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Optically-Controlled Nano-Transducers Based on Cleaved Superlattices for Monitoring Gigahertz Surface Acoustic Vibrations

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1 **ABSTRACT:** Surface acoustic waves (SAWs) convey energy at subwavelength depths along surfaces.
2 Using interdigital transducers (IDTs) and opto-acousto-optic transducers (OAOTs), researchers have
3 harnessed coherent SAWs with nanosecond periods and micrometer localization depth for various
4 applications. These applications include the sensing of small amount of materials deposited on surfaces,
5 assessing surface roughness and defects, signal processing, light manipulation, charge carrier and exciton
6 transportation, and the study of fundamental interactions with thermal phonons, photons, magnons, and
7 more. However, the utilization of cutting-edge OAOTs produced through surface nanopatterning
8 techniques has set the upper limit for coherent SAW frequencies below 100 GHz, constrained by factors
9 such as the quality and pitch of the surface nanopattern, not to mention the electronic bandwidth limitations
10 of the IDTs. In this context, unconventional optically-controlled nano-transducers based on cleaved
11 superlattices (SLs) are here presented as an alternative solution. To demonstrate their viability, we
12 conducted proof-of-concept experiments using ultrafast lasers in pump-probe experiments on SLs made of
13 alternating $\text{Al}_x\text{Ga}_{1-x}\text{As}$ and $\text{Al}_y\text{Ga}_{1-y}\text{As}$ layers with approximately 70 nm periodicity and cleaved along their
14 growth direction to produce a periodic nanostructured surface. The acoustic vibrations, generated and
15 detected by laser beams incident on the cleaved surface, span a range from 40 GHz to 70 GHz,
16 corresponding to the generalized surface Rayleigh mode and bulk modes within the dispersion relation.

17 This exploration shows that, in addition to SAWs, cleaved SLs offer the potential to observe surface-
18 skimming longitudinal and transverse acoustic waves at GHz frequencies. This proof-of-concept
19 demonstration below 100 GHz in nanoacoustics using such an unconventional platform might be useful for
20 realizing sub-THz to THz coherent surface acoustic vibrations in the future, as SLs can be epitaxially grown
21 with atomic-scale layer width and quality.

KEYWORDS: surface and surface skimming acoustic waves, superlattices, opto-acousto-optic
transducers, nanoacoustics, picosecond laser ultrasonics, coherent GHz phonons, ultrafast laser
spectroscopy

22 Surface acoustic waves (SAWs) possess the distinct ability to interact with various state variables
23 within solids. These variables include electrons, spins, photons, magnons, thermal phonons, and
24 strain, enabling the evaluation and/or modification of material properties at subwavelength depths
25 near the surface. The significance of SAWs spans over six decades, beginning with the invention
26 of interdigital transducers (IDTs) in 1965¹ and their subsequent use in combination with lasers for
27 SAW monitoring in 1968.² While lower-frequency SAWs, falling within the ultrasonic range, have
28 been influential in communication and signal processing technologies,³ higher-frequency SAWs
29 have found applications in diverse fields. These application include material characterization,⁴
30 photonic modulation,^{5,6} optomechanics,⁷ the transport of other excitations,⁸⁻¹⁰ and interactions with
31 magnons.¹¹ SAWs are involved in a variety of products for Information and Communication
32 Technologies (ICTs), including mobile devices (utilized in central processing units, signal
33 converters and gyroscopes) and devices associated with the “Internet of Things”, where they find
34 applications in sensors and integrated energy converters.

35 At present, the upper operational frequency limit for coherent SAWs controlled by IDTs or opto-
36 acousto-optic transducers (OAOTs) remains below 100 GHz.^{12,13} Extending this frequency range
37 to sub-terahertz (sub-THz) range in integrated elasto-optic modulators and SAW processors/filters
38 holds significant potential for future ICT devices. Access to higher frequencies and broader
39 channel bandwidths is considered essential for the continued expansion of global mobile
40 communications. Sub-THz SAWs could provide the means to characterize materials at depths in
41 the single-digit nanometer range beneath the surface. They could be also applied to characterize
42 materials or molecules deposited between the SAW emitter and detector along the path of SAW
43 propagation. A primary objective of an ongoing research is to develop techniques for exciting and
44 detecting coherent SAWs with picosecond periods and deeply sub-optical, nanometer-scale
45 wavelengths and localization depths for applications in nanometrology, nanoimaging, as well as
46 in sensing and manipulations for both fundamental and applied research, and the field of ICTs.

47 From a theoretical perspective, the generation of SAWs at a specific frequency necessitates the
48 creation of a spatio-temporal stress distribution in a material at the SAW penetration depth during
49 excitation. The temporal Fourier spectrum of this distribution should include the desired frequency
50 component. Simultaneously, its spatial Fourier spectrum along the surface should encompass the
51 required SAW wavenumbers.¹⁴ The expansion of IDTs to sub-THz frequencies faces limitations
52 due to electronics bandwidth, unlike laser-based methods, which lack such restrictions. Intensity

53 envelopes of laser pulses lasting less than a picosecond contain frequencies up to the terahertz
54 range, effectively incorporating these frequencies into the spectrum of photo-induced stress.

55 Creating the required high wavenumbers in the spatial distribution of photo-induced stress can
56 be accomplished using laser gratings. This technique involves the interference between two
57 overlapping laser beams propagating at an angle, forming an intensity pattern with periodicity that
58 selects the SAW wavenumber.¹⁵⁻¹⁷ In this technique, the ratio of the wavevector of SAW to that
59 of electromagnetic radiation is less than or equal to 2.^{17,18} Notably, in laser methods, SAWs are
60 detected by observing the scattering of probe laser radiation due to SAW-induced changes in the
61 material refractive index and/or the surface topography. The highest-frequency Rayleigh SAW (50
62 GHz) was generated and detected using extreme ultraviolet (EUV) light grating with an 84 nm
63 period.¹⁸ Femtosecond pump pulses at a wavelength of 39.9 nm and a probe pulse at a wavelength
64 of 13.3 nm were derived from the FERMI free electron laser source. EUV radiation from the free
65 electron laser was also employed to induce gratings with a minimum period of 28 nm, although
66 this allowed monitoring of only the longitudinal acoustic waves propagating along the grating
67 direction.¹⁹ Due to the very high velocities of the longitudinal waves in the sample, the 28 nm
68 EUV gratings provided the capability to monitor longitudinal acoustic waves up to 360 GHz in
69 frequency.

70 As an alternative, the nanometer spatial periodicity of laser-induced stresses can be achieved
71 through surface nanostructuring. Historically, material nanopatterning has been accomplished
72 through depositing nanoscale arrays of metal lines (Figure 1a). In these cases, SAWs were
73 predominantly generated via the thermoelastic stress induced by laser heating of such grating.^{20,21}
74 Experiments with metallic gratings having a minimum period of around 40 nm were conducted
75 using 800 nm infrared pump laser pulses and 30 nm EUV probe laser pulses.²² The highest reported
76 coherent SAW frequency was approximately 90 GHz, achieved in 100 nm period metallic gratings
77 pumped and probed using near infrared light (at around 820 nm wavelengths),¹³ indicating
78 successful generation and detection with deeply sub-optical gratings.

79 However, for SAWs to reach 1 THz, the grating period controlling the wavenumber should be
80 shorter than 3 nm, assuming a typical Rayleigh SAW velocity of 3000 m/s in the material. The
81 minimum period of structures currently patterned in leading research laboratories is around 10 nm.
82²³⁻²⁵ Therefore, the primary limitation in generating SAWs at frequencies near 1 THz is the need
83 to reduce surface nanopatterning pitch while improving its quality. We propose that this limitation
84 can be addressed by engineering unconventional OAOTs based on cleaved nanostructured bulk
85 materials. Bulk superlattices (SLs) with a period of several atomic layers and atomic-scale quality
86 at the interfaces can currently be grown by epitaxy. These SLs can be cleaved or sliced (*i.e.*, with
87 microtome) along the surface normal to their layering, producing a nanopatterned surface, with
88 the periodicity and quality of bulk samples, that can control SAWs (Figure 1b). Figure 1c depicts
89 the consecutive steps of a cleaved SL attainment from a bulk SL grown on a substrate through
90 cleavage. While the highest frequencies of coherent bulk acoustic waves up to 3 THz have been
91 generated and detected by lasers in SLs/multiple quantum well structures,^{26,27} they have not yet
92 been applied to control SAWs. Previously, the cleaved SL structure was theoretically

93 investigated.²⁸ This structure is stratified normal to the cleaved surface and has physical parameters
 94 varying periodically not only in the bulk but also at its cleaved mechanically free surface. It was
 95 studied as a SAW phononic crystal, and an increased efficiency of the OA conversion in cleaved

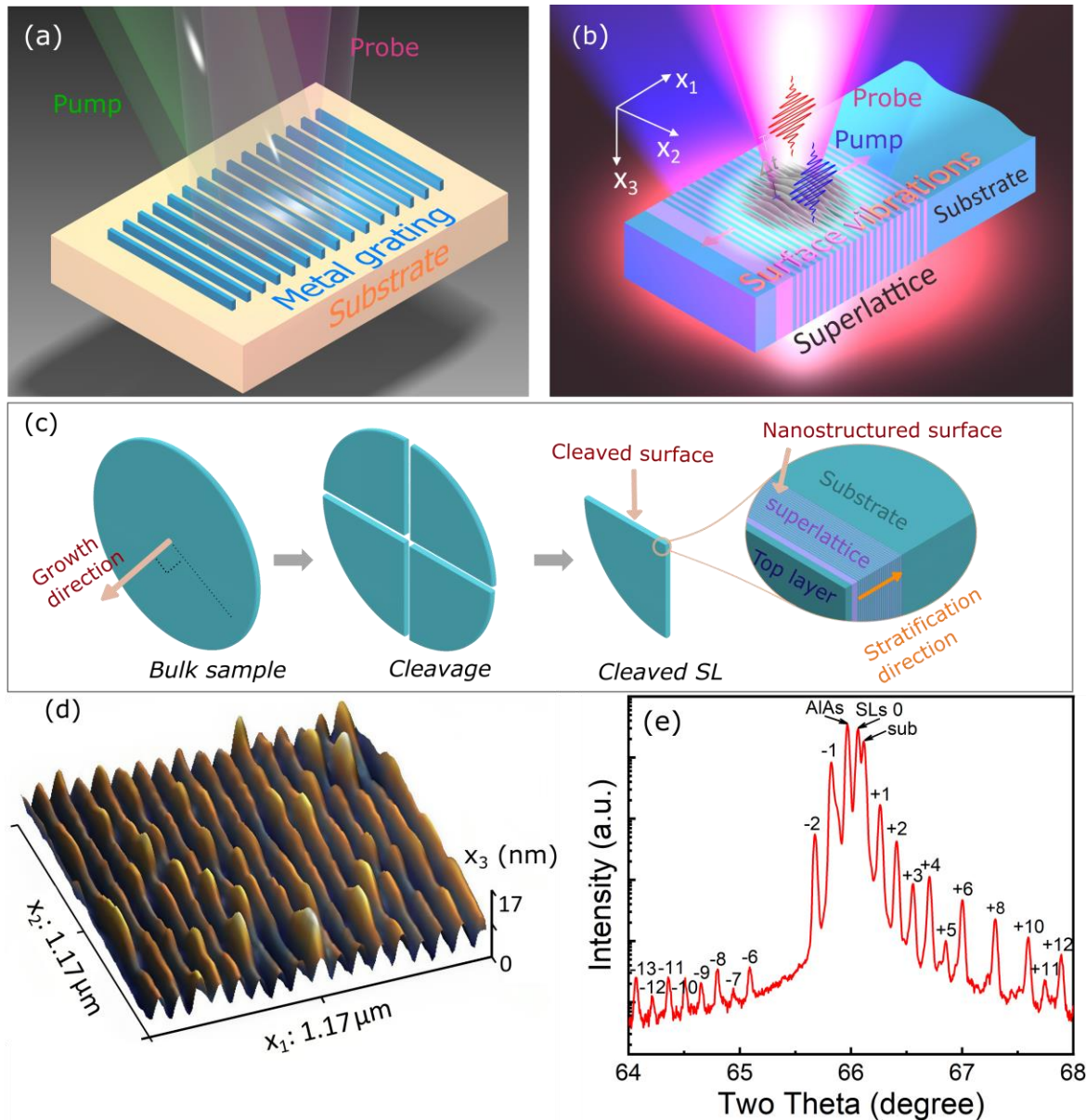


Figure 1. (a) An illustrative display of a metal grating structure. (b) Schematic of pump-probe experiments on the cleaved SL. (c) The illustration of the experimental sample preparation. GaAs substrate with the grown on it SL and cap GaAs layer is denoted as bulk sample. (d) AFM image of a cleaved GaAs/AlAs SL surface modified by oxidation. (e) X-ray diffraction measurement of a GaAs/AlAs superlattice.

96 semiconductor SL for monitoring GHz SAWs was suggested.^{29,30} Note that we will henceforth
 97 refer to the experimental SL structure with notation “SL stratified normal to the surface”, as
 98 suggested in the pioneering theoretical investigation.²⁸ While the theoretical investigations were
 99 conducted more than three decades ago, experimental demonstrations of acoustic waves on such

100 nanostructures have not yet been realized. Here, on the platform of cleaved semiconductor SLs,
101 we conducted proof-of-concept experiments for all-optical monitoring of coherent acoustic
102 vibrations of the surface (Figure 1b), encompassing various acoustic modes, including generalized
103 Rayleigh-type (gR) surface acoustic waves, surface-skimming transversal (T), and longitudinal
104 (L) acoustic waves, all of which are reported within this work. Two samples have been tested. The
105 first, an AlAs/GaAs SL with a 71 nm period, allowed us to simultaneously monitor the gR, T, and
106 L waves at frequencies of 40.2 GHz, 50.9 GHz, and 70.3 GHz, respectively, using near-infrared
107 femtosecond lasers. In the second sample, containing two Al_{0.2}Ga_{0.8}As/Al_{0.4}Ga_{0.6}As SLs of the
108 same period but with a modified content of Al in constituent layers compared to the first sample,
109 the gR wave at 40 GHz frequencies was also monitored.

110 RESULTS AND DISCUSSION

111 The nanostructures were prepared by epitaxial growth along the (001) direction of a semiconductor
112 SL on a GaAs substrate, layer by layer, with precise control of each layer composition (alternating
113 between Al_xGa_{1-x}As and Al_yGa_{1-y}As) and thickness (alternating between d_1 and d_2). This growth
114 resulted in a periodic stack of nanometers-thin layers stratified normal to the substrate surface.
115 Two semiconductor structures were investigated. They contain SLs with a nominally equal period
116 and individual layer thickness of $d_{\text{SL}} = 71.4$ nm, $d_1 = 40.0$ nm, $d_2 = 31.4$ nm, while the constituent
117 materials are GaAs/AlAs and Al_{0.2}Ga_{0.8}As/Al_{0.4}Ga_{0.6}As, respectively. The first sample contains a
118 single SL of 5.35 μm thickness, and the second one contains two SLs of 1.2 μm thickness separated
119 by a 1 μm -thick GaAs layer. Cleavage procedures were applied to the sample along the growth
120 direction, perpendicular to the substrate surface, producing a mirror-finished nanostructured
121 surface for optical monitoring of coherent acoustic phonons (Figure 1b).

122 However, oxidation of Al_xGa_{1-x}As ($x \neq 0$) layers exposed to air occurred. The morphology of
123 the nanostructured surface two months after cleavage was revealed by atomic force microscopy
124 (AFM, see Methods A), showing ripples with a periodicity of 71nm, extracted from a 2D FFT
125 analysis of the image. Apart from local defects, the average modulation depth of the oxidation
126 induced ripples is 7 nm in the GaAs/AlAs SL (see Figure 1d). Therefore, the sample with the
127 nanostructured surface (Figure 1c) was freshly cleaved just before the pump-probe experiments to
128 minimize oxidation effects, although complete prevention was not achieved.

129 It is worth noting that in the second sample, the Al content is sufficiently low in both SL layers
130 to significantly reduce the surface oxidation effect.³¹ The X-ray diffraction (XRD) results on the
131 GaAs/AlAs SL confirmed the good quality and periodicity of the structure (see Figure 1e and
132 Methods A). In contrast to a metal grating deposited on a substrate (see Figure 1a), the cleaved SL
133 structures (Figure 1b) acquire their nanopatterned surface through the nanostructuring of bulk SLs.
134 Common techniques such as lithography and focused ion beam are employed for nanostructuring
135 only the surface.

136 The periodicity of the mechanical/acoustical properties (such as density and elastic moduli) in
137 the cleaved SLs gives rise to Brillouin zone-center acoustic eigenmodes with corresponding lateral,
138 *i.e.*, along x_1 direction on the cleaved surface, periodicity of the mechanical strain/displacement

139 fields. These modes, for example, gR waves, contain lateral unmodulated/homogeneous
 140 components, which correspond to infinitely long modulation period (see Methods B). The same
 141 lateral periodicity of the optical properties, *i.e.*, of the complex optical refractive index,
 142 significantly enhances the possibilities for optically generating these acoustic eigenmodes (see
 143 Methods C). The identical periodicity of both optical and acousto-optical properties, *i.e.*, of the
 144 complex photoelastic tensor, facilitates the optical detection of these modes (see Methods D).

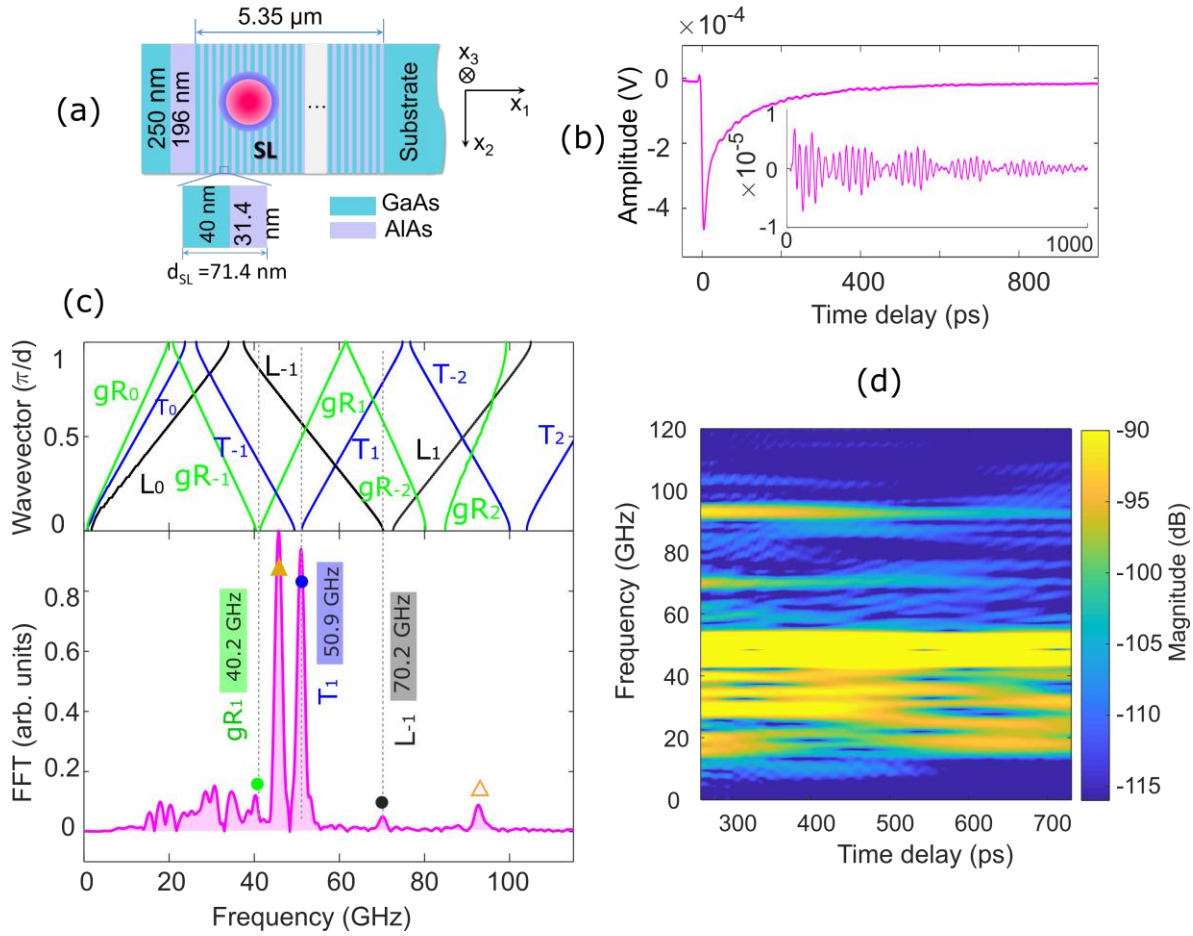


Figure 2. Presentation of the scheme and of evaluation results for the first sample. (a) Schematic depiction of the sample surface and of a potential laser focus position. (b) Time-domain reflectivity signal. Inset: acoustically-induced oscillations in the transient reflectivity signal after removing non-oscillating background. (c) Upper part: calculated dispersion relations of acoustic modes in the SL, which is infinitely thick along x_1 and semi-infinite along x_3 (green: gR, blue: T, black: L). Subscripts indicate acoustic band orders; Lower part: spectrum of the acoustic vibrations presented in the inset of (b). Green, blue and black solid dots indicate the identified first-order $k_1 = 0$ gR, T, and L waves, respectively. Brown solid and hollow triangles represent the frequencies of bulk longitudinal acoustic waves propagating in the zeroth (45.7 GHz) and first (92.6 GHz) diffraction orders, respectively. (d) Spectrogram of the acoustic signal shown in the inset of part (b) obtained by short-time Fourier transform (see Methods F for details).

145 It is important to note that the existence of laterally unmodulated and homogeneous eigenmode
 146 components is theoretically advantageous for their all-optical monitoring. High-frequency acoustic

147 modes at the zone center can be excited even by the laterally homogeneous part of the distributed
 148 photo-induced stresses acting on laterally homogeneous components in the mode structure. These
 149 modes can be detected through time-domain backward Brillouin scattering of the normally
 150 incident probe light, even when the SL motion is laterally unmodulated, meaning that scattering
 151 occurs from the motion of the SL averaged over its period (see Methods C and D). Due to the short
 152 periodicity in their physical properties, the designed SLs stratified normal to the surface (x_1x_2 plane
 153 in Figure 1b)²⁸ offer the opportunity to optically monitor surface acoustic vibrations with deeply
 154 sub-optical lateral periodicity, since they are composed of the acoustic waves with deeply sub-
 155 optical lengths.

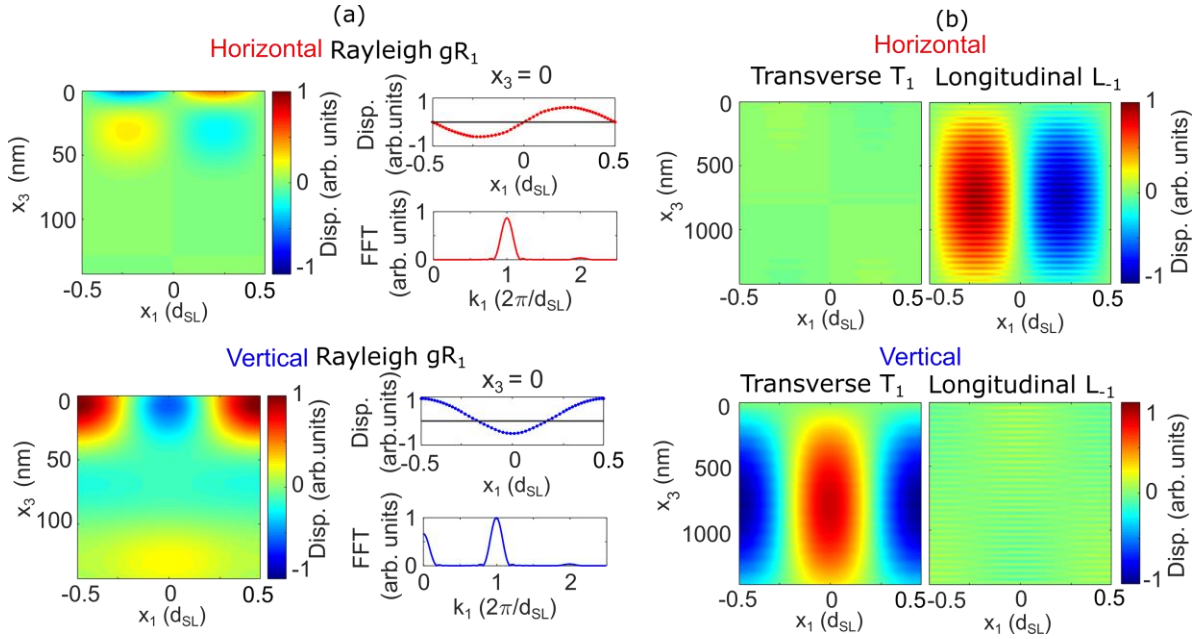


Figure 3. Calculated spatial distributions of the mechanical displacement components in the first-order zone center $k_1 = 0$ symmetric (a) gR (41.4 GHz), (b) T (51.2 GHz) and L (70.3 GHz) acoustic modes. Disp. denotes displacement. The second column in (a) presents the displacements at mechanically free surface and their k_1 spectra, highlighting the presence of the spatially homogeneous component in the gR mode structure.

156 We conducted ultrafast pump-probe transient reflectivity experiments to monitor coherent
 157 acoustic phonons on the cleaved SL surface of the prepared samples (see Figure 1b and Methods
 158 A). Unlike the conventional setup for monitoring bulk acoustic waves in SLs,²⁶ where the pump
 159 and the probe laser beams are incident on the plane x_2x_3 , in this case, they are directed normally to
 160 the cleaved plane x_1x_2 . Laser wavelengths of 405 nm/810 nm were chosen for the pump and probe
 161 laser beams, which were collinearly co-focused by a 100 \times objective onto the sample, creating a
 162 spot with a radius of approximately 0.82 μm (measured at $1/e^2$ by cross-correlation, see Methods
 163 E for details). We monitored the time-dependent reflectivity change of the sample, induced by the
 164 pump excitation on the cleaved structure, using the probe light whose intensity was modulated by
 165 this change, and we captured it with a photodetector.

166 The experimental results obtained from both SL-based structures are now presented and
 167 compared favourably with theoretical expectations. The proof-of-concept experiment was
 168 conducted on the first sample, which was based on the cleaved 5.35 μm -thick GaAs/AlAs SL. In
 169 this configuration, the laser beams were focused entirely on the SL region (see Figure 2a). The
 170 interband absorption of the pump laser pulses in the GaAs creates electron-hole pairs, whose
 171 distribution dominantly contributes to the generation of acoustic waves through the stress induced
 172 by the electron-hole-phonon deformation potential mechanism.³² Notably, this opto-acoustic
 173 conversion mechanism is much more efficient than the thermo-elastic one, most commonly found
 174 in metallic grating OA transducers.^{13,14} The reflectivity change induced by the acoustic waves
 175 (phonons) is monitored by the probe near infrared light as a function of the time-delay between
 176 the pump and the probe pulses.

177 The time-domain signal (Figure 2b, main part) reveals that the acoustically-induced oscillations
 178 (Figure 2b, inset) are superimposed on the monotonous exponential variations in amplitude caused
 179 by the generation/recombination of the electron-hole pairs and transient heating of the sample after
 180 photo-excitation. To enhance the display of acoustic oscillations (Figure 2b, inset), the dominant
 181 background is numerically removed, following the signal processing details in Methods F. Due to
 182 the SL periodicity, the projections k_1 of the wave vectors on x_1 axis, which are larger than π/d_{SL} in
 183 the acoustic modes dispersion relations, are folded into a mini-Brillouin-zone (see Figure 2c, upper
 184 section, and Methods G for details on numerical calculations). In this zone, the gR, T, and L waves
 185 differ in frequencies due to the differences in their propagation velocities. The gR waves become
 186 supersonic above the frequency corresponding to the intersection of the first order optical surface
 187 phonon branch R_{-1} and the bulk acoustical phonon branch T_0 , leading to attenuation through the
 188 emission of bulk acoustic waves (Brekhovskich attenuation).³³

189 However, due to the small relative differences between the parameters of the SL constituent
 190 materials, $\mu \ll 1$, where μ represents the ratios of parameter differences to average parameter
 191 values, such as $\Delta C_{11}/\langle C_{11} \rangle = 0.0117$, $\Delta C_{12}/\langle C_{12} \rangle = 0.058$, $\Delta C_{44}/\langle C_{44} \rangle = 0.00845$ and $\Delta\rho/\langle\rho\rangle =$
 192 0.3368 for GaAs/AlAs, a basic analysis (see Supporting Information S1) demonstrates that the $k =$
 193 0 gR wave experiences a slight frequency shift and a weak attenuation proportional to μ^2
 194 ($\Delta\text{Re}(\omega_R)/\omega_R \sim \mu^2$, $\text{Im}(\omega_R)/\omega_R \sim \mu^2$, with ω denoting the cyclic frequency). The numerical
 195 simulations of the eigenmode dispersion relations in the laterally-infinite cleaved SL shown in the
 196 upper panel of Figure 2c, enable the identification of three peaks. These peaks, found in the
 197 experimental spectrum in the lower section of Figure 2c at frequencies 40.2 GHz, 50.9 GHz and
 198 70.2 GHz, correspond to the zone center vibrational motions induced by the gR, T, L waves,
 199 respectively. Figure 2d depicts the spectrogram of the acoustic signal in Figure 2b and shows how
 200 the frequency contents of the signal vary with time delay. This will be further discussed later on.

201 Figure 3 illustrates the calculated shapes of these three modes, revealing that the vibrations
 202 caused by the gR wave (Figure 3a) are highly localized near the surface (at depths smaller than
 203 one SL period). In contrast, the vibrations induced by the T and L waves (Figure 3b) are non-
 204 localized. To better appreciate the surface displacement at $x_3 = 0$ induced by the gR wave, the
 205 horizontal (top) and vertical (bottom) displacement are displayed in the second column of Figure

206 3a as a function of x_1 . It is important to note here that both components depict spatial periodicity
 207 associated with the SL periodicity d_{SL} . More importantly, the vertical component, unlike the
 208 horizontal one, contains a non-zero mean value as seen from the non-zero surface area and, related
 209 non-zero Fourier component at $k_1 = 0$ of its displacement distribution. The differences in amplitude

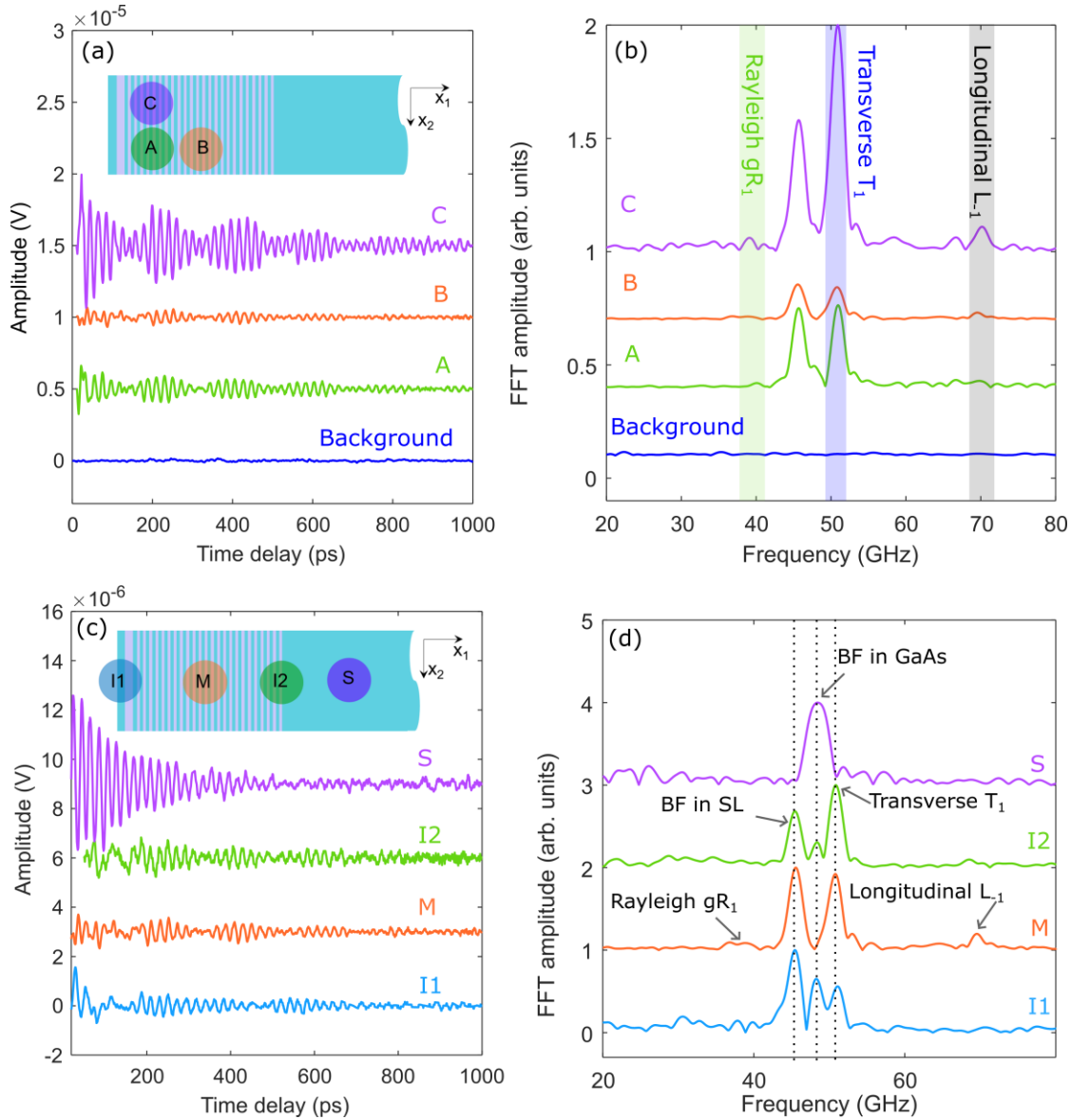


Figure 4. Experimental measurements on the first sample. (a) Detection of acoustic vibrations and (b) their spectra obtained by overlapping the pump and probe foci at positions A, B, C on the SL. These results are compared with background measurements (zero pump light). In (b) the spectral components attributed to the gR , T , and L waves are highlighted in color. (c) Detection of acoustic vibrations and (d) their spectra achieved by overlapping pump/probe foci at positions I1, M, I2, S on top layer/SL interface, SL, SL/GaAs interface, and substrate, respectively. The three spectral peaks indicated by vertical dotted lines correspond to the Brillouin frequency (BF) in the SL (45 GHz, left), the BF in GaAs (48 GHz, middle), and the frequency of the T wave in the SL (51 GHz, right), respectively.

210 of the related transient reflectivity oscillations between the gR and T waves at closely matched
211 frequencies can likely be attributed to the oxidation of AlAs layers and the imperfect cleavage
212 procedures, which introduce surface defects and roughness (as shown in Figure 1d). These factors
213 are expected to exert a more significant influence on the generation, propagation, and detection of
214 surface-localized acoustic eigenmodes compared to their delocalized counterparts.

215 The two additional peaks present in the experimental spectrum in Figure 2b at frequencies of
216 45.7 GHz and 92.6 GHz can be attributed to the Brillouin frequencies induced by the probe light
217 that is backward scattered by the coherent bulk acoustic modes launched within the bulk (*i.e.*,
218 depth) of the SL nanostructure by the pump laser radiation. This is detailed in Methods H where
219 we derive the analytical estimates of Brillouin frequencies for light scattering by quasi-longitudinal
220 acoustic (QLA) waves propagating in the diffraction orders $n = 0$ and $n = 1$, respectively.

221 Several spectral components below 40 GHz, not consistently replicable in the experiments, are
222 likely attributable to random noise originating from the optical system or electronic devices.
223 However, it is not within the scope of this discussion to delve further into those frequencies due to
224 their low repeatability.

225 To assess the reliability and variations in the monitored signals, we adjusted the laser foci
226 laterally and vertically on the cleaved surface. We conducted these adjustments at positions labeled
227 A, B and C within the SL region (as shown in Figure 4a and Figure 4b). Here, we aim to replicate
228 the vibrations induced by the gR, T, and L waves at their anticipated frequencies. However, it is
229 noteworthy that the amplitudes of these vibrations exhibit variations based on the specific positions
230 due to the inherent inhomogeneities within the SL material, as depicted in Figure 1d. Furthermore,
231 these results are compared with the background measurements, and these comparisons indicate
232 that the noise level, approximately at 10^{-7} , remains consistently below the level of the identified
233 folded acoustic phonons.

234 It is also enlightening to shift the lateral position of the laser foci across different regions,
235 encompassing the top layer/SL interface (I1), the SL itself (M), the SL/GaAs interface (I2), and
236 the substrate (S) (refer to Figure 4c and 4d). By demarcating three prominent peaks with vertical
237 dotted lines, we uncover that the middle frequency at 48 GHz is notably absent when probing at
238 position M within the SL. However, this frequency is present at interfaces I1 and I2, and
239 corresponds to the Brillouin frequency detected in the GaAs substrate at position S. This
240 observation affirms our expectations that the Brillouin frequency measured within the SL does not
241 coincide with that in the GaAs substrate. The Brillouin frequency at 45 GHz (represented by the
242 left dotted line) detected within the SL region exhibits a red-shift relative to the Brillouin frequency
243 in the GaAs, primarily due to the SL lower refractive index compared to the bulk GaAs when
244 considering the probe light wavelength. The frequency at 51 GHz (as indicated by the right dotted
245 line) is detected at all positions except the substrate, hinting at its origin as a folded acoustic mode
246 arising from the nanostructure periodicity. Additionally, other folded coherent acoustic modes are
247 partially detected at the interfaces (as seen in the comparison between Figure 4b and Figure 4d).

248 Finally, we investigated the excitation and detection of surface modes on the second sample. A
249 lateral scan of overlapping pump and probe laser beams was conducted on the sample containing

250 the two $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}/\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ SLs (Figure 5a). It is worth noting that the individual thicknesses
 251 of SL₁, SL₂, and the gap are smaller than the focused beam spot. Consequently, the light is always
 252 partially absorbed by GaAs when directed at the SLs.

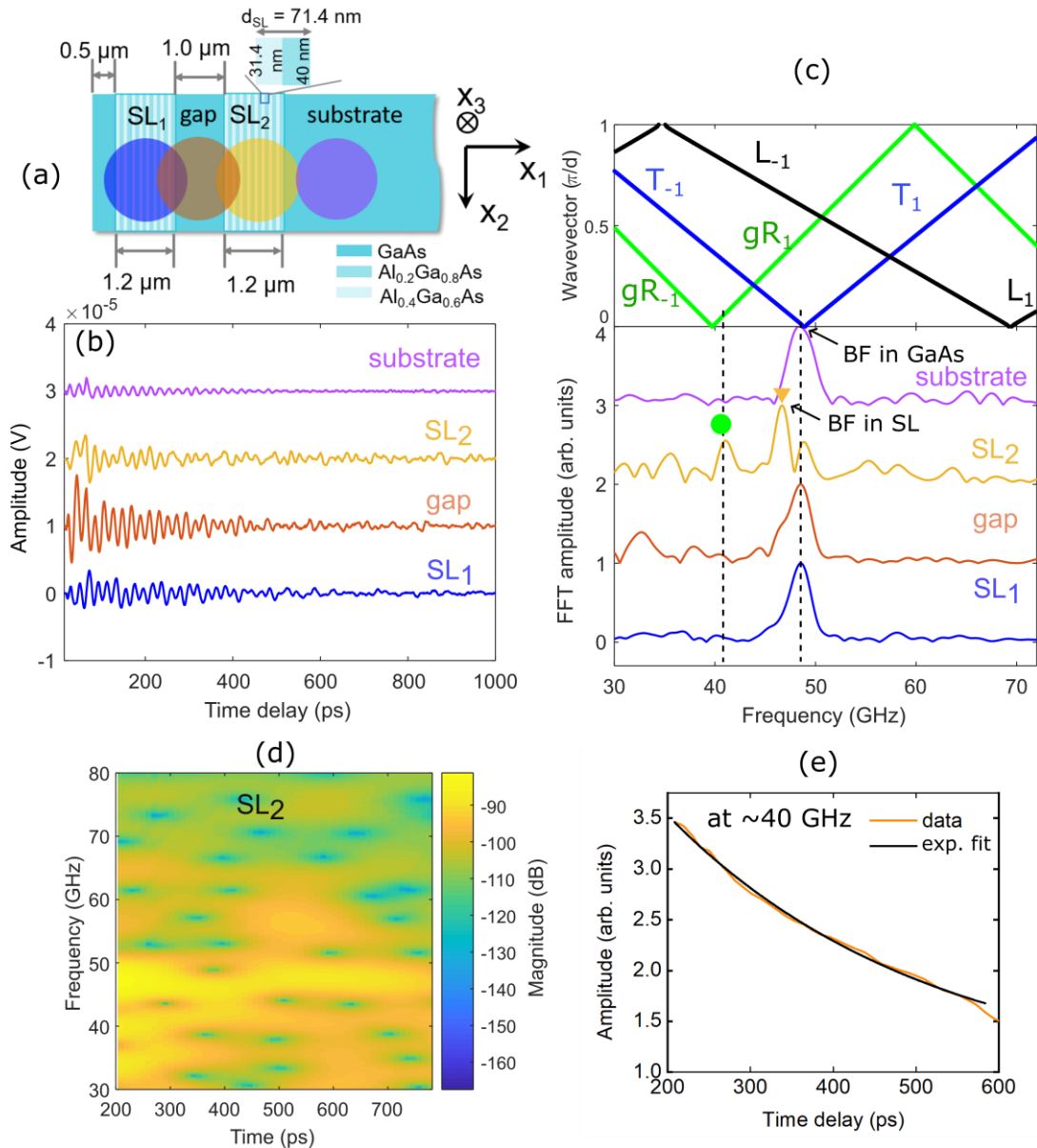


Figure 5. Presentation of the scheme and of the evaluation results for the second sample. (a) Schematic depiction of the sample surface and of the laser foci positions tested on SL₁, gap, SL₂ and substrate. (b) Contributions to transient reflectivity signal induced by acoustic vibrations. (c) Upper part: computed dispersion relations of acoustic modes in the SLs, considered to be laterally infinite along x_1 and semi-infinite along x_3 (green: R, blue: T, black: L); Lower part: spectra of acoustic vibrations shown in (b). (d) Spectrogram of the acoustic signal on SL₂ shown in part (b) obtained by short-time Fourier transform (see Methods F for details). (e) Attenuation of 40 GHz mode as a function of the time delay, extracted from (d).

253 The spectral peak at 48.5 GHz is identified as the Brillouin frequency in the GaAs substrate
254 when the laser foci do not overlap with the SL (purple spot in Figure 5a). However, at the three
255 other positions, a potential contribution to this spectral peak from the T wave, theoretically
256 predicted at 49 GHz (see the upper part of Figure 5c) cannot be ruled out.

257 In SL₂, we monitor the folded first-order $k_1 = 0$ gR wave frequency at 41 GHz (indicated by the
258 green solid dot) along with the Brillouin frequency at 46.6 GHz. The absence of these frequency
259 components on the SL₁ can be attributed to a non-ideal surface quality or SL quality at the sample
260 edge (there is only 500 nm distance between the top layer/air interface and SL₁ left edge).

261 The peak at about 40 GHz, attributed to the gR mode, exhibits an exponential decay with a
262 lifetime τ_S of approximately 300 ps, obtained from the exponential fit, shown in Figure 5e, of the
263 amplitude attenuation with time delay at 40 GHz extracted from the spectrogram in Figure 5d.
264 This gR wave lifetime is comparable to that shown in Figure 2d. The propagation velocity of the
265 gR wave can be estimated as $v_R \equiv d_{SL}\omega_R/2\pi \cong 2930$ m/s. The escape time τ_a of the gR wave
266 from the region where it is monitored, defined by the laser foci, can be estimated as $\tau_a \equiv 2a/v_R \approx$
267 550 ps. Therefore, it is not just the escape time that limits the observed lifetime τ_S of the gR wave
268 signal. The gR wave lifetime τ_R , potentially limited by gR wave diffraction, its absorption in the
269 bulk of the materials due to inelastic processes, its scattering by surface roughness, and emission
270 of the bulk acoustic waves, can be estimated from the relation $\tau_S^{-1} = \tau_a^{-1} + \tau_R^{-1}$, as $\tau_R \approx 660$ ps.
271 The diffraction time of the gR wave, τ_D , can be estimated as $\tau_D \sim (2a)^2/d_{SL}v_R \approx 12.6$ ns. The
272 time of the longitudinal acoustic wave absorption in the bulk of the GaAs at 56 GHz frequency
273 exceeds 2 ns.³⁴ In our experiments, the duration of the beatings in acoustically-induced transient
274 reflectivity signals (Figure 2b and Figure 4a,c), resulting from the superposition of signals of
275 comparable amplitudes due to bulk longitudinal acoustic wave and T waves, demonstrates that the
276 lifetimes of the shear acoustic waves at ~ 50 GHz are, at least ~ 1 ns. Hence, it is expected that
277 neither the diffraction nor the bulk absorption limits the gR wave lifetime in the experiments
278 reported here.

279 This observation suggests that the gR wave lifetime could be further increased by either reducing
280 the gR wave attenuation caused by surface roughness through improved surface quality (*e.g.*, by
281 depositing an oxide nanolayer to protect the surface from oxidation) or by minimizing gR wave
282 energy emission to bulk modes through the design of an optimized SL structure and composition.

283 In the second sample, there is no apparent monitoring of L wave, which could be attributed to
284 the reduced contrast in acoustic properties between the constituent layers of
285 Al_{0.2}Ga_{0.8}As/Al_{0.4}Ga_{0.6}As SLs in the second sample in comparison to the GaAs/AlAs SL of the
286 first sample. Alternatively, other factors related to sample fabrications, currently not fully
287 understood, might contribute to this observation.

288 In summary, the transient reflectivity measurements from lateral scanning on the surfaces of
289 both samples can be employed to unveil defects and the nanostructure geometry (including the
290 interfaces of the SL with the surrounding materials) on micrometer scale through the evaluation
291 of acoustic surface vibrations and bulk acoustic eigenmodes. Simultaneously, the monitoring of
292 the gR wave on both samples strengthens the expectations that, in the near future, dedicated studies

293 and optimized designs of the SL-based SAW nanotransducers will enhance the characteristics of
294 the reported gR waves.

295 It is important to highlight that the signals from monitored T waves are notably prevalent in all
296 our experiments. The following discussion clarifies why this observation has been somewhat
297 unexpected. All forms of bulk acoustic waves, including quasi-longitudinal and quasi-transverse,
298 can be detected using laser-induced gratings in transparent or weakly absorbing materials.^{35,36} The
299 detection of L waves was previously reported in multiple experiments utilizing visible^{11,36} and UV
300 laser-induced gratings,¹⁹ as well as deposited metallic gratings.^{37,38} Concomitantly, these gratings
301 modulate (structure) the near-surface properties of the sample and modulate (ripple) the surface
302 profile. However, there are relatively few reports³⁸ on T waves detection through laser-induced
303 gratings in opaque materials. Additionally, we are not aware of any demonstrations of T wave
304 generation and detection assisted by deposited metallic gratings.

305 A potential explanation for the absence of T waves in experiments with elastically isotropic
306 gratings on an elastically isotropic substrate was previously provided.³⁹ In such elastically isotropic
307 media, the most common laser-induced stresses, *i.e.*, thermoelastic stresses, typically do not
308 generate shear (transverse) acoustic waves within the bulk of the sample.³⁸ Theoretically, only the
309 transverse mode that is mode-converted from the longitudinal mode, either incident at the surface
310 or propagating along the surface, could contribute to shear surface displacement. Based on
311 accumulated experimental^{37,40} and theoretical^{39,41} knowledge, it is anticipated that the structures in
312 which either the substrate, the grating, or both exhibit elastic anisotropy, are better suited for
313 controlling T waves. In the absence of elastic anisotropy, gR and L waves are expected to
314 significantly dominate the acoustic signals in transient reflectivity measurements. This rationale
315 aligns with our experimental demonstrations on cleaved SL structures composed of elastically
316 anisotropic constituents. In our experiments, we have successfully accessed both gR and T waves,
317 which feature extremely short but equal spatial periodicity. This characteristic results in a
318 significant separation of their frequencies, even when their propagation velocities are closely
319 matched. This platform provides promising opportunities to generate and detect T waves, thus
320 avoiding potential signal interference from the presence of the gR wave. For instance, in isotropic
321 media, measuring the velocity of T waves provides information on the shear rigidity of the
322 material, a property entirely absent in L waves and mixed with information on the longitudinal
323 modulus in the gR wave.^{14,42} Unlike L waves, T waves preferentially couple with rotational
324 eigenmodes in materials with complex elasticity.^{43,44} In a non-exhaustive list of other advantageous
325 applications of T waves, one can find the determination of residual stress,⁴⁵ inspection of material
326 cracks,^{46,47} and evaluation of the dynamics of structural phase transitions.³⁶

327 CONCLUSIONS

328 We have successfully showcased the all-optical monitoring of the acoustic vibrations on the
329 samples with periodically structured surface, prepared by cleavage of the SLs along their growth
330 direction. In these structures the periodicity of the material parameters at cleaved mechanically
331 free surface is due to their periodicity in the bulk. These samples, composed of nanometer pitch

332 normally-cleaved $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{Al}_y\text{Ga}_{1-y}\text{As}$ SLs, were examined in pump-probe experiments
333 utilizing visible (blue) pump and near-infrared probe laser beams from an ultrafast laser. These
334 semiconductor SLs featured different $\text{Al}_x\text{Ga}_{1-x}\text{As}$ constituent layers and a period of 71 nm. We
335 observed and studied folded acoustic phonons, including gR, T, and L waves, within the frequency
336 range of 40 GHz to 70 GHz.

337 In this tested platform for studying surface acoustic motion at GHz frequencies, T waves
338 dominate over gR waves and L waves. This dominance is tentatively attributed to the elastic
339 anisotropy of the SL. It is worth noting that this may be related to the specific orientation of the
340 SL layering relative to the nanostructure surface, which appears to favor the excitation of T waves
341 by the gradients of the photo-induced stresses. However, it is essential to emphasize that this
342 hypothesis would require further theoretical development, which is beyond the scope of our current
343 presentation, primarily focused on experimental results.

344 Our platform provides opportunity to monitor T waves and gR waves independently due to the
345 well-separated frequencies by tens of GHz in the nanotransducer. Our proof-of-concept
346 experiments revealed the importance of phenomena such as the oxidation of the freshly cleaved
347 surface, the surface roughness and the non-negligible smallness of the elastic contrast between the
348 layers on the efficiency of SAWs monitoring by laser on the suggested platform. By optimizing
349 these factors along with diminishing the SL period, our platform holds practical opportunities to
350 break the current limitation of SAW frequency, extending it to sub-THz range.

351 Our experimental findings provide a viable solution for monitoring surface acoustic vibrations
352 with GHz frequencies. This approach serves as a valuable alternative and supplement to
353 established techniques involving metal grating and EUV light grating methods. Unfortunately, our
354 initial trial experiments on the same nanostructured samples, aimed at demonstrating the
355 propagation of the gR wave packets between two spatially separated points on the sample surface
356 by focusing the pump and probe laser beams on these points, expectedly failed. The limited length
357 of gR wave propagation on our SLs, estimated to be approximately $l_R \approx 1.9 \mu\text{m}$ based on the
358 estimated gR wave lifetime and velocity, is comparable to the diameter of the laser beams (≈ 1.62
359 μm). These laser beams dictate the lengths of the monitored gR wave packet. Consequently, the
360 pump and probe laser beams essentially overlap, even when the centers of the pump and probe
361 laser beams are separated by the gR wave propagation distance. Moreover, the gR wave
362 propagation time between the focal points is similar to its duration. In simpler terms, the signal
363 increase resulting from the forward movement of the gR wave packet, generated in the pump focus
364 region, toward the detection area controlled by the spatially shifted probe focus, is significantly
365 suppressed or saturated due to the attenuation of gR wave packet happening on the same time
366 scale. This led to gR wave signals falling below the noise level.

367 Several potential approaches exist for future monitoring of gR wave packets. Firstly, it is
368 desirable to increase the propagation lengths of gR waves. This can be accomplished by reducing
369 surface roughness and preventing surface oxidation. To mitigate oxidation, a protective oxide
370 nanolayer can be applied to the surface immediately after cleaving the SL, or the SLs can be grown
371 from oxide material components. Secondly, shortening the length/duration of the monitored gR

372 wave packets is desirable. This can be achieved by employing stronger laser focusing or by
373 reducing the thickness of the SL to be smaller than the diameter of the laser foci.

374 Furthermore, it is crucial to combine these approaches with the optimal selection of materials
375 for the SL layers. Material optimization entails enhancing optoacoustic and acousto-optic
376 transformation efficiencies at specific wavelengths of the pump and probe lasers. By improving
377 these efficiencies, the amplitude of the signal resulting from coherent gR waves would increase,
378 potentially elevating gR wave amplitudes above the noise level at micrometer propagation
379 distances.

380 Sub-THz SAWs are potentially promising for future applications in science and technology,
381 such as the next generation of ICT devices well beyond the 5G standard (600 MHz - 6 GHz). The
382 recent demonstration of picosecond acoustics based on miniaturized lasers¹⁶ represents a
383 significant step forward in applying laser-monitored SAWs to future ICT. A detailed
384 understanding of the propagation of such SAWs on atomically flat surfaces and across material
385 junctions is a prerequisite for accessing the fundamental concepts of thermal load management.
386 With the diminishing periodicity of the SL, the platform of the cleaved SLs presented herein also
387 holds considerable potential for revealing and exploring strong acousto-optic and opto-acoustic
388 interactions due to the simultaneous confinement of acoustic and electromagnetic modes.

389 **METHODS**

390 **A. Sample preparation and characterization by X-ray diffraction and AFM**

391 The SL samples were grown using solid-source molecular beam epitaxy (MBE) with a Veeco
392 GenXplor system. The $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloy was achieved through a digital alloy growth technique,
393 where the composition is determined by the layer thicknesses of GaAs and AlAs. The samples
394 were grown under typical conditions for GaAs and AlAs, maintaining an As-rich environment at
395 580 °C. The growth rates were approximately 1 ML/sec for both GaAs and AlAs. High-resolution
396 XRD scans on the (004) plane were conducted using a Bruker D8 system. AFM measurements
397 were performed with a 5500 AFM from Agilent® in intermittent contact mode in air (cantilever
398 stiffness was 40 N/m). Image processing utilized Gwyddion® freeware.

399 **B. Structure of the generalized Rayleigh SAWs in cleaved SLs**

400 For the physical interpretation of the demonstrated all-optical monitoring of the Rayleigh-type
401 SAW (R) wave on the SL stratified normal to the surface, it is important to emphasize the key
402 differences in the mode structure of this gR wave compared to the Rayleigh SAW (RSAW) on the
403 surface of a homogeneous half-space. As explained by the Bloch-Floquet theorem,⁴⁸ there is an
404 infinite number of the bulk longitudinal and transverse wave eigenmodes in the SL presented in
405 Figure 2c. This is attributed to the periodicity of SL parameters, characterized by the spatial period
406 d_{SL} or by the SL (Floquet) wave number $q_{SL} = 2\pi/d_{SL}$ along the x_1 axis (the direction of SL
407 layering).

408 The m -th order bulk eigenmodes can be viewed as the diffraction orders of the bulk modes with
409 frequency ω and the wave vector \vec{k} (with $k_2 = 0$ for the waves polarized in the sagittal plane
410 (x_1, x_3)), which differ by the projections of k_1 ($k_1 + mq_{SL}, m = 0, \pm 1, \pm 2, \dots$) on the x_1 -axis.

411 Crucially, the displacement components with different m are parametrically coupled due to the
 412 periodicity of the acoustical properties in the SL. Consequently, each eigenmodes, numbered as
 413 the m -th mode, contains components with all allowed $m = 0, \pm 1, \pm 2, \dots$, including, in
 414 general, $m = 0$, although with different amplitudes.

415 The gR wave results from the superposition of the bulk modes, evanescent in the depth direction
 416 x_3 (*i.e.*, with k_3 describing amplitude decay with depth) and satisfying periodic stress-free
 417 boundary condition on $x_3 = 0$ at a particular gR wave eigenfrequency, ω_{GR} . It contains
 418 displacement components with all $k_1 + mq_{SL}$, $m = 0, \pm 1, \pm 2, \dots$ (see Supporting Information S1
 419 for details). Similar to the dispersion relations for bulk acoustic waves, the gR wave dispersion
 420 relation can be folded in the Brillouin zone with the acoustic phonon branches, starting from $\omega =$
 421 0, and the optical phonons branches^{28,49}. The important difference of the gR wave from RSAW is
 422 that the former contains, in the zone-center $k_1 = 0$, laterally non-modulated displacement and
 423 strain components at the frequencies of optical-type SAWs.

424 **C. Photo-generation of the coherent acoustic eigen modes in cleaved SLs**

425 In our experiments, the phonons in the close vicinity of the Brillouin zone center play the dominant
 426 role, *i.e.*, with $k_1 \ll q_{SL} \equiv 2\pi/d_{SL}$, as evident from the fact that the radii of pump and probe laser
 427 beams foci on the SL surface, $a_{\text{pump}} \approx a_{\text{probe}} \approx a$, significantly exceed the SL period d_{SL} . The
 428 \vec{k} -spectrum of the photo-generated SAWs is controlled by the lateral distribution of the stresses,
 429 photo-induced by the pump laser pulses (see Supporting Information S1 for the details). In our
 430 experiments, the lateral modulation of the photo-induced stresses on the nanometer spatial scale is
 431 mostly due to the difference in the optical properties of the two layers.

432 In the infinite SL, the stresses photo-induced by laterally homogeneous pump laser radiation will
 433 have the periodicity of the SL and exhibit the Bloch-Floquet property. Thus, the folded k_1 -
 434 spectrum of the photo-induced stresses would contain only zone center components, $k_1 = 0$.
 435 Focusing the pump laser radiation in a spot with radius a localizes laterally the photo-induced
 436 stresses on the scale $a \gg d_{SL}$, even in the case of a diffraction-limited focusing, because of the
 437 sub-optical periodicity of the SL, *i.e.*, $d_{SL}/\lambda \ll 1$. Therefore, the related broadening of the k_1 -
 438 spectrum components is much narrower than the width of the Brillouin zone π/d_{SL} and the photo-
 439 generated gR wave spectrum is strongly localized near the Brillouin zone center.

440 The condition $a \gg d_{SL}$ also leads to the expectations that symmetric motions of individual SL
 441 layers are photo-excited with larger amplitudes than antisymmetric ones. Here, we denote
 442 symmetric modes as modes that exhibits an odd distribution of the horizontal mechanical
 443 displacement, u_1 , and an even distribution of vertical mechanical displacement, u_3 , relative to the
 444 central plane of the individual layer (see Supporting Information S1 for the details).

445 For the experimental platform of a semi-infinite SL stratified normal to the surface, it is crucial
 446 that the zone-center surface confined modes contain laterally homogeneous components in their
 447 structure. This implies that gR modes can be excited by a laterally homogeneous (averaged over
 448 the SL period) part of the distributed photo-induced stresses or, in principle, even by laterally non-
 449 modulated stresses in the hypothetical case of the absence of optical but existing acoustical contrast
 450 between the SL composing layers.

451 Under the condition of weak acoustical contrast, $\Delta\rho/\langle\rho\rangle \sim \Delta C_{ij}/\langle C_{ij}\rangle \sim \mu \ll 1$, which is
 452 sufficiently well satisfied in our experimental samples, the amplitude ratio of the non-modulated
 453 to modulated zone-center gR wave components scales with $\sim\mu$, the frequency of the lowest zone
 454 center optical-type gR wave, ω_{GR} , deviates from the respective frequency ω_R of RSAW with the
 455 wave vector $k_1 = 2\pi/d_{SL}$ in the medium of the period-averaged parameters, $\text{Re}(\omega_{GR} -$
 456 $\omega_R)/\omega_R \sim \mu^2$, and it acquires an imaginary part $\text{Im}(\omega_{GR})/\omega_R \sim \mu^2$. Thus, the zone center gR waves
 457 are actually pseudo-SAWs^{50,51}.

458 **D. Photo-detection of the coherent acoustic eigenmodes in cleaved SLs**

459 The laterally homogeneous components of the high frequency acoustic eigenmodes in cleaved SLs
 460 can be detected by probe laser radiation, as established in picosecond laser ultrasonics of depth-
 461 stratified samples/structures.⁵² The theoretical analysis of the detection paths of gR wave, when it
 462 is propagating in the SL (identical to the one where it was photo-generated), reveals that the
 463 laterally modulated components of the gR wave can also be efficiently detected due to the lateral
 464 periodicity in the SL of the optical permittivity ε and/or the photoelastic parameter p (see
 465 Supporting Information S2 for the details).

466 The essence of the acoustic wave detection in the ultrafast pump-probe optical experiments (via
 467 measurements of the transient reflectivity at normal incidence of probe laser light) is the
 468 heterodyning of the weak acoustically-scattered probe light by the strong probe light reflected from
 469 the surface of the SL⁵³. Only the probe light reflected in the $m = 0$ diffraction order reaches the
 470 photodetector because the light reflected in the other diffraction orders is evanescent due to sub-
 471 optical periodicity of the considered cleaved SLs. Consequently, only the probe light scattered by
 472 the acoustic wave in the zeroth diffraction order is detectable.

473 An additional consequence of the sub-optical periodicity of the SL is that each diffraction of
 474 light by the modulated components of the SL parameters acquires a multiplier $(d_{SL}/\lambda)^2 \ll 1$ in
 475 the amplitude of the diffracted light. Because of this, the non-evanescent probe light that can
 476 propagate in the direction of the photodetector (in the zeroth diffraction order) after exhibiting
 477 several consecutive diffractions (in reflection and/or scattering) is negligibly small in amplitude
 478 compared to the non-diffracted probe light.

479 Only the scattering of the non-diffracted probe light by the acoustically-induced modulation of
 480 the period-averaged optical permittivity $\langle\varepsilon\rangle$ of the SL should be considered. However, this laterally
 481 homogeneous modulation is caused not only by the modulation of the averaged permittivity by the
 482 laterally non-modulated gR wave component (first path) but also by the demodulation of the
 483 laterally modulated components of the gR wave when it interacts with SL parameters, exhibiting
 484 the same periodicity as the gR wave (second path). From the viewpoint of formal mathematics,
 485 the second path is due to the presence of the constant/infinite-period term in the square of the
 486 sine/cosine functions. In particular, the second path of the possible detection can be due to $\varepsilon^{(1)} -$
 487 $\varepsilon^{(2)} \neq 0$ and/or $(\varepsilon^{(1)})^2 p^{(1)} - (\varepsilon^{(2)})^2 p^{(2)} \neq 0$, *i.e.*, due to optical or/and acousto-optical contrast
 488 between the SL layers (see Supporting Information S2).

489 Note that, in the case $\varepsilon^{(1)} - \varepsilon^{(2)} \neq 0$, the second detection path can be due to the difference in
 490 the individual layer thickness variations induced by horizontal displacements components of the

491 gR wave even in the case of the negligible photoelasticity of both layers ($p = 0$). The theory
492 indicates that either the first or the second detection path could dominate depending on the relative
493 strengths of the acoustical (density ρ , elastic moduli C_{ij}), or optical/acousto-optical (permittivity,
494 photoelastic moduli) contrasts between the SL layers. The first path progressively disappears with
495 diminishing acoustic contrast because of the disappearance of the laterally unmodulated
496 component of gR wave. However, it could still be the dominant path in the hypothetical case of
497 the absence of optical and acousto-optical contrasts (see Supporting Information S2 for the details).

498 Overall, the inevitable diminishing of the detected gR wave signals with the diminishing of the
499 sub-optical SL pitch ($\sim d_{SL}/\lambda$, see Supporting Information S2) is not the direct consequence of the
500 fall in light diffraction efficiency due to nanometer scale periodicity but is caused by the
501 diminishing of the acousto-optic scattering volume proportionally to the penetration depth of the
502 gR wave, $\sim d_{SL}$.

503 **E. Ultrafast pump-probe laser experiments**

504 The acoustic vibrations are monitored through ultrafast pump-probe experiments employing a
505 femtosecond laser. A Ti: sapphire laser (Spectra-Physics) with a central wavelength of 810 nm, a
506 pulse width of ~ 100 fs, and a pulse repetition rate of ~ 80 MHz is utilized. The laser beam at the
507 fundamental wavelength probes the sample, while the laser beam at the second harmonic,
508 generated in the BBO crystal, is employed to pump/excite the sample. A translation stage inserted
509 in the pump path induces the pump-probe delay, with a maximum range of ~ 8 ns.

510 The attenuated pump and probe laser beams are collinearly focused by an objective (100 \times , NA:
511 0.55) on the same surface of the sample. The reflected probe light is detected by a large-area 150
512 kHz-bandwidth photoreceiver (NewFocus, model 2031), whose output is connected to a lock-in
513 amplifier for synchronous detection at the 150 kHz modulation frequency of the pump intensity.
514 This allows for low-noise signal acquisition, transmitted to an analog-to-digital convertor for data
515 collection. To test different lateral positions on the sample surface, the sample is displaced relative
516 to laser foci by a translation stage.

517 **F. Signal processing methods**

518 (a) *Background deletion.* The time-dependent reflectivity change undergoes numerical processing
519 to reveal small-amplitude acoustic oscillations. These oscillations are extracted by subtracting the
520 non-oscillating background from the raw signal. The background is obtained using a moving
521 average method with a Gaussian window duration of approximately 100 ps. Removing the
522 background does not introduce any new frequency appearances or alterations; instead, it suppresses
523 undesired relatively low frequency components or environmental/mechanical/thermal noises.

524 (b) *Fourier transform.* The spectra are obtained by applying fast Fourier transform (FFT) to the
525 time-domain acoustic signals. A Hanning window is multiplied over the signal of interest for FFT.

526 (c) *Short-time FFT.* To observe time-dependent changes in spectra amplitude (Figure 2d) and
527 for the second sample (Figure 5d), a short-time FFT procedure is used. During the procedure, the
528 parameters include Gaussian window of 400 ps duration, a frequency resolution for frequency axis
529 of 0.1 GHz, and a hop time of 10 ps.

530 **G. Finite element model (dispersion relation and acoustic eigenmodes)**

531 The dispersion curves of surface acoustic waves (SAWs) propagating at the free surface of
532 assumed infinitely cleaved superlattices are computed using finite element method (FEM) with
533 eigenfrequency analysis in COMSOL Multiphysics.

534 (a) *Structure, material and boundaries.* A unit of the superlattice, consisting of alternating two
535 layers with a period of d_{SL} is constructed (see Supporting Information S1 for details on the
536 structure). The height of the superlattice along the x_3 direction is set to $20d_{SL}$. The [100]
537 crystallographic orientation of constitutive materials is aligned with the x_1 axis (SL growth
538 direction) in the calculation. Since the displacement component normal to the x_1x_3 -plane equals
539 zero, the model is treated as a 2D problem. The two layers are considered as linear elastic cubic
540 materials based on $Al_xGa_{1-x}As$. Floquet periodic boundary conditions are applied to the left and
541 right sides of the unit cell along the x_1 direction. The top surface is assumed to be a stress-free
542 boundary. A perfectly matched layer (PML) is placed at the bottom of the unit cell to simulate an
543 infinite half-space. The bottom surface of the unit cell is set as a fixed boundary to limit the
544 acoustic eigenmodes localized at that bottom surface. The PML length is set as one period of the
545 SL.

546 (b) *Calculation.* The solid mechanics interface of the structural mechanics module of the
547 COMSOL Multiphysics is used to solve an eigenfrequency problem based on the elastic Helmholtz
548 equation. The eigenfrequencies are searched through a parametric sweep over the first Brillouin
549 zone $k_1 \in [0, \pi/d_{SL}]$ with a step of $\pi/d_{SL}/100$. The eigenvalue solver is MUMPS (multifrontal
550 massively parallel sparse direct solver), set to look for 1000 eigenfrequencies to find all the
551 relevant eigenvalues. The eigen vectors for each solution correspond to the mode shape, including
552 the horizontal displacement u_1 (along x_1 -axis) and the vertical displacement u_3 (along x_3 -axis). The
553 computation time for these settings was 6h33min on a computer (Intel core i9-9980HK CPU@2.4
554 GHz).

555 (c) *Mode sorting.* First, solutions with a large imaginary part of its eigenfrequency are excluded.
556 Second, the mode shape (displacement field or energy spatial distribution) of the remaining
557 solutions is used to distinguish the gR wave (confined close to the surface, see Figure 3a), T wave
558 (with dominant vertical displacement, see Figure 3b) and L wave (with dominant horizontal
559 displacement, see Figure 3b) modes. At last, for validation, the obtained mode branches are
560 compared to their expected phase velocities.

561 **H. Brillouin light scattering in cleaved SLs**

562 The efficient backward scattering of the probe light in the zeroth order of diffraction, the only non-
563 evanescent light field component in the deeply sub-optical SL, is governed by the momentum
564 conservation conditions $k_1 = q_1 = 0$ and $k_3 = 2q_3$, where k_i and q_i denote the components of the
565 acoustic and probe light wave vectors, respectively. The normal components k_3 of the coherent
566 acoustic waves in the different diffraction orders m are given by $k_{3,QTA,QLA} = \sqrt{k_{QTA,QLA}^2 - (2\pi m/d_{SL})^2}$, where
567 $k_{QTA,QLA} = \omega/v_{QTA,QLA}$ are the wave vectors of the quasi-transversal (QTA) and quasi-
568 longitudinal (QLA) acoustic waves propagating along the directions of the respective diffraction
569 orders in the SL, d is the SL period and $m = 0, \pm 1, \pm 2 \dots$. Therefore the velocities $v_{QTA,QLA}$ of the

570 QTA and QLA waves in the above formula should be evaluated along the directions defined by
571 the following angles $\gamma \equiv \gamma(d_{SL}, n) = \tan^{-1}[(2\pi m/d_{SL})/k_{3, QTA, QLA}] = \tan^{-1}[(2\pi m/d_{SL})/$
572 $(2q_3)]$ relative to the x_3 axis. Thus, the knowledge of the probe light wave vector and the slowness
573 curves (anisotropy) of the acoustic modes in the SL provides an opportunity to estimate the
574 Brillouin frequencies.

575 In our estimates, as described in Supporting Information S1 and based on the weak elastic
576 contrast between the layers, we used the dependences of the acoustic velocities on the propagation
577 direction/angle provided by the slowness curves of the GaAs⁴² and averaged optical refractive
578 index $\langle n \rangle$. Two of the spectral peaks in the lower part of Figure 2c can be tentatively attributed to
579 Brillouin scattering of probe by QLA waves in the orders $m = 0$ and $m = 1$, respectively. All these
580 waves are non-evanescent. Note that symmetry considerations prevent the generation of the QTA
581 waves with the order $m = 0$ in our experimental configuration.⁴² It is also worth noting that the SL
582 is optically birefringent,⁵⁴ and thus, potentially each of the Brillouin peaks could be composed of
583 up to three neighboring spectral components, corresponding to different combinations of ordinary
584 and extraordinary probe light propagating from and towards the SL surface.⁵⁵ We have
585 experimentally verified that the position of the most intense Brillouin peak around 45.7 GHz
586 (Figure 2c) depends on the polarization of the probe light. However, the splitting of the peak is not
587 resolved in our experiments because of weak birefringence, which is caused only by structuring,
588 while the materials of the SL layers are optically isotropic.

ASSOCIATED CONTENT

589 Supporting Information

590 The Supporting Information is available free of charge at <https://pubs.acs.org>

- Structure of generalized Rayleigh-type surface acoustic wave on a superlattice stratified normal to its surface; optical detection of generalized Rayleigh-type surface acoustic wave on a superlattice stratified normal to its surface (PDF)

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Author Contributions

C.L. conducted the experiments, executed the simulations and input corresponding sections for the manuscript. N.C conducted the first experimental tests and helped with the experiments. T.T. participated the first trial experiments. V.T helped with the initial simulations. N.D conducted the AFM characterization. H.L and K.Z. prepared and characterized the samples by XRD. S.R. assisted with signal processing, improved the manuscript. V.G. and H.L designed the samples. V.G. designed the methodology, wrote the first manuscript and supervised the project. All authors reviewed the manuscript.

Notes

The authors declare no competing financial interest.

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ABBREVIATIONS

SL, superlattice; SAW, surface acoustic wave; OA, optoacoustic; AO, acousto-optic; OAOT, opto-acousto-optic transducer; EUV, extreme ultraviolet; NIR, near infrared; MBE, molecular beam epitaxy; XRD, X-ray diffraction; AFM, atomic force microscopy; gR, generalized Rayleigh surface acoustic wave; T, skimming surface transverse acoustic wave; L, skimming surface longitudinal acoustic wave; QLA, quasi-longitudinal acoustic; QTA, quasi-transversal acoustic.

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