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Mesoporous-based responsive acoustic resonators operating in the GHz range

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ABSTRACT

The manipulation of ultra-high-frequency acoustic phonons in the tens of gigahertz to sub-terahertz range offers a promising avenue for technological advancements. This control can be achieved by means of semiconductor superlattices, such as GaAs/AlAs, which requires costly fabrication processes in order to achieve atomically flat interfaces to avoid phonon scattering and dephasing. In addition, acoustic resonators based on such systems are bound to fixed frequency operations. Mesoporous materials, with pores at the nanoscale, are good candidates to overcome such limitations. It has been shown that multilayered resonators based on mesoporous SiO₂ and TiO₂ support acoustic resonances in the 5-100 GHz range. These materials are susceptible to liquids and vapors infiltration into the pores, causing the modification of the effective optical and elastic parameters. Here we present a design of open-cavity acoustic resonators of SiO₂ mesoporous thin films (MTFs) responsive to the environment. The resonator is composed of an acoustic distributed Bragg reflector, to avoid phonon diffusion towards the substrate, followed by a nickel acousto-optical transducer, and the MTF on top. We characterize the resonator by using coherent phonon generation and detection experiments in a reflectivity pump-probe setup, and compare it with a simplified structure. The experimental results show that the acoustic signals in the open-cavity resonator have improved performance. Our findings unveil a promising platform for nanoacoustic reconfigurable optoacoustic devices based on soft, inexpensive fabrication methods. As a perspective to this work, we aim at studying the dependence of the acoustic resonances on the environment humidity.

Keywords: Nanophononics, Coherent Acoustic Phonons, Mesoporous Materials, Responsive Acoustic Resonators

1. INTRODUCTION

The manipulation of ultra-high-frequency acoustic phonons in the tens of gigahertz to sub-terahertz range presents a promising frontier for technological innovation across various domains.¹⁻⁴ Standard acoustic resonators based on superlattices such as GaAs/AlAs,⁵⁻⁸ face limitations stemming from costly fabrication processes and fixed-frequency operations. These challenges have motivated the exploration of alternative materials and architectures to overcome these constraints and unlock new capabilities in nanoacoustic devices.

Mesoporous materials, characterized by their nanoscale pores,⁹⁻¹² offer a compelling avenue for addressing the shortcomings of semiconductor superlattices. In particular, mesoporous SiO₂ and TiO₂ have emerged as viable candidates due to their ability to support acoustic resonances in the 5-100 GHz frequency range while also exhibiting susceptibility to environmental stimuli, such as liquid and vapor infiltration.¹³⁻¹⁶ Leveraging these properties, we propose a novel design of open-cavity acoustic resonators based on mesoporous SiO₂ thin films (MTFs), engineered to be responsive to changes in environmental humidity.

Here, we present the design and characterization of these mesoporous SiO₂ thin film resonators. By employing coherent phonon generation and detection experiments we access the acoustic response of such resonators. As

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perspective, the proposed structures can be studied at different ambient conditions. Mesoporous materials have their elastic properties modified under liquids and vapors infiltration.^{12,17,18} Therefore, these studies could potentially elucidate the influence of environmental factors (such as ambient humidity) on the resonator's performance. Our findings not only demonstrate the feasibility of utilizing mesoporous materials for nanoacoustic applications but also pave the way for the development of reconfigurable optoacoustic devices with enhanced functionality and versatility.

2. RESULTS AND DISCUSSION

We fabricated open-cavity acoustic resonators composed of mesoporous SiO₂ thin films using a sol-gel method in a dip coating approach.^{9,13,14} The resonator architecture consisted of an acoustic distributed Bragg reflector (DBR) of 3 periods of dense SiO₂/TiO₂ multilayers to prevent phonon diffusion towards the substrate, a 30-nm-thick nickel acousto-optical transducer for efficient acousto-optical transduction, and the mesoporous SiO₂ thin film serving as the resonant cavity, as shown in Fig. 1(a). The porous nature of the SiO₂ thin film facilitates the infiltration of liquids and vapors, enabling modulation of the effective optical and elastic parameters. We compare this design with a reference structure¹³ composed by the substrate, mesoporous thin film, dense SiO₂ and Ni, as depicted in Fig. 1(b). In this configuration, the dense SiO₂ protects the mesoporous layer against Ni diffusion upon deposition.

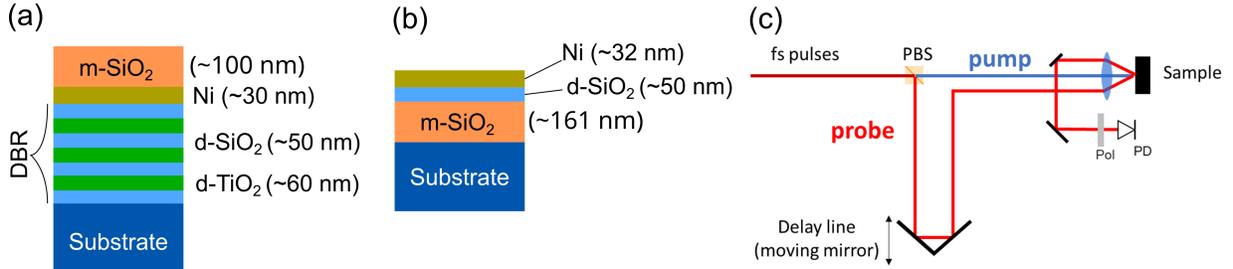


Figure 1. (a-b) Multilayer structure schematics of the (a) top-cavity acoustic resonator and (b) reference sample. (c) Schematics of the pump-probe experiment. PBS, Pol and PD stand for polarizing beam splitter, polarizer and photodetector, respectively.

For the resonators characterization, a coherent phonon generation and detection experiment was performed in a reflectivity pump-probe setup,^{19,20} shown in Fig. 1(c). A strong pump pulse, focused on the structure, interacts with the Nickel thin film that expands via thermoelasticity and generates a strain pulse.²¹ The strain wave propagates along the structure, modulating the optical refractive index of the materials. A delayed probe pulse senses the modulation of the refractive index, resulting in a modulation of its reflectivity. By scanning the relative time delay between pump and probe, the reflectivity timetrace is reconstructed, as displayed in Fig. 2(a). For the reference structure, the photoelastic interaction in the Ni layer was not sensitive to coherent phonons confined in the MTF, therefore an interferometer has been adapted to the system, to detect the surface displacement.^{13,22}

Figure 2(a) shows the reflectivity timetraces for both studied structures. At zero time delay, the rapid reflectivity change is due to the electronic excitation. It is followed by a slow thermal relaxation, superposed with some coherent oscillations due to the presence of coherent acoustic phonons. Fast oscillations in the first 100 ps, present in the reference sample and absent in the top-cavity resonator, are associated with an acoustic resonance between the metallic transducer and the dense layer.¹³

Upon Fourier transforming the time traces, we derive the corresponding phononic spectra, as illustrated in Fig. 2(b). In the case of the top-cavity resonator, three distinct peaks emerge within the 10 to 30 GHz range, related to the fundamental mode and higher harmonics of the acoustic phonons confined in the MTF. Similarly, the reference sample exhibits acoustic resonances within the mesoporous layer, alongside a higher

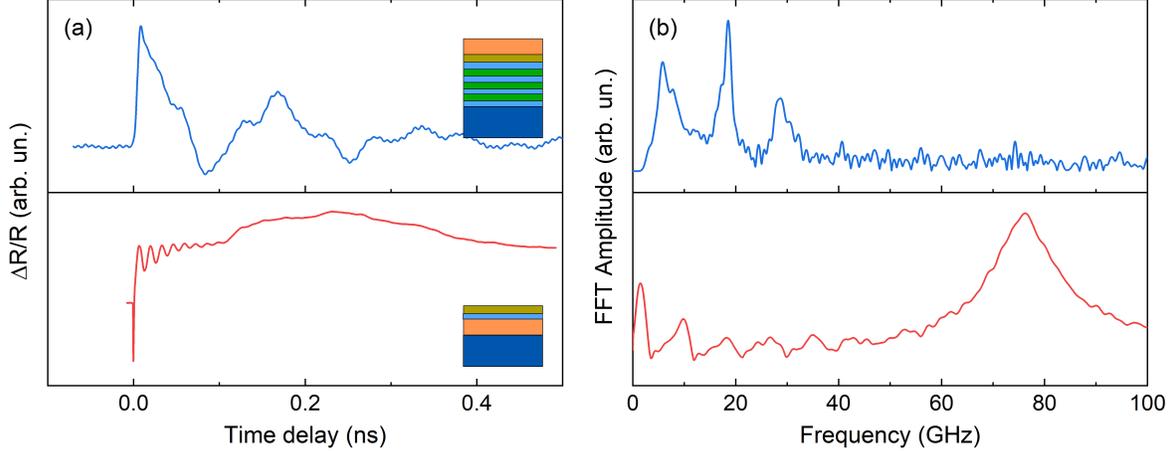


Figure 2. (a) Reflectivity time trace of the acoustic resonators. Top and bottom panels correspond to the measurements performed on the top-cavity and reference resonator, respectively. (b) Fourier transform of the respective time traces displayed in (a).

frequency peak (around 75 GHz) associated with the resonance in the Ni film.¹³ A notable observation when comparing both structures is the clear definition of peaks in the open-cavity resonator, and the possibility of accessing the resonances with standard reflectivity measurements. Additionally, positioning the mesoporous thin film as the outermost layer of the structure facilitates easier infiltration of liquids and vapors.¹⁵ Our findings highlight the practical utility of this device for realizing tunable and reconfigurable optophononic devices within the GHz range. As a perspective, we aim to investigate the resonance dependency on liquid infiltration through pump-probe experiments conducted at varying relative humidity levels.

3. CONCLUSION

In conclusion, we have demonstrated the design and characterization of open-cavity acoustic resonators based on mesoporous SiO₂ thin films, showcasing their capability to support acoustic resonances in the GHz range. Our findings underscore the potential of mesoporous materials as a versatile platform for reconfigurable optoacoustic devices, offering advantages in terms of cost-effectiveness, and adaptability. By harnessing the unique properties of mesoporous SiO₂ thin films, such as their nanoscale porosity and sensitivity to external stimuli, we have established a promising framework for the development of novel nanoacoustic technologies. These devices hold considerable promise across a range of applications, including sensing, and signal processing, where dynamic responsiveness to environmental changes can be important.

Moving forward, further research is required to explore additional functionalities and optimize the performance of mesoporous SiO₂ thin film resonators. This may involve investigating the devices at different humidity conditions, exploring alternative pore size distributions, optimizing fabrication processes, and exploring novel integration schemes with complementary materials, geometries, and components.^{7, 15, 23, 24}

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