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Femtosecond ultrasonic spectroscopy using a piezoelectric nanolayer: Hypersound attenuation in vitreous silica films

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We report ultra-broadband ultrasonic spectroscopy with an impedance-matched piezoelectric nanolayer, which enables optical generation and detection of a 730-fs acoustic pulse (the width of ten lattice constants). The bandwidth improvement facilitates THz laser ultrasonics to bridge the spectral gap between inelastic light and x-ray scatterings (0.1-1 THz) in the studies of lattice dynamics. As a demonstration, this method is applied to measure sound attenuation α in a vitreous SiO_2 thin film. Our results extend the existing low-frequency data obtained by ultrasonic-based and light scattering methods and also show a $\alpha \propto f^2$ behavior for frequencies f up to 650 GHz. © *2011 American Institute of Physics*. [doi:10.1063/1.3620879]

Controlling GHz-THz acoustic waves using femtosecond optical pulses (so-called *picosecond ultrasonics* or *nanoultrasonics*) has been a substantial way to study lattice dynamics[1–6] and to manipulate electron-phonon and photon-phonon couplings in condensed matters.[7] Application of such high-frequency ultrasounds to the field of material sciences is especially prominent. This is because of the nanoscaled wavelengths and their unique potential for examining vibrational dynamics in the sub-THz range, where the conventional inelastic light and x-ray scatterings cannot be applied, i.e., a *spectral gap*.[3,5,8] It is thus highly desired to develop *broadband* laser ultrasonic spectroscopies to understand artificial nanostructures and materials with mesoscopic ordering,[9] such as glasses and liquids.

Photoacoustic coupling through a deposited metallic thin film is representative of the picosecond ultrasonic technique. With this scheme, Zhu *et al*.[2] measured the sound attenuation in amorphous SiO_2 at frequencies f up to 440 GHz. An alternative way to detect hypersound is based on the time-resolved Brillouin scattering,[1,3,5] which originates from the interference of light reflected from fixed interfaces and regions modulated by strain waves. To increase the detected acoustic frequency, Devos *et al*.[3] proposed to monitor Brillouin oscillations in stiff substrates and with short-wavelength light. It enabled investigations of hypersound attenuation in amorphous SiO_2 at $f \sim 250$ GHz. The corresponding spectroscopic analysis ($f < 200$ GHz) was recently performed by Pontecorvo *et al*. using a white-light continuum probe. Even with these developments, broadband ultrasonic experiment with frequencies up to 1 THz is still challenging at present.

In this Letter, we report a femtosecond ultrasonic spectroscopy using acoustic pulses with a full width at half maximum (FWHM) shorter than 730 fs, corresponding to a spatial width of ten lattice constants. Measurement of the hypersound attenuation α in vitreous (v-) SiO_2 is taken as an example for demonstrating the capabilities and limitations of the technique, which indicates a $\alpha \propto f^2$ behavior for frequencies f up to 650 GHz.

Following the principle of the nanoultrasonic technique,[6,10] the broadband optoacoustic transduction relied on the usage of a piezoelectric nanolayer. This nanolayer provided a localized detection sensitivity function[11] and can serve as a target for selective photoexcitation. In addition, it was essential to avoid acoustic interferences, which would otherwise narrow the detection bandwidth, by minimizing impedance mismatch at interfaces of the nanolayer. These requirements can be achieved using an $In_{0.14}Ga_{0.86}N/GaN$ single quantum well (SQW). The indium content dramatically modified the band structure as well as the photoacoustic coupling, even though the impedance mismatch between the well and the GaN barriers was only 3%.[12]

A detailed sample structure is shown in an inset of Fig. 1, including a 3-nm-thick $In_{0.14}Ga_{0.86}N$ SQW and a 7-nm-thick GaN cap layer. Both layers were grown on a c-plane sapphire substrate with a GaN buffer layer by metal-organic chemical vapor deposition. A v-SiO_2 film with a thickness of 22 nm was then prepared by plasma-enhanced chemical vapor deposition at 280 °C (the gas was a mixture of O_2, SiH_4, and Ar). Good bonding between GaN and the v-SiO_2 film was confirmed by the transmission electron microscopy. The atomic force microscopy illustrated clear atomic terraces on the GaN surface (before depositing SiO_2) with a root-mean-square roughness σ of ~2.5 Å. This discrete feature disappeared after SiO_2 deposition due to the amorphous character, while the sample surface (v-SiO_2/air) was still atomically flat ($\sigma \sim 2.5$ Å). It was noticed that the roughness-induced phonon scattering was responsible for reducing the measurement bandwidth[6] and could cause an overestimation of the α.[2] Careful investigations on the two interfaces enabled us to estimate the roughness contribution,[6] which is essential for precise estimations of the propagation attenuation.

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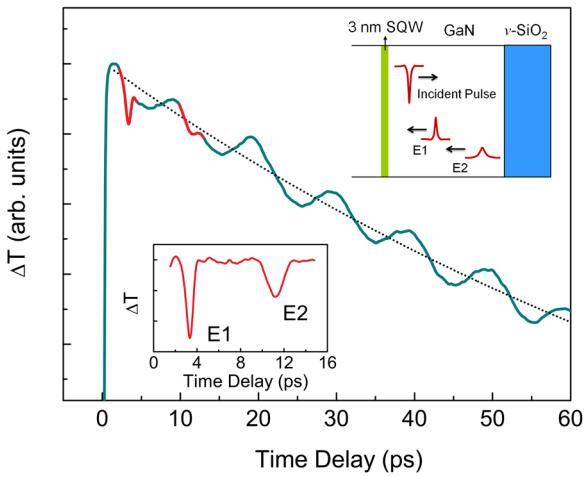

FIG. 1. (Color online) Measured optical transmission changes ΔT as a function of time delay (solid line). The slowly decaying background is depicted by the dotted line. The two acoustic echoes at the first 15 ps are plotted in red. The insets show the echo signal clarified by removing the charge and Brillouin backgrounds (Left) and schematic diagram of the nanoultrasonic experiment (Right), where the two echoes are labeled by E1 and E2.

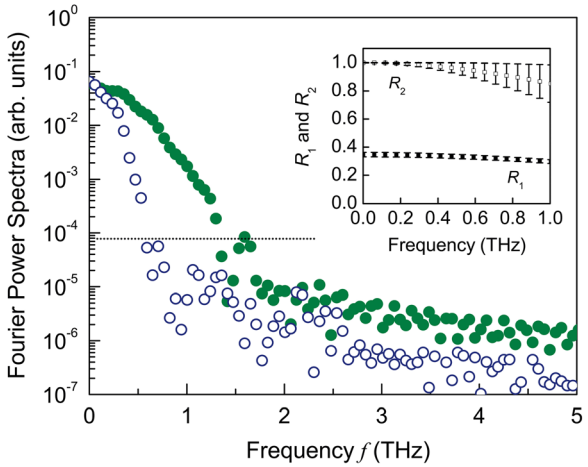

FIG. 2. (Color online) Fourier power spectra of the echo E1 (solid circles) and E2 (open circles). The horizontal dotted line denotes the noise level of the system, corresponding to $\Delta T/T \sim 10^{-6}$. The inset shows the power reflectivity of coherent waves at the two interfaces.

We used frequency-doubled optical pulses (~200 fs width) from a Ti:sapphire laser as the light source, to generate and detect femtosecond acoustic pulses. The experiment was performed at room temperature with a typical optical pump-probe setup.[6] The wavelengths of the optical pump and probe were 410 nm, corresponding to the interband transition of the SQW. Both beams were focused onto the sample with a spot diameter of 17 μm and an excitation fluence of ~180 $\mu J/cm^2$.

Fig. 1 shows a magnified measurement result. After optical excitations, thermalization and recombination dynamics of the photocarriers caused a temporal decay of the transmission changes (ΔT). On top of this carrier dynamics background, we can find a monotonic oscillation with a frequency of 102 GHz and two dips in the first 15 ps. The former was contributed from Brillouin scatterings in GaN and has been previously discussed.[6] The features concerned in this letter are the dips at 3.3 and 11.2 ps (labeled by E1 and E2, respectively), which are the acoustic echoes reflected from the two interfaces of the v-SiO_2 film.

With a sound velocity of 7.53 nm/ps,[12] the characteristic time for sounds traveling through the SQW was ~0.40 ps. Combined with the property of impedance matching, this nanolayer had the capability to excite and detect ultrasounds with frequencies higher than 1 THz. To analyze the echoes in detail, we extracted the echo signals by removing the carrier dynamics and the Brillouin backgrounds from the measured ΔT (an inset of Fig. 1). The result illustrated a FWHM of 0.73 ps for the first echo from the GaN/SiO_2 interface, corresponding to a signal bandwidth up to 1.3 THz, as shown in Fig. 2. The impedance-matched nanolayer, therefore, opens a way to bridge the spectral gap (0.1-1 THz) in the lattice dynamics studies. Furthermore, the penetration of carrier wave functions into GaN barriers resulted that the initial acoustic pulsewidth and distribution of the detection sensitivity function were broader than the film thickness.[7,11] This property, together with scattering loss during the sound propagation and a finite optical pulsewidth, made the observed echo width broader than the calculated characteristic time.[13]

Analysis of the two echoes led to the acoustic properties of the v-SiO_2. The (power) attenuation coefficient of the v-SiO_2 film can be expressed by[2]

$$\alpha = \frac{1}{2L}\ln\left[\frac{R_2 T_1^2}{R_1}\frac{P_1}{P_2}\right], \quad (1)$$

where L is the film thickness, P_i is Fourier power spectra of the echo reflected from interface i, and R_i and T_i are the power reflectivity and transmission of coherent waves for the interface i (the subscript $i=1$ or 2 for the GaN/SiO_2 and SiO_2/air interface, respectively).

Roughness-induced scattering makes the phonon-interface interaction deviate from predictions of the acoustic mismatch model, as quantitatively confirmed in the recent literature.[6] The interface diffuse scattering destructs the coherence of ultrasounds. The fraction of phonons preserving their coherence after interface reflection/refraction can be described by a specular scattering probability (SSP).[6,14] R_i and T_i are thus formulated as $R_i = SSP \times R_{i,sp}$ and $T_i = SSP \times T_{i,sp}$, where $R_{i,sp}$ and $T_{i,sp}$ obey the acoustic mismatch model and represent the reflection and transmission probability of a specularly scattered phonon.[15] With the reported material properties,[12] $R_{1,sp}$ was calculated to be 0.35 ± 0.02 ($R_{2,sp} = 1$). Moreover, SSP was mainly determined by the interface irregularity, and the corresponding formulation has been proposed using the wave-scattering theory (small-slope approximation).[6] Using this theory and the measured statistics of the interface roughness, we estimated SSP as well as T_i and R_i (the inset of the Fig. 2). Uncertainties of the estimation were predominated by the inhomogeneity of σ on different sample locations. In addition, R_i shows a decaying trend with an increase in frequency, as a character of geometric scatterings.

The attenuation coefficient (Fig. 3) can then be calculated using Eq. (1) and the measured power ratio of the echoes (P_1/P_2). α is converted into the linewidth $\Gamma/2\pi$ by $\Gamma = \alpha c$, where $c = 5.5 \sim 5.97$ nm/ps is the sound velocity in v-SiO_2.[4] Reliable bandwidth of the estimated Γ (0.18–1.3 THz) is limited by the finite accuracy of the background

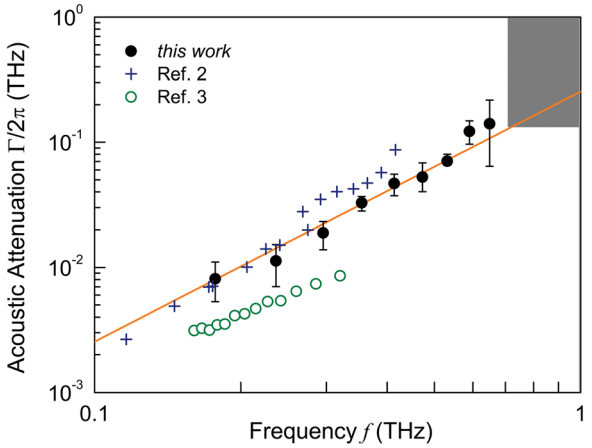

FIG. 3. (Color online) Acoustic attenuation coefficient of v-SiO_2. The colored region shows the sound attenuation at 0.65-1 THz where only the lower bound of Γ is determined from the presented experiment. Data from the previous picosecond ultrasonic measurements are also shown for comparison. The thin line with a slope of 2 is a guide for eye.

fitting and the noise level (the dotted line in Fig. 2), while at $f > 650$ GHz only the lower bound of α can be provided. We thus show the sound attenuation at $f > 650$ GHz by a colored area in Fig. 3. It was found that the ultrasonic studies on this material system[1–3] have not reached $f > 500$ GHz at the present date.

Our data in the range of 180–650 GHz show a clear $\Gamma \propto f^2$ behavior, suggesting that the responsible damping mechanism for hypersounds is anharmonicity.[3] According to the x-ray scattering experiment ($f > 1$ THz)[8] and the observed plateau in thermal conductivity of glasses around 10 K,[9] Γ should transit from f^2 regime to f^4 regime at a frequency less than the Ioffe-Regel limit (~1 THz). Without observing this transition at $f < 0.5$ THz, our ultrasonic data thus implies that the high-order damping process ($\Gamma \propto f^4$), caused by the interaction with quasilocal vibration and/or phonon scattering by frozen-in disorder,[3] could become predominant in an even higher frequency range, i.e., 0.5–1 THz.[16]

Fig. 3 also shows the literature values taken from the reported picosecond ultrasonic measurements. The presented Γ was found to be 1~2 times higher than the value from the Ref. 3, but close to that from the Ref. 2. Both reports[2,3] proposed that roughness-induced interface scatterings would result in an overestimation in Ref. 2, while this effect was calibrated in this presented work using the method confirmed in our previous effort.[6] Stronger damping observed here was, therefore, attributed sorely to the characteristic of the studied specific sample. We noticed an existing doubt on whether nanoscaled thin film can be considered as the bulk material,[8] especially as the film thickness ($L \sim 22$ nm) was comparable to the correlation length of the density fluctuation domains in v-SiO_2 (10-100 interatomic spacings).[9] Our results revealed a stronger sound attenuation, i.e., a shorter phonon mean free path, in nanoscaled glasses.

Here, we discuss limitations of the presented method to provide a guideline for the future technical development. Low spectral densities of the generated acoustic power and the detection sensitivity are general difficulties for broadband spectroscopies. They weaken the detected acoustic signal and thus restrict this metrology to the study of thin films ($L < \alpha^{-1}$). In addition, the carrier dynamics and Brillouin signals influence the low-frequency cutoff of the spectral analysis, which can be disposed using interferometric detection. Sample non-uniformity, causing the error bars (11%~54%) in Fig. 3, is also an issue to be resolved for precise analyses of nanoscaled thin films. Co-localization of the surface characterization with optical measurement can release this constraint.[6] (Compared with the inhomogeneity, Δc of ~4% in SiO_2 plays a minor role in the estimation of the error bars.)

In summary, femtosecond ultrasonic spectroscopy was demonstrated and applied to investigate hypersound attenuation in a v-SiO_2 thin film. This technique was based on an impedance-matched nanolayer, providing photoacoustic coupling with bandwidths broader than 1 THz. The measurement in the 22-nm SiO_2 indicated a $\alpha \propto f^2$ behavior in the frequency range of 180–650 GHz and a shorter phonon mean free path, compared with bulk v-SiO_2.

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