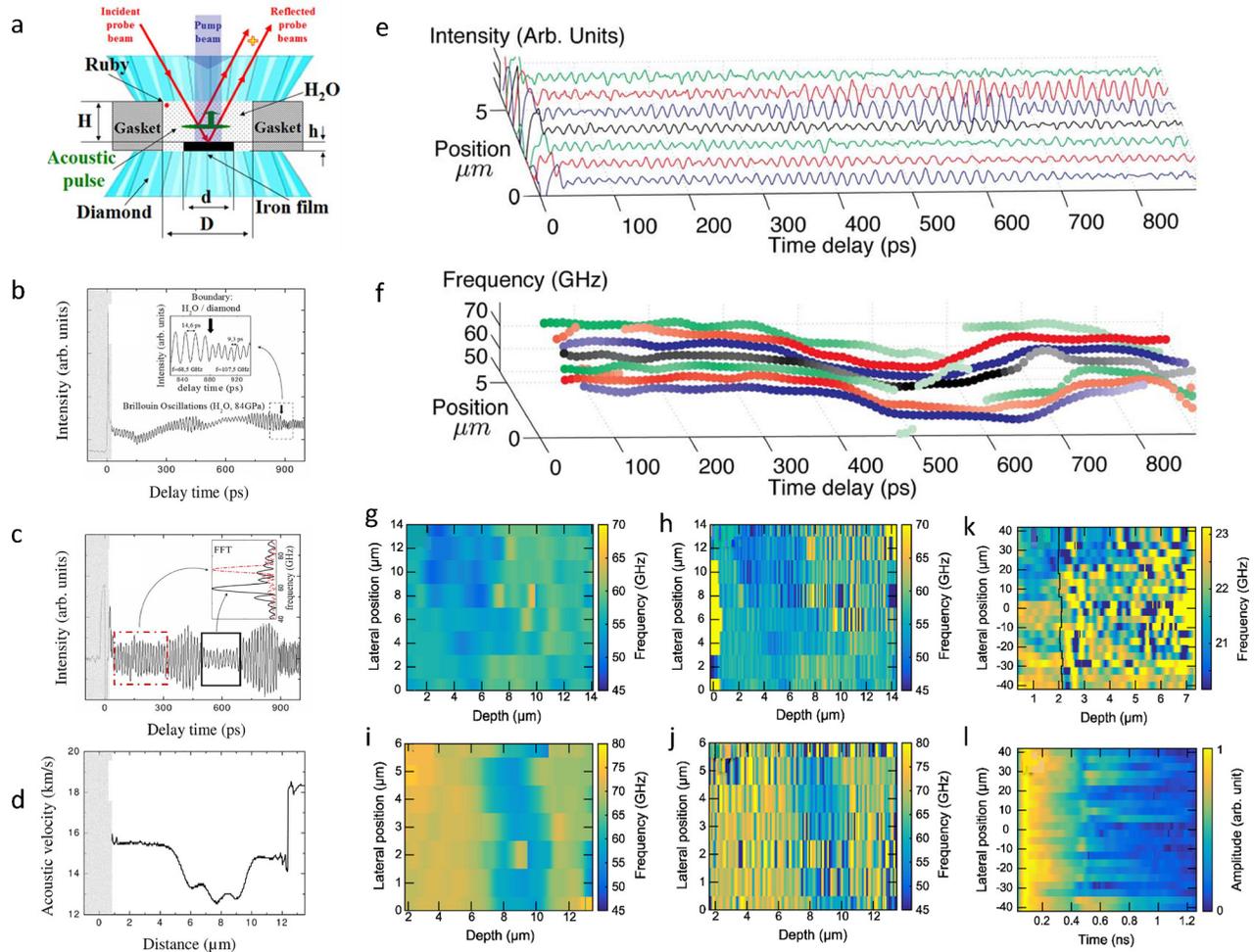


FIG. 8. Application of TDBS imaging in high pressure research. (a) Transparent, spatially inhomogeneous medium (polycrystalline H₂O ice⁴⁷ compressed in a diamond anvil cell. A picosecond pulse from the pump laser (violet) hits the absorbing transducer launching a coherent acoustic pulse (green) in the sample volume. Propagation of the pulse is monitored by a delayed probe laser pulse (red) reflected from stationary (transducer) and moving (acoustic pulse) interfaces. (b) and (c) Demonstration of the capability of TDBS for revealing spatial inhomogeneities in H₂O ice X at megabar pressures. (b) Typical time-resolved reflectivity signal in ice compressed in a DAC to 84 GPa. The vertical arrow marks the time of transmission of the laser-generated coherent acoustic pulse across the interface of ice with diamond. Inset: Zoom of the signal in the vicinity of the ice/diamond interface. (c) TDBS signal obtained by subtracting from the signal in (b) the time-varying thermo-reflectance contribution caused by transient heating of the sample. Inset: Fourier spectra of the signal inside two temporal windows, marked by rectangles showing the shift of the Brillouin frequency from 60 GHz to approximately 73 GHz, indicating spatial inhomogeneity in the ice sample. (d) Spatial variation of the longitudinal sound velocity obtained from the temporal dependence of the BF, as revealed by a Fourier transform performed in a rectangular moving temporal window of 100 ps duration, using extrapolated refractive index values of ice.⁹⁶ (e) and (f) Demonstration of μm -scale texture in H₂O ice aggregate at megabar pressures. (e) Intensity versus time delay signals in H₂O ice at 84 GPa, obtained by displacing the sample relative to the co-focused pump and probe laser beams in the lateral direction, i.e., parallel to the ice/diamond interface in 1 μm steps. (f) Two-dimensional images of the Brillouin frequency magnitude obtained by processing the signals in (e). Isometric representation of the TDBS signals and of the temporal dependence of the Brillouin frequency of ice X. In (f), the signal amplitude is correlated with the symbol color: the darker the symbol, the higher the maximum amplitude of the BO, and vice versa. Image (f) reveals a clear, large-scale, 3–5 μm , layering of the ice aggregate at 84 GPa in the direction normal to the diamond anvil culets. Reproduced with permission from S. M. Nikitin *et al.*, *Sci. Rep.* 5, 9352 (2015). Copyright 2015 Nature Publishing Group. (g)–(l) 2D-images of Brillouin frequency inhomogeneities in water ice compressed in a DAC to 57 GPa [(g) and (h)] and 84 GPa [(i) and (j)]. Absolute values of the frequencies can be derived from the color bars on the right hand side of each plot. The in-depth spatial resolution is about 1.05 μm in (g) and (i) and 0.26 μm in (h) and (j). (k) and (l) 2D-images of Brillouin frequency (k) and of the Brillouin oscillation amplitude (l) of solid argon compressed in a DAC to 15.4 GPa.⁴⁸ The in-depth spatial resolution is about 0.36 μm . In (k), the black line stands for delimiting the depth after which the influence of the second acoustic pulse on the images could be important. Reproduced with permission from M. Kuriakose *et al.*, *Ultrasonics* 69, 259 (2016). Copyright 2016 Elsevier.

was correlated²⁸ with its non-monotonic dependence on the UV radiation dose. The photoelastic coefficient $P(z)$ was estimated to be the dominant contributor to the variations in $U(z)$. Overall, the experimental results presented in Fig. 7(c) reveal a wealth of nanoscale subsurface information. An improved understanding of the depth-dependent UV curing effect is a key factor for successful integration of ultra-low- k layers in advanced microelectronic technology, while nondestructive depth-profiling of the mechanical, optical, and acousto-optical properties is essential for achieving this purpose.

D. Imaging of inhomogeneity and texture of polycrystalline materials at high pressures

TDBS was introduced for the first time in high pressure research in 2008.^{92,93} However, its applications to depth-profiling and two-dimensional imaging of a polycrystalline aggregate of ice compressed in a diamond anvil cell to megabar pressures were reported only quite recently⁴⁷ [Fig. 8(a)].

The knowledge of the pressure dependences of sound velocities and elastic moduli of liquids and solids at megabar pressures and the evolution of the texture of polycrystalline samples under compression is of extreme importance for a few branches of the natural sciences, such as condensed-matter physics, physics of the Earth and planetology, as well as for the monitoring and prediction of earthquakes and tsunamis, and for monitoring nuclear weapons tests. Experimental data on high-pressure materials parameters can be substantially influenced by spatial inhomogeneities and texture variations in polycrystalline aggregates. The aggregate sound velocity depends on the characteristic dimensions of individual crystallites, which are elastically anisotropic,⁹⁴ and even a partial alignment of the crystallites, i.e., orientational texture, which biases the measured velocities.⁹⁵ These factors prevent precise evaluation of elastic moduli and require the development of experimental methods for three-dimensional imaging of microscopic samples *in situ* when compressed in a diamond anvil cell (DAC) for the purpose of characterizing both their morphological and orientational/directional texture.

Figures 8(b)–8(d) illustrate how the inhomogeneity of H₂O ice was revealed in TDBS experiments.⁴⁷ Figures 8(e)–8(j) present the images of the spatial structuring of ice. In Figs. 8(g) and 8(h), the two-dimensional images of ice obtained with different spatial resolution reveal different scales in material inhomogeneity and texturing. In Fig. 8(k), the 2D image of Ar ice in DAC⁴⁸ is presented for comparison with those of H₂O ice revealing greater lateral than axial structuring in polycrystalline Ar. The most detailed updated information on the analysis of the TDBS imaging signals measured in ice^{47,48,97} can be found in Ref. 97. The synchronous detection technique (SDT) was applied in the temporal window equal to one Brillouin oscillation. The dependence of the accuracy in frequency determination on the signal-to-noise ratio (SNR) and the number of signal points inside the window was described. Two goodness criteria were applied in the signal analysis. The first one requires that the SNR exceeds 10 dB and the second one that the normalized root-mean-square error (NRMSE) remains below 10%. Under these criteria, the

frequency in the temporal window containing one Brillouin cycle was determined with the accuracy better than 6%.

The TDBS method allowed examination of the characteristic dimensions of ice texturing with sub-micrometer spatial resolution in the direction normal to the diamond anvil surfaces and with micrometer spatial resolution in the lateral directions. It is worth noting here that the axial spatial resolution in classic BS microscopy¹⁴ applied to three-dimensional imaging¹⁵ currently exceeds 2 μm . In chemically homogeneous transparent aggregates, TDBS imaging^{47,48} provides for each crystallite (or group of crystallites) usable information on its orientation (if the material is elastically anisotropic) as well as the elastic modulus along the direction of sound propagation. From the viewpoint of quantitative measurements, the TDBS technique is significantly superior to the classical frequency-domain BS (FDDBS) technique as a result of its far higher spatial resolution. For example, in Ref. 47, the values of the longitudinal sound velocity, $v_s(z)$, in H₂O ice were recorded with an axial resolution of $\sim 0.26 \mu\text{m}$ at 82 GPa, corresponding to the length of one BO. Thus, the TDBS signal collected at one given lateral position from a 15 μm thick sample contained up to 60 independent v_s measurements. In order to accumulate statistically reliable limits of variation of v_s lateral scans over distances up to $\sim 70 \mu\text{m}$ can be performed. This capability dramatically increases the statistical confidence levels of the recorded maximal and minimal velocities in cubic single crystals, $v_s^{(111)}$ and $v_s^{(100)}$, permitting reliable determination of pressure dependences of single crystal elastic moduli $c_{ij}(P)$ of ice at high pressures.⁹⁷

E. Subcellular imaging inside plant and animal cells

Owing to its demonstrated capability for determining materials properties and imaging spatially localized inhomogeneities in solids, TDBS was explored for probing liquids.^{98,99} After the first experiments in water⁹⁸ were reported, the interest shifted towards demonstration of the utility and biocompatibility of the technique for probing cells,^{79,81,100–103} the goal being development of a tool for imaging at a subcellular scale for biology and medicine. Different BFs were measured separately for the vacuole and nucleus *in vitro* in individual onion cells⁷⁹ suggesting that, in principle, there could be sufficient acoustic contrast within the cell for imaging. The estimated acoustic stress generated with TDBS is much lower than the adhesion stress of cells to their surroundings making the technique completely non-invasive.^{80,102}

Although the TDBS has been applied to the spatially resolved imaging of the cells only recently^{49–51} the method has already shown promise. In Ref. 49, TDBS allowed three-dimensional imaging (x, y, and t) of the CAP propagation with $\propto 1 \mu\text{m}$ lateral and $\propto 150 \text{ nm}$ depth resolutions *in vitro* animal cells [Fig. 9(a)]. The acquisition time for a single image ($\propto 30 \text{ s}$) is of the same order as that in super-resolution microscopy techniques such as photoactivated localization microscopy (PALM) and stochastic optical reconstruction microscopy (STORM).^{12,13} The BOs were wavelet transformed using the Morlet mother wavelet. The spatial depth resolution was limited to the order of the Brillouin wavelength. The frequency of the oscillations was measured with

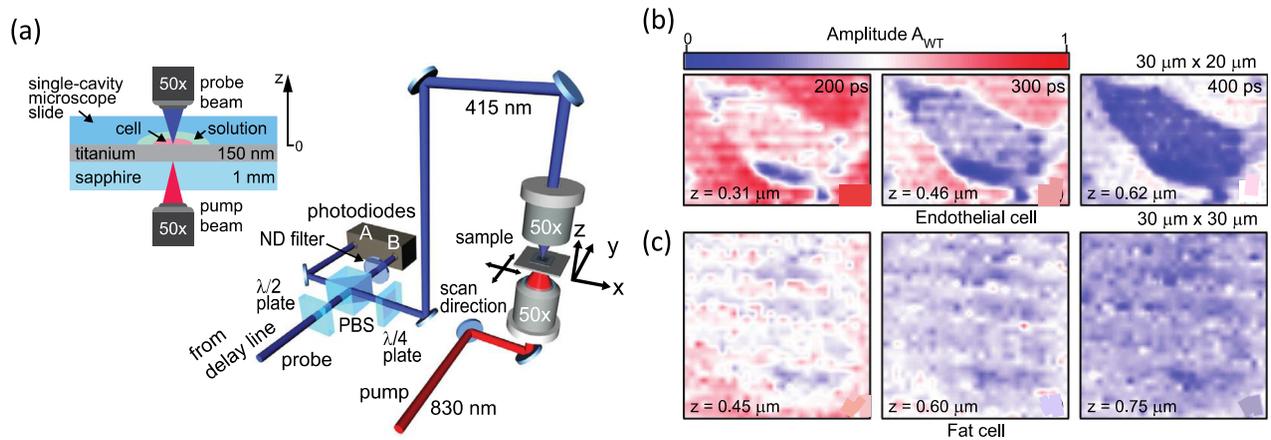


FIG. 9. TDBS imaging in cells in probe reflection configuration. (a) Experimental setup for cell imaging.⁴⁹ PBS: polarization beam splitter. ND filter: neutral density filter. Each (unstained) cell was covered by a single-cavity microscope slide filled with saline and buffer solutions for the endothelial and fat cells, respectively. Each slide is then placed on an x - y translation stage, allowing 2D raster scans. The probe beam is temporally scanned up to 800–1000 ps (after each ultrasonic pulse arrives at the Ti-solution interface) through use of a mechanical delay line. Spatial and temporal scanning allow 3D imaging (x , y , and t) over $40\ \mu\text{m} \times 40\ \mu\text{m}$ areas with $\approx 1\ \mu\text{m}$ and 1 ps resolutions. Two types of mammalian biological tissues—a bovine aortic endothelial cell and a mouse adipose cell, were investigated. Normalized wavelet transform amplitude images $A_{WT}(x, y)$ at the Brillouin frequency at different times: (b) at a frequency of 10.3 GHz for the endothelial cell at 200, 300, and 400 ps and (c) at 9.9 GHz for the fat cell at 300, 400, and 500 ps. The calculated average propagation distances z of the ultrasonic pulses within the cells at these times are also shown. Reproduced with permission from Appl. Phys. Lett. **106**, 163701 (2015). Copyright 2015 AIP Publishing.

the accuracy $\approx 1\%$. Spatially interpolated images of normalized wavelet transform amplitude (AWT) at three separate times are shown in Fig. 9(b) for an endothelial cell and in Fig. 9(c) for a fat cell. The spatial dependence of $A_{WT}(t)$ in Figs. 9(b) and 9(c) shows evidence of inhomogeneity inside the cells. The application of PAI allowed⁴⁹ determination of important physical parameters of cell samples including sound velocities, bulk moduli, and ultrasonic attenuation coefficients.

The TDBS technique described in Ref. 50 used a three-layered metal-dielectric-metal film¹⁰⁴ as a transducer to launch acoustic waves into the cell [Fig. 10(a)]. The design of this transducer and measuring system was optimized to

overcome the vulnerability of a cell to the exposure of laser light and heat without sacrificing the signal-to-noise ratio. The transducer substrate shields the cell from the laser radiation, efficiently generates acoustic waves, facilitates optical detection in transmission, and aids with heat dissipation away from the cell. The experiment⁵⁰ was based on a dual laser ASOPS system,^{83,84} where the sample was scanned by moving electromechanical stages with a minimum step motion of 100 nm. In a single scan, typically 15 000 averages per point were taken, which took 4 s for the acquisition. Figures 10(b)–10(c) show two fixed 3T3 fibroblast cells and the result of a 2D scan of their BO. The correlation between features in the optical image (b) and the TDBS map (c) is

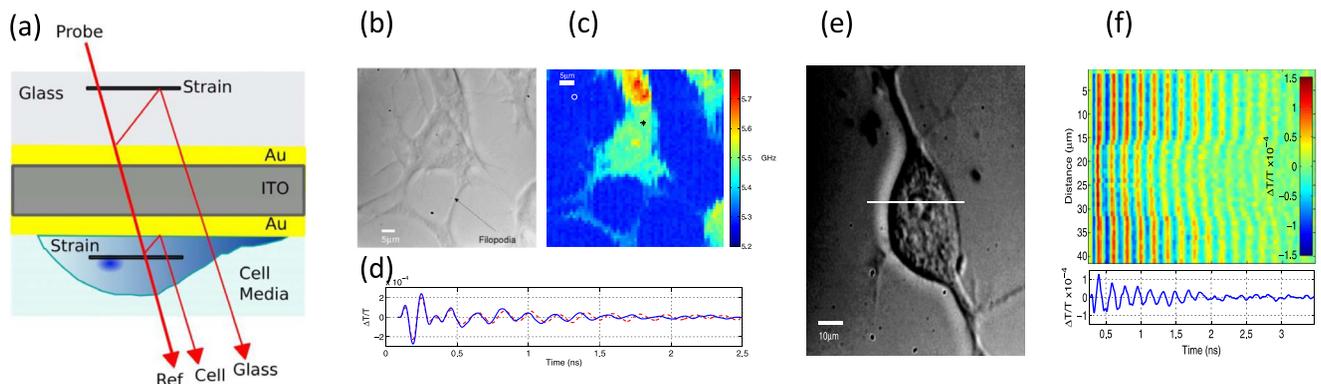


FIG. 10. TDBS imaging in cells in probe transmission configuration. (a) Schematic of the setup for operating in the transmission mode using a thin-film transducer.⁵⁰ Two signals are detected—one from the specimen (ref + cell) and one from the substrate (ref + glass). The dimensions of the transducer are exaggerated and beams are shown at an angle for clarity. 3T3 fibroblast cells were cultured on the transducers for 24 h using standard culture conditions prior to fixation in paraformaldehyde (4%) in phosphate buffered saline (PBS). The fixed cells were kept in PBS during the experiment, allowing a gentle flow that prevented air bubble formation. (b) Optical image of a 3T3 fixed cell, and (c) its TDBS map obtained using the transmittance approach. The scanned area was $60 \times 60\ \mu\text{m}$, and was recorded with one-micron steps. (d) A representative trace obtained from the cell (blue) and the medium (dashed red), revealing possible spatial inhomogeneities in the axial direction, which, however, were not analyzed.⁵⁰ The locations of these traces are indicated by a star and a circle, respectively. (e) and (f) TDBS scanning over living 3T3 cells. A microscope image of the cell before the scan is given in (e). A B-scan of the acquired cross-section with a sample time trace taken from the $21\ \mu\text{m}$ position is given in (f). The powers for pump and probe were 0.4 and 1 mW, respectively. The cell is exposed to 0.04 and 0.3 mW from the pump and probe beams owing to the transducer transmittance characteristics. The morphology of the cell after the scan remained structurally intact. Reproduced with permission from F. Pérez-Cota *et al.*, Appl. Opt. **54**, 8388 (2015). Copyright 2015 Optical Society of America.

good. It can be seen that the nuclei of both cells had clearly different Brillouin frequencies. This change is much less visible in the optical picture demonstrating the potential of acoustical contrast. Data acquisitions were also possible with living cultures of 3T3 cells as shown in Figs. 10(e) and 10(f).

The experiments^{49–51} reported to date suggest extension of TDBS techniques for high-resolution acoustic intercellular imaging which should prove to be invaluable for investigating the mechanical properties of cell organelles. The application of TDBS allows determination of important physical parameters including sound velocities, bulk moduli, and ultrasonic attenuation coefficients of animal^{49,100} and vegetal^{50,51,79,103} cells. Characterization of the physical properties of cells is essential for understanding mitosis, apoptosis, adhesion, and mobility.

IV. IMAGING OF NANO-ACOUSTIC WAVES TRANSFORMATION IN HOMOGENEOUS MEDIA

A. Attenuation of coherent hypersound

TDBS possesses a capability for following the transformation of CAPs during their propagation in optical transparent media as can be seen from examination of the basic formula for acoustically induced transient reflectivity variations given in Eq. (1), where spatio-temporal distribution of the strain field enters under the integral. A well-known example of acoustic wave transformation is evaluation of coherent sound attenuation from the temporal decay of the Brillouin oscillation amplitude.^{23,25,92,98} We cite here just some of the measurements of sound absorption in glasses,²⁵ solids,⁹² polymers,^{80,105} glass-forming liquids,¹⁰⁶ liquids,^{98,107} and biological cells.^{49,79} It is important to note that the experimental results presented in Ref. 80 indicated deviation in the decay of the detected BO from an exponential one. The deviation was increasing with increasing propagation distance of laser-generated CAP from the emitting opto-acoustic transducer. The authors hypothesized that the revealed attenuation of the BO, additional to acoustic absorption, was caused by the diffraction of the coherent acoustic waves as it had been expected theoretically.²⁵ This hypothesis was supported by the estimates of the characteristic diffraction length of the coherent acoustic wave at BF and is very plausible. The additional attenuation of the TDBS signal caused by the diffraction of the CAP was also reported later.¹⁰⁸ However, no attempts to image the CAP diffraction process by TDBS, via quantitative fitting of the BO attenuation, have been undertaken.

B. Nano-focusing of coherent hypersound

A very recent example is the application of TDBS for monitoring of the focusing of coherent nanoacoustic pulses.⁶³ Infrared pump light pulses were used to generate CAPs at the surface of a sub-micron, single metal-coated silica fibre [Fig. 11(a)]. Partially transmitted probe light pulses allowed the CAPs to be continuously monitored revealing acoustic focusing in the time domain as a result of the curvature of the fibre surface. An analytical model, supported by three-dimensional simulations, suggested that focusing of the 44-GHz BF of the acoustic beam was possible down to a

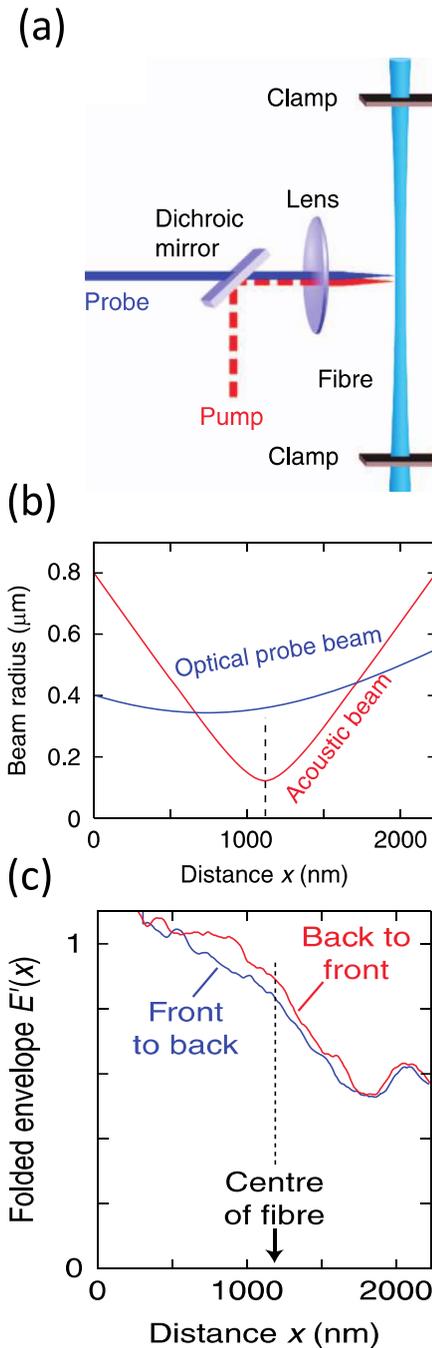


FIG. 11. Optical tracking of picosecond coherent phonon pulse focusing inside a sub-micron object.⁶³ (a) Experimental sample and setup. (b) Theoretical optical probe (blue) and acoustic-strain (red) $1/e^2$ beam radii as a function of propagation distance x (over the region from the front surface of the fibre to the back) as determined from Gaussian-beam theory for a fibre of radius $R = 1150$ nm. (The dashed line indicates the position of the centre of the fibre.) (c) Experimental results for the BO envelope as a function of the propagation distance x . Reproduced with permission from T. Dehoux *et al.*, Light: Sci. Appl. 5, e16082 (2016). Copyright 2016 Nature Publishing Group.

~150-nm diameter waist [Fig. 11(b)]. The experimental results, shown in Fig. 11(c), are in good agreement with the theoretical predictions.

The work with fibre generated CAPs demonstrates the high lateral resolution of TDBS imaging possible with focused picosecond CAPs, which, otherwise, is normally

limited by the diffraction limit of the pump optical pulses to $\sim 1 \mu\text{m}$. In particular, until now TDBS imaging has not been able to overcome the optical diffraction limit for lateral resolution. As suggested in Ref. 63, with the use of metal-coated cylindrical or spherical sub-micron hollowed regions on the end of an immersed tapered fibre in a standard near-field scanning optical microscope geometry, TDBS could follow focused GHz acoustic phonon pulses in liquids, extending the resolution of 3D acoustic imaging^{49–51} of biological structures to the nanoscale. In addition to focusing of CAPs^{63,109} other possibilities for improving the lateral resolution of TDBS imaging include photo-generation of CAPs significantly narrower than the pump laser beam dimensions as a result of nonlinear processes in opto-acoustic transformation¹¹⁰ and application of near-field optical schemes.^{111,112}

C. Nonlinear transformation of coherent nano-acoustic pulses

Variation of the CAP spatio-temporal distribution in homogeneous media can also be caused by nonlinear acoustic phenomena, which are known to lead to the formation of weak shock fronts in non-dispersive media.¹¹³ Frequency mixing processes arising from the elastic nonlinearity of the material modify the spectrum of the CAP, its shape, and length. The nonlinear transformation of bipolar picosecond CAPs had been first observed by classical frequency-domain BS¹¹⁴ through measurement of the distribution of 22 GHz coherent longitudinal phonons along the CAP trajectory via a step-wise increase in distance between the opto-acoustic transducer and a continuous probe light focusing region. The resolution in the propagation direction, on the order of $4 \mu\text{m}$, was determined by the waist of the focused probe laser. This spatial resolution is 1–2 orders of magnitude lower than that achievable by TDBS. Reference 61 describes experiments where nonlinear bipolar CAPs propagating in a SrTiO₃ (STO) substrate were monitored by TDBS over a range of optical wavelengths from 700 nm to 470 nm. Beats in the amplitude of the detected BO were observed at high laser fluences [Fig. 12(a)].

The theory^{61,69} indicates that the minima in the BS spectrum at a certain time delay at particular frequencies [Fig. 12(b) right panel] can be interpreted as mutual compensation of the probe light fields scattered by leading and trailing fronts of the bipolar CAP. Correspondingly, the observed beats [Fig. 12(a)] are a consequence of the constructive and destructive interference of the probe light scattered by two acoustic fronts moving relative to each other.^{61,69} Monitoring of the nonlinear evolution of the unipolar CAPs by TDBS was achieved in Ref. 62. A unipolar CAP theoretically^{69,113} transforms in the time-domain, as indicated in the upper inset of Fig. 13(a), to a triangular form with a weak shock front and a trailing edge of far longer duration. In the frequency domain nonlinear phenomenon of frequency-mixing leads to continuous generation and absorption of phonons at the BF accompanied by the phase shift. In the time-domain, continuous generation and absorption processes result in the BF predominantly localized at a weak

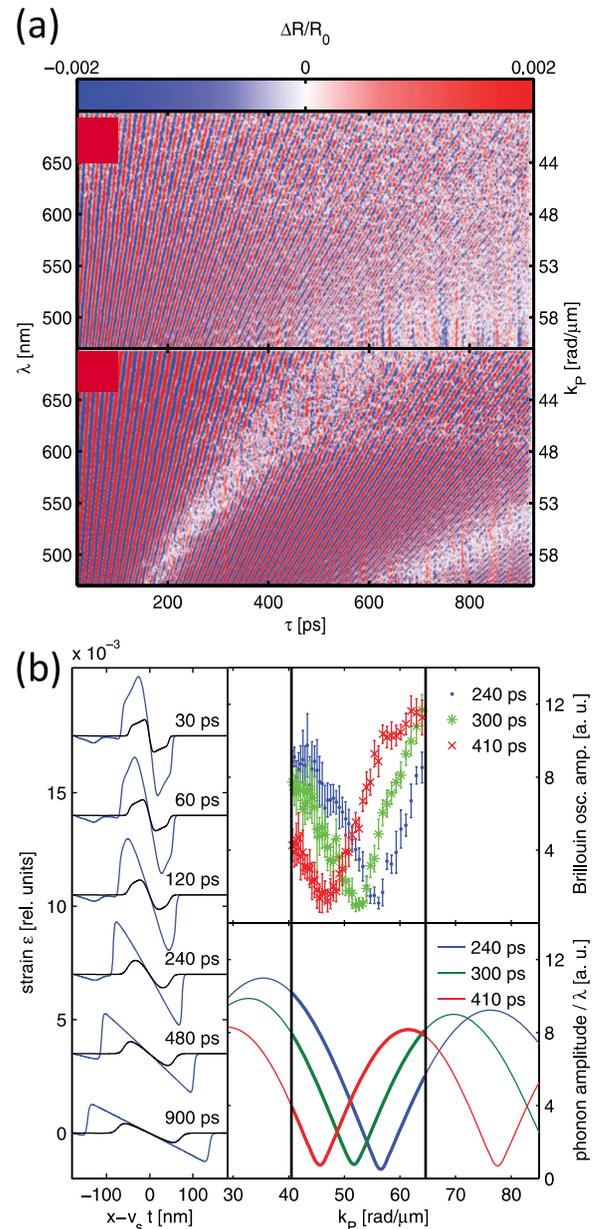


FIG. 12. Nonlinear transformation of a bipolar coherent acoustic pulse as followed by time-domain Brillouin scattering.⁶¹ (a) Relative optical reflectivity change of the sample as a function of delay time for a pump fluence of 14 mJ/cm^2 (upper panel) and 47 mJ/cm^2 (lower panel). The low-frequency background was subtracted from the image by high pass filtering. The ordinate gives the probe pulse wavelength λ (left axis) and the wavenumber of a phonon k_p , causing Brillouin scattering (right axis). For strong excitation conditions (lower panel), a beating was observed in the TDBS signals. (b) (left panel) Spatial profile of the bipolar strain pulse in the STO for different propagation times in a frame of reference propagating with the speed of sound v_s for high amplitude (blue line, 0.47% strain) and low amplitude (black line, 0.14% strain). Upper right panel: Measured amplitude of the Brillouin oscillation for each k_p . The region between the vertical black lines indicates the wave vectors that can be accessed by the optical white light. Lower right panel: Phonon amplitude divided by the wavelength λ as a function of wave vector calculated from Fourier transforms of the simulated strain profile. The data show good agreement with the measurement in upper panel. Reproduced with permission from A. Bojhr *et al.*, Phys. Rev. B **86**, 144306 (2012). Copyright 2012 American Physical Society.

shock front. Consequently, the velocity of weak shock front detected by TDBS matches the velocity of BF component.⁶⁹ Thus, the difference $\partial\varphi_{\eta}/\partial t$ between the cyclic frequency detected by TDBS $\omega_{\text{DR/R}}$ and the linear BF ω_B Eq. (6), is due

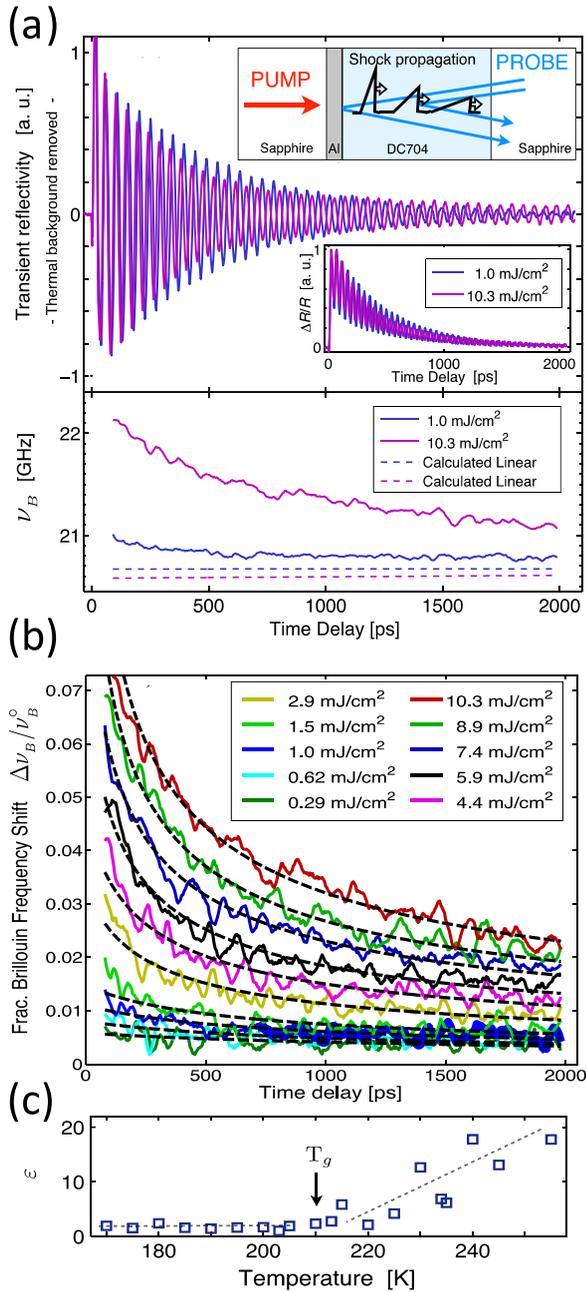


FIG. 13. Nonlinear transformation of a unipolar coherent acoustic pulse (CAP) as followed by time-domain Brillouin scattering (TDBS).⁶² (a) Upper panel: Normalized TDBS data recorded at 200 K in silicone oil (DC704) at two representative low and high pump fluences with the thermoreflectance background removed.⁶² Inset (top): Experimental setup with the $\sim 100 \mu\text{m}$ liquid squeezed between two sapphire substrates, one holding a 33 nm aluminum photoacoustic transducer film. A propagating nonlinear unipolar CAP is depicted as well. Inset (bottom): Diagram of the nonoscillatory signal component arising from thermally induced changes in reflectivity of the Al film. Lower panel: Extracted values for the time variation of the experimentally determined Brillouin frequency. The frequency-down chirp with increasing time at high fluence is caused by nonlinear acoustic propagation of a weak shock. The dashed lines show the calculated up-chirp due to sample heating at 1.0 and 10.3 mJ/cm² pump fluence (top and bottom lines, respectively). (b) Measured results of the fractional Brillouin frequency shift $(\nu_{dR/R} - \nu_B)/\nu_B$ in DC704 for different laser pump fluences. Dashed lines are theoretical fits based on an analytical solution for nonlinear transformation of a triangular CAP into a weak shock. (c) The dependence of the nonlinear acoustic parameter of the fragile glass former DC704 at around 20 GHz at temperatures extending through the glass transition. Reproduced with permission from C. Klieber *et al.*, Phys. Rev. Lett. **114**, 065701 (2015). Copyright 2015 American Physical Society.

to the additional nonlinear transport of the BF phonons and other high frequency phonons contributing to weak shock front. In other words, the experiments⁶² and the theory⁶⁵ demonstrate that the time-dependent phase of the transient reflectivity signal can be predominantly determined by the position of the weak shock front. The shock speed, and, as a consequence $\nu_{dR/R}$ increase with CAP amplitude, which increases with laser fluence and diminishes with the propagation distance, as was observed experimentally in Ref. 62 [Figs. 13(a) and 13(b)] in accordance with theoretical expectations.⁶⁹ Non-exponential reduction of the frequency shift $\nu_{dR/R} - \nu_B$ with time shown in Fig. 13(b) for high excitation fluences has also an origin in nonlinear acoustics.^{62,69} It is worth noting here that in the experiments⁶² the duration of the acoustic pulse initially launched in the medium was approximately ten times shorter than the period of the Brillouin oscillation, T_B , but could become comparable to T_B at the largest imaged depths for the highest pump laser fluencies. At the same time for the highest laser fluencies, the width of the shock front, which is controlled by the competition of the nonlinearity and dissipation, is shorter than the initial duration of the acoustic pulse even at the end of the experimental time window, i.e., at largest depths. The nonlinear dissipation of acoustic energy^{69,113} takes place in the weak shock front and the points of the shock front profile are the only points of the CAP profile, which are moving at time-dependent velocity. Thus, revealing by the TDBS of the time-dependent $\omega_{dR/R}$ ⁶² provides an experimental evidence of the TDBS sensitivity to the processes taking place in the shock front and of the TDBS imaging with spatial resolution much higher than one limited by the Brillouin wavelength, λ_B . TDBS enabled⁶² determination of the acoustic nonlinearity parameter¹¹³ to which the wave-amplitude-dependent quantity $\partial\phi_{\eta}/\partial t$ is proportional⁶⁹ [Fig. 13(c)]. A material-specific nonlinear acoustic parameter in a solid characterizes the anharmonicity of the intermolecular interaction potentials; in a liquid the same parameter should reflect an additional nonlinearity arising from structural relaxation. The experiments reported in Refs. 61 and 62 demonstrate convincingly that TDBS opens the door to versatile measurements of gigahertz nonlinear acoustic phenomena.

V. PERSPECTIVES AND CONCLUSIONS

It is evident from the extensive experimental advances seen to date that the possibilities for application of TDBS to nanoscale imaging are numerous. There exist some limitations and drawbacks of the TDBS scattering technique to imaging and some prospects in enhancing its functionalities. As it has been explained in Sec. II, the spatial depth resolution of the TDBS imaging can be significantly better than one limited by the wavelength of the acoustic phonon at the BF, λ_B . In all currently known TDBS experiments, the duration of the laser pulses and the time interval Δt between neighbor points in the signal are shorter than the duration of CAP, so the length of the CAP or of its sharp front localization are expected to be the limiting factors in the spatial resolution. However, the spatial resolution was limited by the duration of the moving window, τ_w , in which the theoretical

signal was fitted to the experimental one, and in most of the experiments the window, τ_w , was not narrower than the Brillouin oscillation period, $T_B = 2\pi/\omega_B$. Thus, in most of the TDBS imaging experiments, the spatial resolution was not better than λ_B . There could be several possible reasons for choosing $\tau_w \geq T_B$. First, in this large window a quasi-sinusoidal character of the signal is well determined and the well-developed/known approaches based on the Fourier and wavelet transforms could be applied. Second, in the case of the probe laser radiation at wavelength $\lambda_p \propto 400$ nm, the Brillouin wavelength, λ_B , is around 100 nm, in the case of large optical refractive index, and this spatial resolution could be sufficient for the experimental goals. Third, the shortening of τ_w additionally 4–5 times to a typical duration of the CAP could require improvement/modification of the experimental setup and would require application of less common/known methods for signal fitting. In the inhomogeneous medium, in general, both the amplitude and the phase of the TDBS signal are varying and each of them has to be approximated by a polynomial, for example, increasing the number of the fitting parameters. Then, the minimum duration of τ_w , which could be achieved, in general, depends on the number $N \approx \tau_w/\Delta t$ of the experimental points in the moving window, on the signal-to-noise ratio (SNR) and on a desired accuracy in the determination of the parameters. The larger is SNR and N the smaller could be uncertainty in the evaluation of the parameters. Shortening of τ_w without lose in accuracy could require increase in N through the diminishing of Δt and the increase in SNR. Both could be achieved by increasing the experimental time, if it is not limited by the stability of the laser or of the sample. When limited by the available experimental time, the SNR could be still increased by longer acquisition time in each experimental point and Δt could be diminished conserving the total duration of the experiment but diminishing the imaged depth of the sample.

It is also obvious that to achieve the ultimate spatial resolution it would be more and more complicated if the number of CAPs propagating simultaneously in an optically transparent sample increases, because of the necessary increase in the number of the fitting parameters. For example, this situation could take place when thin film optoacoustic transducer (OAT) in Figs. 1(a) is not well matched acoustically to the substrate or the tested inhomogeneous medium and emits a sequence of CAPs comparable in amplitude and periodically separated in time by the interval τ_d equal to the time required for the CAP to cross the film twice.^{25,48,69} If it is impossible to avoid this type of the OAT ringing, then it could be worth trying to accomplish first the extraction of the distributions of material parameters from the data accumulated in the time window $0 \leq t \leq \tau_d$ and then to use the obtained results for theoretical prediction of the contribution to TDBS signal from the second CAP when $\tau_d \leq t \leq 2\tau_d$ and its deletion from the total TDBS signal. Thus, the imaging by a single first CAP could be extended to the interval $\tau_d \leq t \leq 2\tau_d$, and so on. However, to keep the necessary level of accuracy in determining the material parameters could require broadening of τ_w to compensate the uncertainties accumulating at each next time interval.

Similar approach could be suggested if a single CAP initially travelling in a sample composed of several inhomogeneous layers of different transparent materials splits at the interface between the layers into two CAPs (reflected and transmitted) providing comparable contributions to the TDBS. However, in the latter case, if the BFs in the two simultaneously probed layers are sufficiently different, it could be more profitable to fit the TDBS signal from two CAPs simultaneously, by extending the theoretical formulas and increasing the number of the fitting parameters. In any case, it is obvious that for achieving the higher spatial depth resolution it is highly desirable to realize in the TDBS experiments the conditions for imaging by a single CAP at a time.

The limitation on the maximum depth of TDBS imaging could be caused by the attenuation of the CAP and/or of the probe light, and by decoherence of the reflected beams.²⁵ The probe light pulses scattered by the CAP and the sample interfaces should overlap in time in order to interfere at the photodetector. For example, in the simplest situation depicted in Fig. 3, if TDBS signal does not disappear at shorter distances due to limited penetration depths of CAPs and/or probe light, it will disappear at depths of CAP penetration exceeding half of the probe laser pulse length in the medium. For typical laser pulses of 200 fs duration, these depths are $\geq 10 \mu\text{m}$ for the refractive indexes $n \leq 3$. This limitation is rather technical than physical. If required, it could be lifted off by using longer laser pulses. However, if the durations of the probe and pump laser pulses are mutually related and comparable, then possible elongation of the laser pulses is itself limited, because it could cause elongation of the CAP.^{36,37} Another way to avoid the considered limitation could be splitting of a part of the probe beam before its incidence on the sample, delaying this part in time and using it for the heterodyning of the probe light scattered by the CAP.

From the technical point of view, new developments in TDBS imaging can be expected due to possible acceleration in signal acquisition, which should provide faster access to larger amounts of data than is presently possible (e.g., to a larger number of scans and more pixels per image), improvement in the signal-to-noise ratio through increased averaging at a particular measurement point, and a decrease in maximum thermal and optical loadings of the samples. The ASOPs technique^{73,83,84} has already been applied for TDBS imaging,^{50,51} while multichannel¹¹⁵ and wide-field¹¹⁶ systems have not yet seen experimental application. Also from the technical point of view, samples for TDBS imaging could be prepared in such a way that most of the spurious signals degrading the TDBS signal, such as contributions to transient reflectivity caused by photo-excitation of the optoacoustic transducer, are suppressed. For example, it would be advantageous to have spatial separation between the regions where the CAP's are generated and the points where the inhomogeneities lie, as was done in some earlier picosecond ultrasonics experiments.^{117,118}

From the technical-physical point of view, the prospects are related to the application in TDBS imaging of ultrafast time-resolved optical interferometry^{119,120} and optical polarimetry/ellipsometry.^{33,121–127} Theory⁶⁵ shows the advantages of depth profiling that could be gained if measurements of

both the real and the imaginary parts of the transient reflectivity $dr(t)/r$ are determined by optical interferometry. In the ideal case, ultrafast interferometry presents the opportunity to separate exactly the slowly varying amplitude from the equally slowly varying frequency of the BO. It is worth noting here that the separate measurements of the phase and of the amplitude of the TDBS signal by optical interferometry will require corresponding modifications of the current methods applied to TDBS signal processing. For example, all the methods exploiting somehow the Fourier transform become senseless, because neither amplitude nor phase is periodic in time. The explicit theoretical solutions for local BF as a function on time derivative of the measured phase of the TDBS signal could be applied for $\omega_B(t)$ extraction.⁶⁵ Thus, the achievement of the ultimate spatial resolution would be related to the minimization of the temporal window in which the phase (or phase derivative over time) could be adequately fitted to provide the required accuracy in the determination of $\omega_B(t)$ when substituted in the analytical formulas. However, in most of the experiments, this approach for absolute measurements of $\omega_B(t)$ requires additional independent measurement of some optical parameters of the samples and dedicated reduction/elimination of non-acoustic contributions to the TDBS signals.⁶⁵ For some cases, where the non-acoustic components masking the “wanted” TDBS signal are present, theory proposes how multiple measurements at different angles and/or with different probe polarizations could be applied in order to extract the wanted variations of the amplitude and frequency. These prospects are not simply technical because the application in TDBS of the polarimetry, for example, provides access to imaging of the material parameters that are not accessible by common TDBS technique, such as optical refractive index measurements for ordinary and extraordinary light waves and determination of the different components of the photoelastic tensor, including those controlling acousto-optic mode-conversion of probe light.¹²⁶ By conducting TDBS at several polarizations of the probe light it has already been possible to reveal spatially inhomogeneous optical anisotropy induced by loading of the material in a DAC⁴⁷ and to characterize spatial distributions of different photoelastic parameters of GaAs implanted by energetic protons.¹²⁷ Significant physical-technical progress also could be expected through the application of shear CAPs for TDBS imaging. Although optoacoustic transducers for shear CAPs are more elaborate than those used for compression/dilatation waves^{33,121–133} and detection of shear CAPs as well,^{33,122,133} their application would provide access to imaging of the complex shear rigidity of inhomogeneous media at nanoscale.

From the physics point of view, applications of TDBS imaging for deeper understanding of physical phenomena present themselves not only in the systems reviewed above but also in other materials, media, and structures that are spatially inhomogeneous at nanoscale. TDBS could be applied, for instance, to investigation of the physics of inhomogeneity of various transparent films, not simply those where the inhomogeneity is caused by post-deposition processing. The variation in the film texture with the increasing film thickness is considered a common phenomenon for film growth, in

general.¹³⁴ An inhomogeneity can also be introduced intentionally for the production of multilayered optical antireflective and highly reflective coatings.¹³⁵ Another possibility for the extension of physical applications of the TDBS technique is related to depth-profiling of the stress distribution near the surface of a crystal.¹³⁶ Knowledge of this distribution is not only of fundamental importance for surface and interfaces science but is also essential in microelectronics fabrication technologies. Stress in thin films in large scale integrated circuits can result in delamination of the film from the substrate, and cracking or buckling of the film itself. There can also be significant stress in the substrate as a result of the manufacturing process. Stress can come from the temperature gradients generated during solidification or as a result of deposition of structures on the surface. Thus, TDBS imaging of stress distributions in space could become an important technique for noncontact and nondestructive testing and control in micro and nanotechnology. Finally, the proven ability of the TDBS to monitor subsurface materials modifications in time,^{137,138} opens the path for the development of 3D spatial imaging of transient processes at nanoscale.

ACKNOWLEDGMENTS

V.E.G. acknowledges the support from the Alexander von Humboldt Foundation, JSPS, IUF, and ANR grant LUDACism. The authors thank A. Lomonosov, O. B. Wright, T. Dekorsy, C.-K. Sun, A. V. Akimov, A. J. Kent, A. Zerr, C. Mechri, G. Vaudel, N. Chigarev, S. Raetz, A. Bulou, V. Tournat, V. Juvé, and V. Temnov for collaborations and discussions. The authors thank G. J. Diebold and J. Thoen for critical reading and editing of the manuscript.

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