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Phonon dynamics in novel materials and hybrid structures

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ABSTRACT

Nanometer-thick multilayer structures, characterized by contrasts in elastic properties, present promising avenues for engineering and manipulating acoustic phonons at the nanoscale. Semiconductor nano-acoustic cavities, particularly those based on Distributed Bragg Reflectors (DBRs), have demonstrated unique capabilities in simultaneously confining light and acoustic phonons. This dual confinement enhances the generation and detection phononic fields, making these structures attractive for ultra-high-frequency applications and as platforms for simulating solid-state systems. In this study, we further explore the possibilities of hybrid nanostructures that could be both tunable and responsive to ultrafast changes in elastic properties induced by external stimuli such as temperature, humidity, and electrical fields. Building upon our theoretical simulations, our experimental investigation focuses on the dynamics of acoustic phonons spanning the frequency range of 5-500 GHz, utilizing near-infrared pump and probe ultrafast transient reflectivity experiments. The materials under investigation include mesoporous SiO₂/TiO₂ multilayers with a Nickel transducer, GaAs/AlAs DBR incorporating mesoporous SiO₂ as an open cavity layer, YBCO/STO multilayers, and other potential responsive materials. Our long term objective is to uncover the interplay between these nanostructures and external stimuli through systematic experimentation, shedding light on their tunability and responsiveness. Our experimental findings pave the way for developing nanoacoustic sensing technologies and reconfigurable optoacoustic nanodevices.

Keywords: Nanophononics, Coherent Acoustic Phonons, Ultrafast pump-probe technique, Reconfigurable Acoustic Resonators, Nanoacoustic sensing devices

1. INTRODUCTION

Nanotechnology has facilitated the exploration of acoustic phenomena at the nanoscale, offering unprecedented opportunities for engineering and manipulating phonons in novel materials and hybrid structures.¹⁻⁵ Semiconductor nano-acoustic cavities, notably those based on Distributed Bragg Reflectors (DBRs), have emerged as promising platforms for confining both light and acoustic phonons, paving the way for ultra-high-frequency applications and simulating solid-state systems.⁶⁻¹³ In this paper, we extend this exploration by focusing on hybrid nanostructures potentially responsive to external stimuli, to develop nanoacoustic sensing technologies and reconfigurable optoacoustic nanodevices.

Our investigation is centered on hybrid nanostructures that incorporate materials with potentially tunable elastic properties into DBR-based resonators. These materials would enable tunable and responsive acoustic behavior,^{14,15} thereby expanding the functionality of nanoacoustic devices. We explore materials such as mesoporous SiO₂/TiO₂ multilayers and GaAs/AlAs DBRs with mesoporous SiO₂ serving as an open cavity layer.

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The mesoporous material is sensitive to environmental stimuli, such as liquid and vapor infiltration, making it engineered to be responsive to changes in environmental humidity.^{5,16,17} Furthermore, YBCO/STO multilayers are under investigation, the elastic properties of YBCO are expected to vary with temperature.

To elucidate the anticipated behavior of these hybrid structures, we present the design of the hybrid nanostructures along with simulations. Additionally, we discuss the experimental methodologies currently under investigation to study the dynamics of acoustic phonons.

2. TOWARDS RECONFIGURABLE NANOACOUSTIC RESONATORS

Here we discuss the concept of nanoacoustic resonators to achieve reconfigurability. Two configurations are proposed: In the first configuration, an open cavity design is employed, featuring a responsive material as a top spacer layer (Fig. 1a). In the second configuration, the responsive material is positioned atop a Fabry-Perot cavity (Fig. 1b). In both scenarios, the responsive material is placed at the top of the structure to facilitate easy access for inducing phase transitions via optical means or direct interaction with external stimuli. The frequency of the confined mode is expected to shift as the material undergoes a phase transition or in response to external stimuli, due to changes in its elastic properties. We have calculated the surface displacement caused by the acoustic phonons using the transfer matrix method, demonstrating the variation of surface displacement as the material undergoes a phase transition or responds to external stimuli. Ref. 16 provides a detailed exploration of mesoporous material-based acoustic resonators, showcasing their sensitivity to variations in relative humidity.

In contrast to the design discussed above, similar reconfigurability can be achieved by designing the structure using phase transition material as one of the constituent materials in the Distributed Bragg Reflector (DBR) of the cavity structure. For example, in the case of YBCO/STO-based DBR samples, the properties of YBCO (a high-Tc superconductor) are expected to vary with temperature, causing changes in the phonon reflectivity stopband corresponding to different contrasts in elastic properties. Both changes in the elastic properties of the spacer layer and variations in the acoustic stop band can be experimentally accessed by ultrafast pump-probe transient reflectivity technique,² which is detailed in the next section.

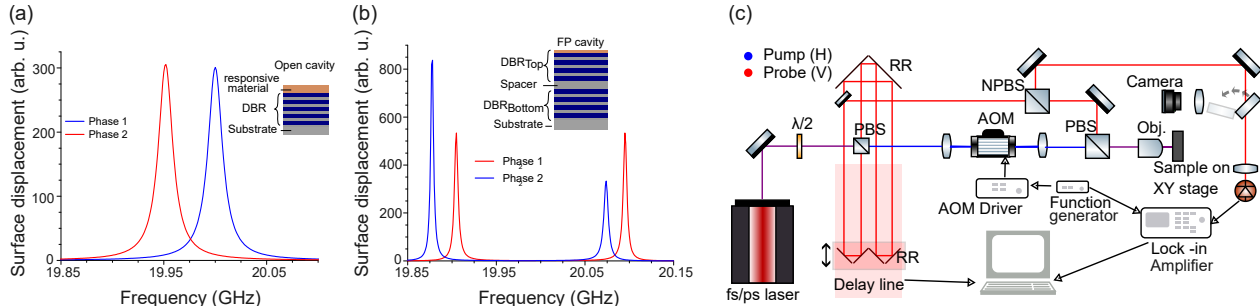


Figure 1. (a) and (b) depict surface displacement variations of the responsive material undergoing phase transition or exposed to external stimuli. In (a), the responsive material acts as the spacer layer of an open cavity structure, while in (b), it is placed on a Fabry-Perot cavity structure. (c) Schematic of the transient reflectivity pump-probe setup.

3. EXPERIMENTAL RESULTS

Ongoing experimental investigations are being conducted using pump-probe transient reflectivity technique with both femtosecond and picosecond lasers. These experiments aim to elucidate the phonon dynamics across a wide frequency range (5-500 GHz) in the proposed nanostructures. The experimental setup is detailed in Fig. 1c. An ultrashort laser pulse (100 fs to a few ps) serves as the pump pulse, inducing thermal stress and launching acoustic phonons in the nanostructure.¹⁸ The dynamics of generated acoustic phonons are detected via transient reflectivity measurements using a time-delayed probe pulse. Changes in reflectivity, resulting from the modulation of the structure's refractive index by propagating acoustic phonons, offer insights into their dynamics. Furthermore, time-frequency analysis of the transient reflectivity signal can reveal the responsiveness of the nanostructures to external stimuli.

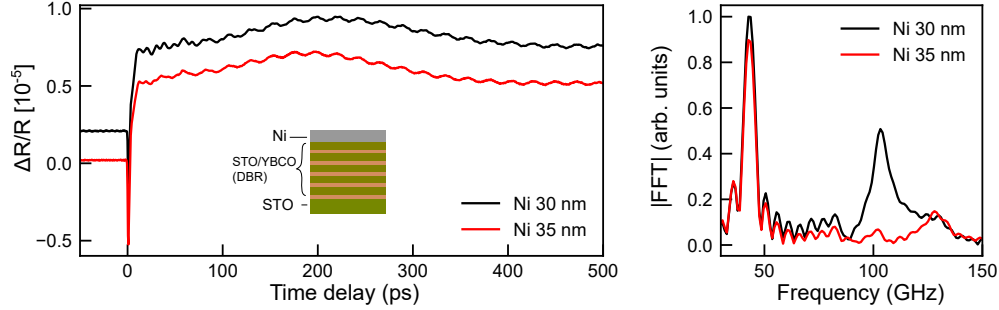


Figure 2. (a) Pump-probe reflectivity signal as a function of the delay between the pump and the probe for different thicknesses of Ni on YBCO/STO phonon mirror. (b) Fourier spectra of the transient reflectivity signal shown in (a).

Fig. 2a depicts transient reflectivity signals observed in hybrid open resonators comprising a Ni layer atop 5 periods of YBCO/STO layers as a mirror superlattice, which is designed for a stop band from 90 GHz to 140 GHz. We tested the hybrid resonator with two different Ni thicknesses, nominally 30 nm and 35 nm. The oscillatory part of the signal after the zero time delay corresponds to the acoustic modes generated by the pump laser pulse. The Fourier spectrum (Fig. 2b) of the acoustic contribution of the signal reveals two dominant modes in both samples. The lower frequency mode, independent of the thickness of Ni, located at 45 GHz in the frequency spectrum corresponds to Brillouin oscillations in the structure. These arise from the interferences between the probe light reflected by the sample's stationary interfaces and that scattered by the propagating longitudinal acoustic wave in the structure.^{19–21} This frequency value aligns well with the estimates for the refractive index in STO and for the 840 nm wavelength of the probe laser beam. The mode which is inhibited or enhanced within the designated stop band range of the mirror superlattice, depending on the thickness of the Ni layer, can be attributed to the confined mode in the Ni spacer of the hybrid cavity structure.² These preliminary results indicate promising prospects for the development of tunable acoustic resonators in future projects. In this case, it can be achieved by varying the temperature. We anticipate variations in the elastic properties of high-Tc YBCO superconductor at phase transition temperatures. This will lead to changes in the phonon reflectivity stopband of the mirror superlattice, thereby tuning the cavity mode.

4. CONCLUSION

In conclusion, our research explores hybrid nanostructures responsive to external stimuli like temperature, humidity, and electrical fields. The investigation, guided by theoretical simulations, employs pump and probe ultrafast transient reflectivity experiments to examine acoustic phonon dynamics (5–500 GHz) in materials such as mesoporous SiO₂/TiO₂ multilayers, GaAs/AlAs DBR with mesoporous SiO₂, and YBCO/STO multilayers. Findings reveal the interplay between nanostructures and stimuli, revealing their tunability and responsiveness, with potential applications in nanoacoustic sensing technologies and reconfigurable optoacoustic nanodevices.

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