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Design of responsive mesoporous-based open-cavity acoustic resonators operating in the GHz range

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ABSTRACT

Designing acoustic resonators at ultra-high frequencies is a promising pathway for important technological advancements, including quantum technologies. Resonators operating in the tens-of-gigahertz range are usually based on semiconductor multi-layered distributed Bragg reflectors (DBRs). However, their inherent lack of tuning capability imposes constraints on specific applications, such as sensing. Mesoporous materials, with pores at the nanoscale, could potentially bridge the gap for responsive acoustic resonators. These cost-effective materials support gigahertz acoustic resonances, and their high surface-to-volume ratio allow for engineering resonators with novel functionalities, such as optoacoustic sensors or switches. For instance, the infiltration of liquids and vapors into the pores modifies the material's optical and elastic parameters, leading to direct modification of the acoustic resonances. Here, we theoretically propose open-cavity multi-layered acoustic resonators based on SiO₂ mesoporous materials that are responsive to the environment, e.g., relative humidity changes. The resonators are formed by a DBR of dense SiO₂/TiO₂ multilayers, followed by a Ni acousto-optical transducer, to both generate and detect acoustic phonons, and finalized by the mesoporous as the last layer, so it has access to the environment. In this design, gigahertz acoustic phonons can be effectively confined at the mesoporous layer. On one hand, the acoustic DBR prevents their leakage toward the substrate, and on the other hand, they cannot propagate through the air, creating an ideal acoustic mirror at the mesoporous-air interface. We present two case studies based on this approach and show, by numerical simulations using transfer matrix method, the responsiveness and tunability of such resonators upon ambient humidity changes. The simulation results also indicate that these devices can be characterized through transient reflectivity experiments. Our study could guide the way for designing effective nanoacoustic sensing and adaptable devices, utilizing cost-effective fabrication techniques.

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

1. INTRODUCTION

The engineering and control of acoustic phonons at the nanoscale is at the core of nanophononics fundamentals, and holds the potential to breakthrough applications in areas such as quantum communication and data processing.¹ Tens-of-gigahertz nanoacoustic resonators are usually based on multilayered distributed Bragg reflectors (DBRs).²⁻¹⁰ The lack of tunability of such resonators can be the bottleneck for further advancements, and therefore, developing strategies for dynamical tuning of acoustic resonators, a nearly unexplored field,^{1,11,12} is of utmost importance in the nanophononics domain. A potential candidate for active tunable acoustic resonator are the mesoporous thin films (MTFs), with pores at the nanoscale.^{13,14} Oxide-based MTFs can be conceived with soft fabrication processes based on sol-gel and dip coating approach.^{15,16} The high surface-to-volume ratio plays a major role on the chemical functionalization of these materials.¹⁷ On the nanoacoustics domain, the liquid and vapor infiltration into the nanopores changes the optical and elastic properties of these materials.¹⁸⁻²⁰ By designing an acoustic resonator with MTFs as the active elements, the acoustic resonance shifts in frequency and amplitude under liquid infiltration.^{11,12} Here, we theoretically propose the design of acoustic resonators based on such structures, and demonstrate two case studies.

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2. DESIGN OF MESOPOROUS OPEN-CAVITY ACOUSTIC RESONATORS

We design the open-cavity acoustic resonators by implementing the transfer matrix method (TMM) along with the photoelastic model.^{21,22} This implementation allows us to study pump-probe experiments for the generation and detection of coherent acoustic phonons. Experimentally, a strong pump laser pulse interacts with the structure, triggering the phonon dynamics in the system.^{23–25} The modulation of the material refractive index due to these vibrations takes place. Finally, a delayed probe reaching the structure detects transient reflectivity changes due to the presence of coherent acoustic phonons.

For the model implementation, a white phonon spectrum propagating from the substrate towards the surface is considered. With the TMM, we compute the normalized stationary solutions for each phonon frequency. This allows us to calculate the acoustic phonon transmission spectrum and the frequency-dependent acoustic field distribution along the structure. The incident laser electric field profile can also be calculated, by considering Maxwell’s equations for electromagnetic waves.²⁶ Finally, the acoustic phonon generation spectrum ($G(\omega)$) is determined by an overlap integral of the incident pump’s electric field ($E(\nu, z)$), the strain field ($\eta(\omega, z)$), i.e., the derivative of the acoustic displacement, and the material-dependent photoelastic constant ($k(z)$):

$$G(\omega) = \int k(z)\eta(\omega, z)|E(\nu, z)|^2 dz, \quad (1)$$

where ω and ν are related to the acoustic and optical frequencies, respectively. Similarly to equation 1, the coherent phonon detection spectrum ($D(\omega)$) is calculated with an overlap integral of the probe’s complex electric field ($E(\nu, z)$), the strain field, the photoelastic constant, and the generation spectrum, in the form:

$$D(\omega) = \int G(\omega)p(z)\eta(\omega, z)E(\nu, z)^2 dz \quad (2)$$

The acoustic impedance mismatch between the layers, associated with the contrast in mass density and speed of sound of the involved materials, leads to frequency-dependent changes of transmission or reflection of the sound waves at the interfaces. Therefore, in the case of acoustic resonators based on mesoporous materials, liquids and vapors infiltration into the mesopores can significantly change the resonance frequency by changing the material average sound velocity. A simple approach could be to submit the device to different relative humidity (RH) conditions.

The difference in acoustic impedance between the layers, related to variations in mass density and speed of sound of the materials involved, causes frequency-dependent alterations in the transmission or reflection of sound waves at the interfaces. Consequently, in acoustic resonators based on mesoporous materials, infiltration of liquids and vapors into the mesopores can notably modify the resonance frequency by altering the average sound velocity of the material., which could be achieved by subjecting the device to varying relative humidity (RH) levels.

3. RESULTS AND DISCUSSION

Here, we have engineered an acoustic resonator formed by an acoustic DBR based on dense SiO₂ and TiO₂ materials, followed by a nickel layer (acousto-optical transducer), one extra dense TiO₂ layer, and the mesoporous thin film on the top, as seen in Fig. 1(a). All the layers have thickness of $\lambda/4$, associated with the acoustic wavelength (λ) at a design frequency of 100 GHz. In this configuration, the DBR at the bottom of the structure reflects phonons in a frequency range around 100 GHz, and the mesoporous-air interface is a perfect mirror for acoustic phonons, as they cannot propagate into the air. This results in a confinement of acoustic phonons at the mesoporous layer, as depicted by the maximum acoustic displacement at this layer in Fig. 1(a).

In addition we have designed another structure similar to the previous one, but with a mesoporous cavity composed by a $3\lambda/4$ spacer. Figure 1(b) displays the coherent phonon detection spectrum of the two structures, where a pronounced peak at 100 GHz is apparent. This result is a purely photoelastic interaction, indicating that in a pump-probe experiment the coherent acoustic phonons can be detected in a standard reflectivity configuration, in contrast with what has been reported for a similar structure, in which the surface displacement was the major contribution to the signal, requiring an interferometric detection setup.^{13,14}

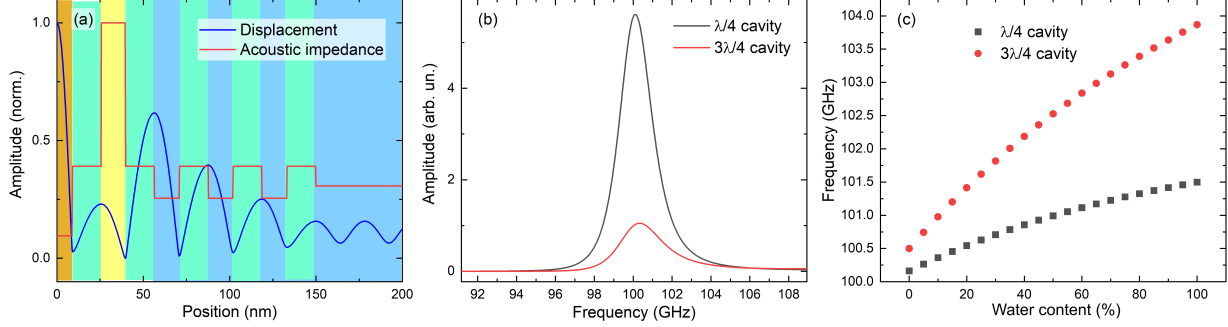


Figure 1. (a) Layer structure of the acoustic resonator with $\lambda/4$ cavity spacer. Normalized displacement pattern and acoustic impedance of the structure are displayed in blue and red lines, respectively. The colored areas indicate the layer composition as follows: dense SiO_2 , dense TiO_2 , Nickel and mesoporous SiO_2 are respectively related to blue, green, yellow and dark yellow. (b) Coherent phonon detection spectra for the $\lambda/4$ and $3\lambda/4$ cavity spacer resonators. (c) Frequency shift of the mode at 100 GHz as a function of water content inside the pores for both structures.

Finally, we have studied the influence of humidity changes in the acoustic response of both resonators. To account for liquid infiltration we have used the effective medium approximation, where weighted averages of the mass density and speed of sound between the solid SiO_2 matrix, air and water composing the MTF, have been taken into account, while the layer thickness is kept constant. We then perform the simulation with a variable humidity between 0 and 100 %, in which the reference 100 GHz mode is calculated at 0 % humidity. The frequency dependence on water content inside the pores for both resonators is displayed in Fig 1(c). One can see that a blueshift of the acoustic resonance is present on both cases, mainly associated to an average increase of the sound velocity with liquid infiltration. It is worth noting that the resonator with a $3\lambda/4$ cavity has a stronger response. This can be related to a penetration of the field to the previous layer, leading to a slightly different cavity length in each case, which results in different frequency shifts.

4. CONCLUSION

In summary, we have introduced a novel design for optoacoustic sensing devices incorporating mesoporous thin films as the active element. By infiltrating the thin film with liquid, its elastic properties undergo gradual modification, resulting in a shift in the resonance characteristics of an acoustic resonator employing these materials. Through theoretical analysis, we have showcased the promising potential of these devices in environmental sensing applications. We have also shown that different designs of the acoustic resonator can lead to different acoustic responses. The distinctive attributes and tunability of MTFs enable the conception of responsive acoustic resonators. This capability paves the way for the development of low-cost optoacoustic tunable nanodevices. Our research elucidates a novel framework for nanoacoustic sensing and adaptable devices, leveraging cost-effective fabrication techniques.

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