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High elastic impedance terasonic hybrid superlattices

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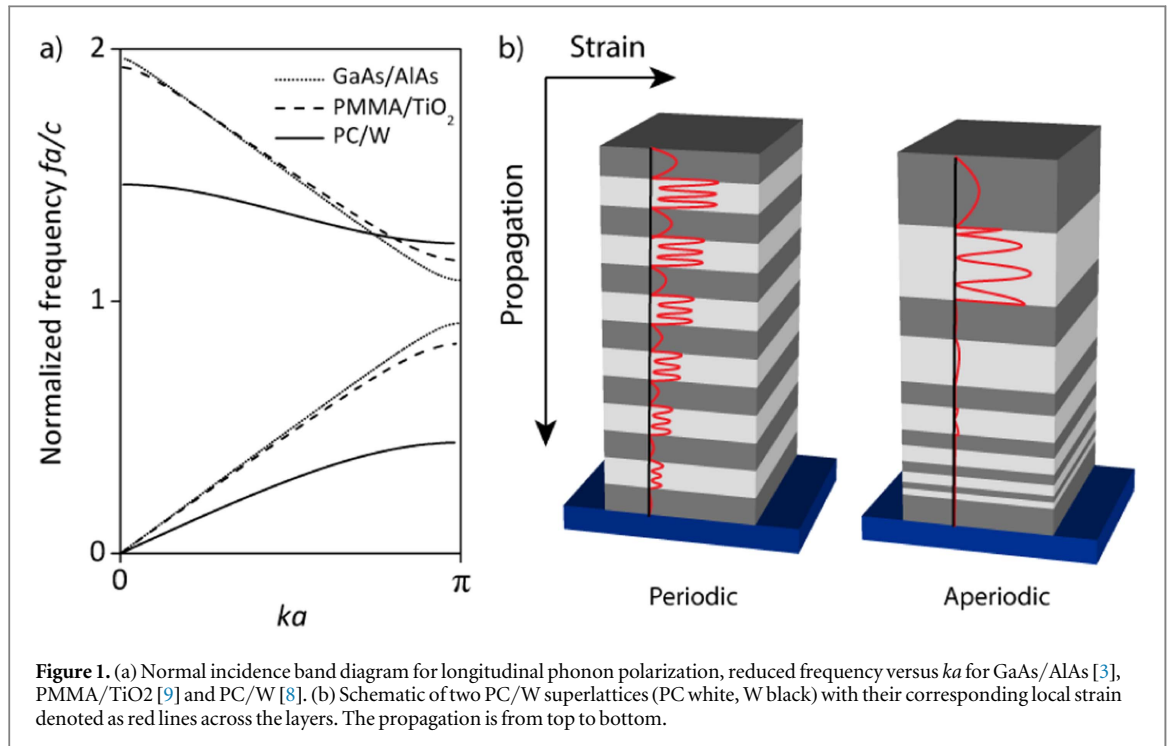
Abstract

The fabrication of new hard/soft hybrid superlattices (SLs) by pulsed laser deposition strongly suppresses interfacial intermixing, as compared to the strong infiltration occurring in their analogue periodic structures prepared by spin-coating. Döring *et al* (2016 *New J. Phys.* **18** 092002) report on the growth of SLs, optimally realizing the wealth of hard and soft materials to boost the terasonic bandgap width and enhance phonon localization upon designed layer thickness sequence in aperiodic SLs. Optimal utilization of the fabrication technique could drive the design of architected SLs with strong photon–phonon coupling and tuned heat conduction.

The selective transmission of high frequency elastic waves in GaAs/AlGaAs superlattices (SLs) in 1979 has inaugurated the idea of phonon filtering [1]. The research of phononic band diagrams started earlier than the theoretical discovery [2] of complete phononic band gaps in 1993, with the study of folded phonons in a superconductor (GaAs/AlAs) SL using Raman spectroscopy [3]. More recently, some evidence of a hypersonic band gap and zone-folded phonons was reported for porous silicon [4] and a polymer [5] SL using Brillouin and pump-probe spectroscopy. For hybrid SLs, however, the first observation of sizable normal incidence hypersonic band gap was reported in porous silica (p-SiO$_2$)/poly (methylmethacrylate) (PMMA) [6]. While the new soft matter based SL allow facile engineering of the band gap region [7] through layer thickness, elasticity and sequence, inevitable infiltration of the hard layer (SiO$_2$, TiO$_2$, BaTiO$_3$) by the polymer due to the spin-coating fabrication sets an upper limit in the band gap width as shown in figure 1(a).

Writing in *New Journal of Physics,* Döring *et al* [8] report the growth of hard tungsten, (W)/soft (polycarbonate, PC) SL by pulsed laser deposition (PLD) with low interfacial roughness and sharp periodicity of the order of 20 nanometers. The utilization of the fabrication technique for polymer deposition is not trivial because of material degradation and ill-defined packing that can significantly impact the soft layer properties. As shown by Döring *et al* [8], laser fluence can impact on both surface roughness and elastic Young's modulus (E) of the PC layer, but the mechanism is still elusive; it is hardly conceivable that deposited PC can assume about twenty times higher mechanical strength than it is commonly accepted. Notably, this parameter is critical for the identification of the acoustic resonances in their pump-probe reflectivity experiment. A thorough material characterization at different fabrication conditions should be a relevant topic for future investigation.

The almost pure hard W layers leads to very large elastic impedance contrast, $Z = (\rho c)_W / (\rho c)_{PC} > 10^3$ between W and PC where ρ denotes the mass density. The impact on the normal incidence bandgap width for longitudinal waves is illustrated in figure 1(a) along with the highest record in SLs prepared by spin-coating. The band gap width $\Delta f/f_G$, normalized to the frequency f_G in the middle of the bandgap should increase from about 30% for spincoated symmetric TiO$_2$/PMMA [9], to 140% for the periodic PC/W SL fabricated by the PLD technique. The full dispersion $\omega(k)$ (frequency versus phonon wave vector) for transparent SLs can be recorded by Brillouin spectroscopy [4, 6, 7, 9]. Using pump-probe spectroscopy, Döring *et al* [8] did not obtain $\omega(k)$ of a periodic W/PC SL with a lattice constant of $d = 17.5$ nm. Instead, only a single resonance frequency at 0.32 THz dominated the time-dependent reflectivity signal. It has been envisaged as a Bloch-like eigenmode $\omega_n = nc\pi/a$ with c being the longitudinal sound velocity in the nanocomposite material and $n = 1, 2, 3...$ From the initial slope of the band diagram in figure 1(a), $c = 3620$ m s^{-1} and the estimated $f_n = nc/2d$ for $n = 3$ captures the resonance frequency. This single frequency falls in the fourth band and the unexpected missing of the predicted

Figure 1. (a) Normal incidence band diagram for longitudinal phonon polarization, reduced frequency versus *ka* for GaAs/AlAs [3], PMMA/TiO2 [9] and PC/W [8]. (b) Schematic of two PC/W superlattices (PC white, W black) with their corresponding local strain denoted as red lines across the layers. The propagation is from top to bottom.

lower bands in the experiment is attributed to their weak contribution to the reflectivity signal. An experimental access to $\omega(k)$ of this new SL is necessary for a thorough comparison with the theory that will also allow a validation of the nanolayers elastic parameters.

Driven by numerical FDTD simulations, Döring *et al* [8] have addressed the blocking of the resonance mode (0.32 THz) whilst penetrating into the last W layer (weak strain in the periodic W/PC in figure 1(b))of an aperiodic W/PC SL, schematically shown in figure 1(b). Again, a single resonance mode was experimentally resolved at 0.135 THz that theoretically can hardly reach the second W layer (figure 1(b)). This interesting direction–dependent elastic wave propagation blocking could be experimentally proved by observing the reflectivity signal from the top of the SL. Indirectly, Döring *et al* [8] have revealed this effect though the localization of the observed resonance mainly into the thick (20.2 nm) W layer on the top of the SL.

Apart from the attractive phononic properties, which should be fully elucidated along and normal to the periodicity direction, the present SL might also exhibit interesting thermal properties. The heat conductivity (κ) of these materials can be dependent of the structural SL characteristics as suggested by Döring *et al* [8]. As the layer thicknesses commensurate typical length scales of phonon mean free paths, it is even possible to increase κ in SLs beyond the constituents value through the manipulation of the interfacial roughness [10, 11]. Yet, the unidirectional bandgap can bias cross-plane and in-plane heat transport that can vary in the case of aperiodic SLs. In addition, the reported strong phonon localization might be beneficial for strong coupling [12] to photons in the THz region. Thus challenges remain with respect to the elucidation of the relative role of the structure, material parameters and interfacial thermal resistance to the thermal properties and optomechanics of these SLs. All this will provide exciting possibilities for the fabrication of multifunctional nanohybrid materials.

Acknowledgments

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