Visualizing Coherent Phonon Propagation in the 100 GHz Range: a Broadband Picosecond Acoustics Approach.

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Abstract: Building on a kHz Ti:sapphire source, we developed a novel pump-probe setup for broadband picosecond acoustics using a white-light continuum probe coupled to an optical multichannel analyser to take snapshots of phonon dynamics.©

OCIS codes: 290.5830 (Scattering, Brillouin), 110.5125 (Photoacoustics)

The behaviour of high-frequency acoustic excitations in disordered materials is one of the most provocative and less understood aspects of glass science. The frequency dependence of sound damping, in particular, is the result of the interplay of several physical mechanisms crucial for many theoretical models. The emerging picture suggests the existence of a frequency crossover in sound attenuation identifying a transition from a macroscopic continuum-like behaviour, due to crystal-like anharmonic damping effects, to a microscopic regime in which the dynamics is dominated by topological disorder, characteristic of the amorphous phase. The very nature of this hypothetic crossover, its frequency position and its relation to other anomalies of the amorphous state is highly debated.

In an effort to address this puzzling scenario, we developed a novel broadband setup for interferometric picosecond acoustics [1], which allows accessing in a single measurement a nearly octave-spanning range of the acoustic frequency wave-packet with unprecedented sampling efficiency (Fig. 1a). A longitudinal acoustic pulse is launched at time zero due to thermal expansion of a thin (10-20 nm) absorbing transducer layer deposited onto the surface of the sample, irradiated by an 800nm, 150-fs pump pulse obtained by a regeneratively amplified 1-kHz Ti:sapphire laser source. The acoustic pulse then travels inside the sample deposited, in turn, on a Si substrate [2]. The wavepacket motion is monitored by a delayed white-light continuum probe pulse generated by the Ti:Sa fundamental focused onto a rotating CaF_2 plate, with a spectrum extending in the 350-700 nm range. Adjacent probe pulses with and without the pump pulse are detected by a multichannel optical analyser and the (normalized) reflectivity difference spectrum is averaged over multiple exposures. We obtain two-dimensional maps as a function of probe delay and wavelength (Fig. 1b) [3] of the interference fringes between the reflections of the probe pulse at the metal transducer and at the position of the travelling acoustic strain. Each optical frequency in the broadband probe phase-matches a single acoustic frequency of the travelling acoustic wavepacket, following the momentum conservation law of stimulated Brillouin scattering. Such frequency is different in the sample and in the Silicon substrate, as it depends on sound velocity and refraction index. The amplitude of the oscillation of each spectral component in Si depends on how much it has been attenuated while travelling through the sample layer.

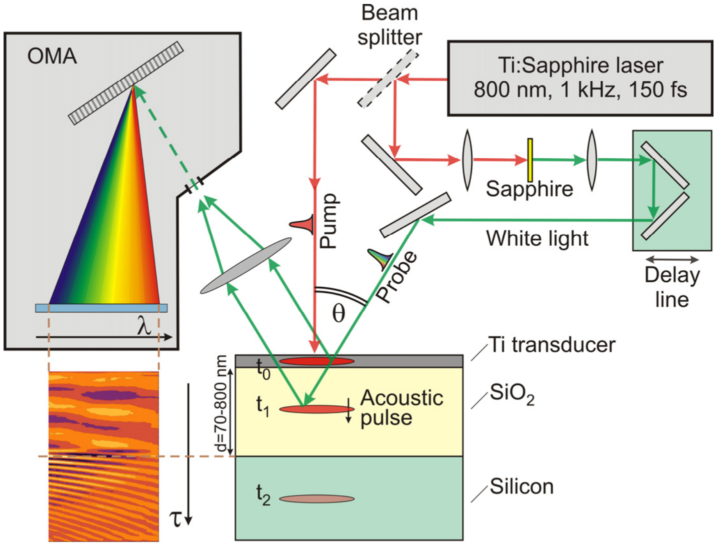

Fig. 1 Schematic layout of the broadband picosecond acoustics setup.

By repeating the measurements for set of samples with different thicknesses, we extract the sound attenuation in the 30-300 GHz frequency region. Data collected in amorphous SiO_2 and GeO_2 films obtained by chemical vapour deposition will be shown as an example. In particular, the acoustic attenuation has been measured at room temperature and at 80K, unravelling the evolution of the hypersonic sound propagation from a low frequency region, in which an elastic continuum description can be invoked, to a microscopic, high frequency region in which phonon-like modes strongly interact with structural disorder.

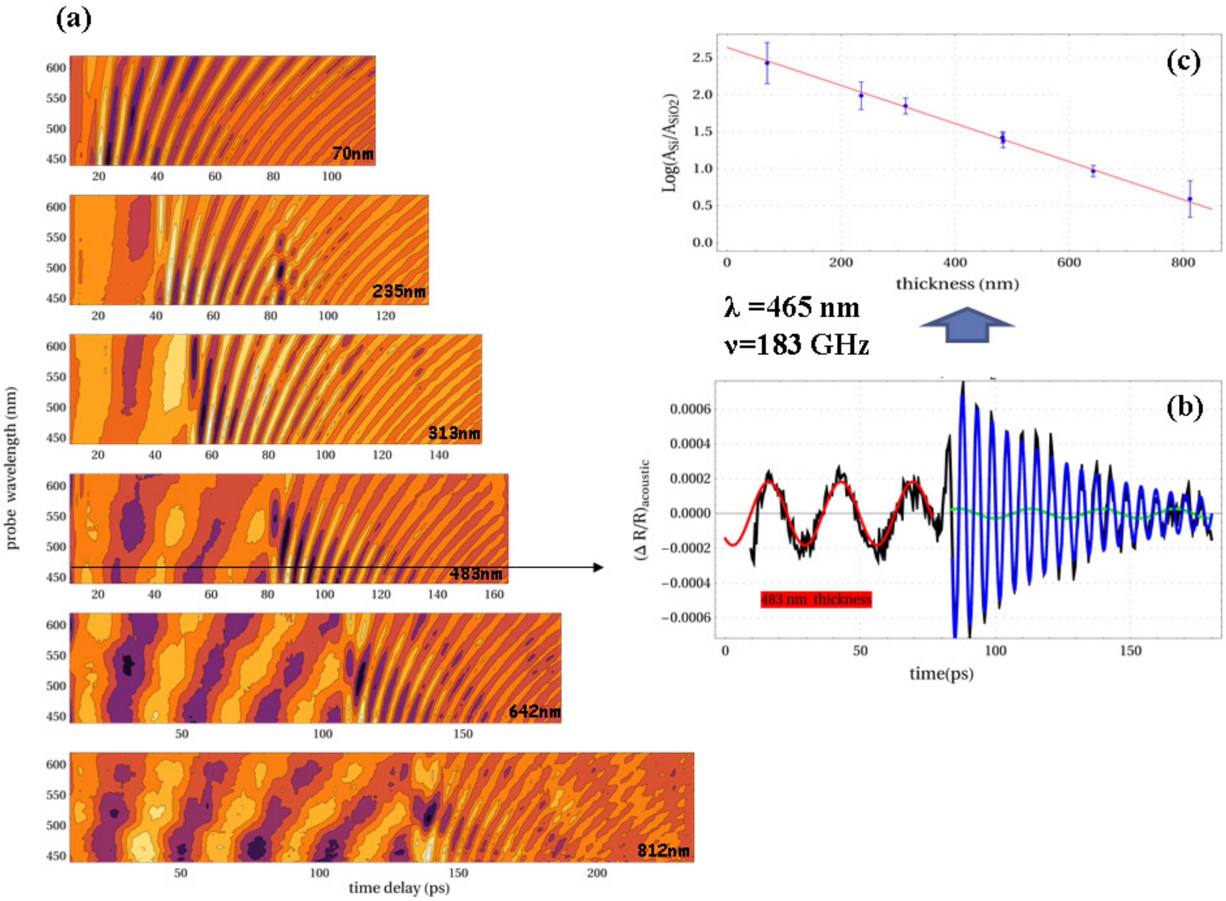

Fig. 2. a): Differential reflectivity maps as function of pump-probe delay and probe wavelength for different sample thicknesses (labelled). b) Single wavelength cut for the 483nm thick sample: the different phase-mathched phonon frequency upon crossing the sample/substrate interface is clearly visible (ν=183 GHz in the substrate). While the sample is practically transparent at the probe wavelength, strong optical absorption from the substrate can be observed. c) Amplitude of the acoustic oscillation at the interface for different sample thicknesses. The frequency dependent sound attenuation is the slope of the exponential decay at any given probe wavelength.

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