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SCANNING BRILLOUIN MICROSCOPY: ACOUSTIC MICROSCOPY AT GIGAHERTZ FREQUENCIES

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Abstract: Starting from acoustic microscopy, which bases on ultrasonic techniques, this document outlines the development of an alternative method, partially surpassing the classical acoustic microscopy: the scanning BRILLOUIN microscopy. Since scanning BRILLOUIN microscopy is an optical technique, it provides an entirely non-destructive access to the hypersonic velocity and hypersonic attenuation within transparent or translucent bulk samples under linear-response conditions. It is a striking feature of this technique that absolute values of hypersonic properties can be investigated. One-, two- or three-dimensional profiles of the acoustic or elastic inhomogeneities or heterogeneities can be generated for transparent materials. The properties of all acoustic modes can be recorded simultaneously, provided that the opto-acoustic coupling is sufficient. For non-transparent materials the acoustic properties of the surface, like surface acoustic waves, can be scanned. As BRILLOUIN spectroscopy is an optical technique, the resolution of maps of the hypersonic properties is given by the optical diffraction limit, so that a lateral resolution of about 1 micron can be achieved. Hence, scanning BRILLOUIN microscopy allows for combining the spatial, temporal and angular resolution of hypersonic properties. In other words, the complete information about the space- and time-dependence of the full elastic tensor can be resolved in transparent condensed matter.

Several examples from the domains of soft and hard matter physics illustrate the worth of this experimental technique: the resolution of transcrystalline domains within a semi-crystalline polymer, the spatial analysis of mechanical interphases of epoxy-based adhesives deposited on different metal substrates, and the spatio-temporal characterisation of solvent uptake of glassy polymeric networks. The determination of the angle dependence of the three acoustic modes, in addition to their space dependence, delivers valuable insight into the spatial evolution of the elastic tensor of anisotropic inhomogeneous or heterogeneous matter. This will be demonstrated for uniaxially stretched polymer specimens and for concentration profiles of mixed crystals. Using polycrystalline diamond plates, it will be evidenced that scanning BRILLOUIN microscopy provides a quantitative access to the spatial distribution of internal stress fields. It is the extremely low level of optical and acoustic perturbation imposed by the BRILLOUIN technique which principally enables to resolve fragile structure formation processes, e.g. at unstable interfaces between liquids, in space and time.

Keywords: Scanning BRILLOUIN microscopy, acoustic microscopy, BRILLOUIN spectroscopy, hypersonic properties

Zusammenfassung: Ausgehend von der klassischen akustischen Mikroskopie, basierend auf der Ultraschalltechnik, hat die vorliegende Schrift das Ziel die Entwicklung einer alternativen, über die klassische akustische Mikroskopie in mancher Hinsicht hinausgehenden Methode vorzustellen: die Raster-BRILLOUIN-Mikroskopie. Als optisches Messverfahren erlaubt die Raster-BRILLOUIN-Mikroskopie eine zerstörungsfreie Bestimmung der akustischen Eigenschaften von optisch transparenten oder halbdurchlässigen Proben im Rahmen der linearen Antwort. Es ist ein besonderes Merkmal der Raster-BRILLOUIN-Mikroskopie, dass Absolutwerte der Schallgeschwindigkeit und der Schalldämpfung gemessen werden können. Infolgedessen können ein-, zwei- oder dreidimensionale akustische Eigenschafts-profile von mechanischen Inhomogenitäten oder Heterogenitäten in durchsichtigen Materialien erstellt werden. Vorausgesetzt, dass die opto-akustische Kopplung ausreicht, werden die spektralen Eigenschaften aller akustischen Moden gleichzeitig aufgenommen. Von intransparenten Proben können die akustischen Oberflächeneigenschaften, also akustische Oberflächenwellen, charakterisiert werden. Die räumliche Auflösung ist durch das optische Messverfahren beugungsbegrenzt, so dass die laterale Auflösung bei etwa einem Mikrometer liegt. Raster-BRILLOUIN-Mikroskopie erlaubt neuerdings räumliche, zeitliche und winkelauflösende Messmethoden zu kombinieren. Das ermöglicht die kompletten elastischen Tensoreigenschaften von kondensierter Materie räumlich und zeitlich zu analysieren.

Mehrere Beispiele zur Illustration der Leistungsfähigkeit der Methode aus den Bereichen der weichen und harten Materie werden im vorliegenden Artikel diskutiert: die räumliche Auflösung transkristalliner Bereiche in einem teilkristallinen Polymer, die räumlich-zeitliche Charakterisierung polymerisations-bedingter Netzwerkbildung in Epoxid-klebstoffen an metallischen Grenzflächen und die räumliche Verteilung der Lösungs-mittelaufnahme von glasartigen polymeren Netzwerken. Die Möglichkeit zur Bestimmung der Winkelabhängigkeit der drei akustischen Moden als Funktion der Ortskoordinate liefert wertvolle Informationen über die räumliche Entwicklung des elastischen Tensors von anisotroper inhomogener oder heterogener Materie. Dies wird anhand von uniaxial verstreckten Polymerproben und Konzentrationsprofilen in Mischkristallen demonstriert. In polykristallinen Diamantplatten erlaubt die Raster-BRILLOUIN-Mikroskopie eine quantitative Bestimmung der räumlichen Verteilung von inneren Spannungsfeldern. Im Falle der BRILLOUIN-Mikroskopie sind die der Probe durch das Messverfahren aufgeprägten Störungen vergleichbar zu den thermischen Fluktuationsamplituden. Folglich können auch gegen Störung äußerst empfindliche Strukturbildungsprozesse untersucht werden. Dies wird am Beispiel von Transport- und Struktur bildungsprozessen an instabilen Grenzflächen zwischen Flüssigkeiten vorgestellt.

1. Introduction

Scanning BRILLOUIN microscopy (e.g. KRÜGER *et al.* 2001, HILLEBRANDS 2005, PHILIPP *et al.* 2011) is an experimental technique which belongs to the wide class of acoustic microscopy techniques. In the classical sense, the term 'acoustic microscopy' refers to ultrasonic techniques that employ high-frequency acoustic signals for the high-resolution acoustic imaging of materials (LEVIN *et al.* 1988, MAEV *et al.* 2002, MAEV 2008, BRIGGS & KOLOSOV 2009). The employed frequency range nowadays reaches from the MHz into the GHz regime; therefore the maximal frequencies are comparable to that encountered in BRILLOUIN microscopy.

However, ultrasonic imaging and BRILLOUIN imaging do exhibit some fundamental differences. Classic acoustic microscopy is based on a confocal pulse-echo setup. The pulse is reflected at any more or less strong change in the acoustic impedance, giving birth to an echo. Such changes of acoustic impedance could be material defects, e.g. voids or cracks, inhomogeneities or heterogeneities. The information obtained consists in the time of flight, the amplitude, and the polarization of the recorded echo. With this information alone it is impossible to determine the acoustic properties locally, like the sound velocity or sound attenuation. The collected data only allow an assessment of the change of the acoustic impedance and give a rough idea at which depth it occurs within the sample. If the sample exhibits no variation of acoustic impedance, this technique, in its classical use, provides no information at all. The imaging character comes into play when the acoustic lens system is scanned across the sample's surface. While employing high-frequency acoustic signals, a lateral resolution in the sub-micron range can be achieved and the depth resolution, for known sound velocity, is even higher. But some disadvantages are linked to the use of high-frequency signals: the sound attenuation may become so strong at high frequencies that the maximum penetration depth is of the order of a millimetre or even less. Especially many liquids and soft materials suffer from extremely high acoustic attenuation. For the well-known medical ultrasonography the probe frequency is usually around 10 MHz, thus permitting a much greater penetration depth at the cost of resolution, which is roughly a tenth of a millimetre (MAEV 2008, BRIGGS & KOLOSOV 2009).

Beside the different probe frequencies the working principles of ultrasonography and acoustic microscopy are almost the same. Regarding suitable samples for classical acoustic microscopy three requirements are to be met: first the region of interest inside the sample should be close to, i.e. at maximum 1 mm below the sample's surface, second the surface should be smooth and planar to avoid acoustic scattering and refraction, and third the sample usually has to be compatible with the use of an immersion liquid so as to facilitate acoustic coupling.

In essence, the information provided by classical acoustic microscopy is comparable to that given by light microscopy, electron microscopy or atomic force microscopy in the sense that one obtains an image related to contrasts of a certain material property. But despite all of the mentioned drawbacks, acoustic microscopy possesses a wide range of applications due to certain unique advantages: for instance this technique can be employed to optically non-transparent samples and three-dimensional imaging is possible. The main fields of application of classical acoustic microscopy are (i) non-destructive testing, like the detection of defects in welded or adhesive joints and depth profiling, (ii) electronics, like the imaging of enclosed integrated circuits, and (iii) biomedicine, like the imaging of cells or the semi-quantitative determination of elastic properties of biological tissues (MAEV 2008, BRIGGS & KOLOSOV 2009).

As demonstrated below in this article, from the viewpoint of accessible information scanning BRILLOUIN microscopy may appear as a major extension of classical acoustic microscopy. Indeed, it yields absolute values of hypersonic velocity and attenuation directly from the targeted information volume and measures these data strictly in linear response (BRILLOUIN 1922, MANDELSTAM 1926, GROSS 1930, FABELINSKII 1968, CHU 1974, BERNE & PECORA 1976, SANDERCOCK 1982, KRÜGER 1989). It is delineated below that the combination of scanning BRILLOUIN microscopy with angle-resolved BRILLOUIN spectroscopy enables the investigation of elastic tensors, also for spatial changes of the macroscopic symmetry within a sample (PEETZ 1987, MARX *et al.* 1988, 1989, KRÜGER 1989, JIMÉNEZ RIOBÓO *et al.* 1990, KRÜGER *et al.* 1990, 1994). Furthermore, this technique can distinguish the amplitudes of spatially varying stress fields (KRÜGER *et al.* 2000, 2001). Scanning

BRILLOUIN microscopy even remains operational when liquids and soft matter with high acoustic attenuation are in the focus of interest. It can nowadays actually probe molecular acoustics not only in dependence of frequency, but also in the course of time-dependent processes (SANCTUARY *et al.* 2009, 2010, PHILIPP *et al.* 2009, 2011). However, as an optical method, it requires transparent or at least translucent samples in order to study bulk acoustic waves. Note that other common terms for scanning BRILLOUIN microscopy are BRILLOUIN microscopy, spatially resolved BRILLOUIN spectroscopy, micro- BRILLOUIN spectroscopy, and BRILLOUIN imaging (e.g. AHART *et al.* 1999, 1996, JIANG & KOJIMA 2000, KRÜGER *et al.* 2001, 2004b, SANCTUARY *et al.* 2003, DEMIDOV *et al.* 2004, KIM *et al.* 2005, KOSKI & YARGER 2005, PERZLMAIER *et al.* 2005, SAKAMOTO *et al.* 2008, VINCENT *et al.* 2005a, MÜLLER *et al.* 2008, SANDWEG *et al.* 2008, SCARCELLI & YUN 2008, SO 2008, PHILIPP *et al.* 2009).

The outline of this manuscript is as follows. For the sake of completeness and overall comprehensibility, an introduction into the rather uncommon technique of classical BRILLOUIN spectroscopy and its physical background is given in chapter 2. Note that the sections marked with an asterisk* can be skimmed over during a first reading. In chapter 3, ultrasonic imaging and BRILLOUIN imaging techniques are compared. An overview of scanning BRILLOUIN microscopy and comments about different setups frequently discussed in literature follows. The subsequent historical overview, chapter 4, sketches the evolution of BRILLOUIN imaging, starting with the first attempts in 1977 (KRÜGER & UNRUH 1977). Furthermore, the broad applicability of scanning BRILLOUIN microscopy to diverse scientific fields is confirmed by illustrative examples from our own research group. Manifold recent applications of scanning BRILLOUIN microscopy concerning interphase formation within amorphous and semi-crystalline polymers and unstable interfaces between liquids are treated in chapters 5 and 6. The conclusion, chapter 7, summarizes how scanning BRILLOUIN microscopy opens a complete new world of physical acoustics for isotropic and anisotropic condensed matter.

Glossary

DETA	Diethylene triamine
DFTCE	Difluorotetrachloroethane
DGEBA	Diglycidyl ether of bisphenol A
P4MP1	Poly(4-methyl-1-pentene)
PET	Poly(ethylene terephtalate)
SBM	Scanning Brillouin microscopy
$\{x_1, x_2, x_3\}$	Orthogonal sample coordinate system
$\{y_1, y_2, y_3\}$	Symmetry coordinate system, according to IRE standards for crystals
$\{z_1, z_2, z_3\}$	Orthogonal laboratory coordinate system
λ_0	Vacuum wavelength of light
n	Refractive index
$\vec{E}_i$, $\vec{E}_s$	Incident/scattered electric field vector
$\vec{k}_{i,s}$, $\omega_{i,s}$	Wave vector/angular frequency of the incident/scattered electric field vector
$\vec{q}$	Wave vector of the sound wave
Λ	Wavelength of the sound wave
$\Omega_{qL,qT}$, $f_{qL,qT}$	Angular frequency/frequency of the quasi-longitudinally/ quasi-transversely polarized sound wave
$v_{qL,qT}$	Quasi-longitudinal/quasi-transversal hypersonic velocity
$\Gamma_{qL,qT}$	Attenuation of the quasi-longitudinally/quasi-transversely polarized sound wave
$\underline{\underline{c}}$	Elastic modulus tensor; given in Voigt notation