Monochromatic transverse-polarized phonons from femtosecond pulsed optical excitation of a GaAs/AlAs superlattice

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We have generated pulses of ~0.5 THz, monochromatic transverse-polarized acoustic (TA) phonons by resonant excitation of a (001) GaAs/AlAs superlattice (SL) structure using femtosecond optical pulses. The phonons were detected using superconducting bolometers. Spectral resolution was obtained by using the filter effect of the frequency-dependent phonon scattering in the GaAs substrate and also using a second superlattice as a notch filter placed between the generator and bolometer. We propose that the detected TA phonons are due to the leakage of optically excited coherent SL modes into propagating phonons. We discuss various mechanisms by which TA phonons, which are not Raman active in (001) GaAs, may couple with the optical pump pulse.

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I. INTRODUCTION

Beams of coherent terahertz acoustic phonons have numerous potential applications in science and technology, including: THz light modulation, generation of THz electromagnetic radiation, sources for phonon spectroscopy, acoustic microscopy of solid-state nanostructures, etc. One way to generate such phonons is by ultrafast optical excitation of semiconductor superlattice (SL) structures, for reviews see Refs. 1,2: the periodicity of the acoustic impedance of the layers making up the SL gives rise to folding of the acoustic phonon dispersion into a mini-Brillouin zone extending from $q=0$ to $q=\pi/d_{SL}$, where d_{SL} is the superlattice period, see Fig. 1. This makes it possible to optically excite zone-folded acoustic phonons having $q=0$ but with high (~THz) frequency. The (001) GaAs/AlAs SL has proved an ideal system for coherent phonon generation because it is possible to accurately grow by molecular-beam epitaxy (MBE) SL structures with excellent interface quality and which show optical absorption at wavelengths within the tuning range of Ti-sapphire ultrafast oscillators. The generation process has been investigated using ultrafast pump-probe measurements,[4] in which a high-intensity femtosecond pump pulse is followed, after an adjustable time delay, by a low-intensity pulse which probes changes in the surface reflectivity. It was observed that when the pump photon energy was resonant with the E1-HH1 optical transition in the GaAs quantum wells (QW's), coherent longitudinal acoustic (LA) phonons were excited in the SL.[5] These were observable through oscillations in the surface reflectivity with a period equal to the reciprocal of the phonon frequency. Detailed measurements of the spectrum of generated phonons[6] revealed a triplet of modes consisting of a dominant line at frequency $\nu=c_{LA}/d_{SL}$ (c_{LA} is the speed of LA phonons) corresponding to the first mini-zone-center mode at $q=0$ and a pair of sidebands at $q=2k_{laser}$ due to backscattering. The results were found to be consistent with the generation process being impulsively stimulated-Raman scattering (ISRS) in which the photoexcitation of an electron-hole pair is accompanied by LA phonon emission. However, other generation mechanisms, e.g., displacive excitation,[7] have been proposed.

The SL vibrations were observed to decay within a nanosecond after the pump pulse. This was due to leakage of the SL modes into propagating monochromatic phonons. Using coherent (pump-probe) detection schemes, these phonons have been detected at distances of up to 0.5 μm from the SL.[8] Propagation of the monochromatic LA phonons across macroscopic distances, ~1 mm, at liquid helium temperatures has also been observed using incoherent detection by superconducting bolometers.[9–11] Superconducting bolometers have a response time of the order nanoseconds which made it possible to resolve the ballistic LA and TA phonon modes due to their different speeds and therefore different times of flight between the SL and bolometer. The intensity of the ballistic LA mode was observed to increase sharply when the condition for coherent phonon generation in the SL was met, i.e., the excitation photon energy was resonant with the E1-HH1 transition in the GaAs QW's.[9] The spectrum of the propagating phonons was studied using the frequency

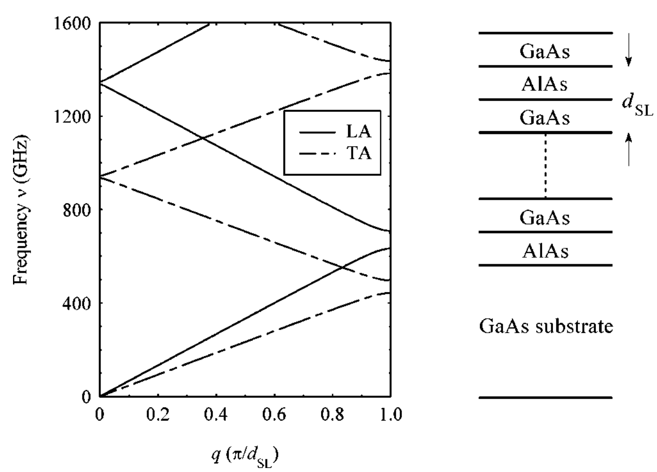

FIG. 1. Folded acoustic phonon dispersion, calculated using the method described in Ref. 3, for a typical superlattice structure consisting of 40 periods each containing 7 ML of GaAs and 7 ML of AlAs ($d_{SL}\simeq 4$ nm).

dependent phonon scattering in the substrate as a low-pass filter[10] and by using a second SL as a notch filter between the generator SL and the bolometer.[11] These measurements showed that the propagating phonon beam was monochromatic at the design frequency of the generator SL. As noted in Refs. 9–11, a strong transverse acoustic (TA) signal was also detected by the bolometer. This was attributed to the decay products of optic (LO) phonons emitted during energy relaxation by carriers photoexcited in the GaAs substrate. However, more recent measurements with improved temporal resolution found that the intensity of the ballistic TA mode was also enhanced when the condition for coherent phonon generation was met.[12] Three possible reasons for this have been suggested: (1) the TA phonons were emitted by carriers photoexcited in the GaAs QW's relaxing their excess energy; (2) the TA phonons were produced by the anharmonic decay of the THz LA SL modes; and (3) coherent transverse acoustic SL modes were being excited optically and these were leaking into propagating TA phonons in the same way as for the LA mode. At first, the latter seemed unlikely because the generation of coherent TA phonons has not been observed in pump-probe measurements on (001) GaAs/AlAs SL's. This is believed to be due to TA phonons propagating along [001] in GaAs not being Raman active.[13]

The aim of the work described in this paper was to determine the origin of the additional ballistic TA phonon signal which appeared under resonant photoexcitation of the SL. We report measurements, made using superconducting bolometer detectors and filter layers, of the temporal, frequency, angular, and excitation wavelength dependence of the TA signal intensity. The results are discussed in relation to the three possible generation processes given above.

II. EXPERIMENTAL METHOD

The experimental samples were all grown by MBE on semi-insulating GaAs substrates. Four different samples were produced. Sample *A* contained a single 40-period phonon generator SL, each period consisting of 22 monolayers (ML) of GaAs and 4 ML of AlAs. In sample *B*, two 40-period SL's were grown: each period of the SL closest to the substrate consisted of 7 ML of GaAs and 7 ML of AlAs; on top of this was grown a 0.5 μm GaAs spacer and then a generator SL (the same structure as in sample *A*). The lower SL was designed to act as a phonon notch filter with a narrow stop band centered on $\nu_{stop}=\pi/2d_{SL}$, i.e., the first minizone boundary mode. Sample *C* differed from sample *B* only in the generator layer composition; 24 ML of GaAs and 8 ML of AlAs. The SL of sample *D* was as sample *A*, but grown on a 2-mm-thick substrate. The SL's were characterized by photoluminescence and Raman spectroscopy.

The back surface of the substrates, opposite the SL's, were polished and 40×40 μm^2 active area granular aluminum bolometers were fabricated by photolithography. Sample *D* was polished at an angle, 2 mm thick at one end and 0.5 mm thick at the other. A row of bolometers were fabricated along the wedge allowing us to study the effect of changing phonon propagation distance, by optically exciting the SL at points opposite different bolometers.

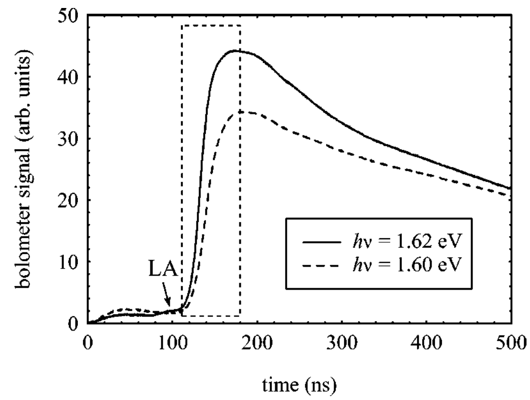

FIG. 2. Bolometer signals for pump photon energy below resonance and resonant with the E1-HH1 transition (1.62 eV) in sample *A*. The box highlights the region where the increase in the ballistic TA signal is seen.

The measurements were carried out with the samples mounted in an optical-access helium cryostat and held at a constant temperature on the superconducting transition of the bolometers, ≈ 2 K. Pulses from a tunable Ti-sapphire oscillator were focussed to a 40 μm-diameter spot on the SL surface, opposite a bolometer. The pulses had a duration of ≈ 100 fs and a maximum energy of 2 nJ. An acousto-optic pulse picker was used to reduce the pulse period from 82 MHz at the laser to <100 kHz at the sample. This was to allow the sample and bolometer sufficient time to return to thermal equilibrium between pulses. Multiple reflections of the beam between a pair of double-chirped negative-GVD mirrors precompensated the pulse for dispersion caused by optical components in the beam path.

The bolometer was biased with a constant current of 10 μA. Phonons incident on the bolometer caused a transient change in its temperature and hence its resistance. The corresponding voltage pulse was amplified and recorded using a high-speed digital signal averager. Figure 2 shows typical signals after photoexcitation of the SL in sample *A*. The two traces are for phonon energies corresponding to the on-resonance condition ($h\nu=E_0=1.62$ eV) and the off-resonance condition ($h\nu<E_0$). The peak at $t=0$ is due to direct optical excitation of the bolometer by PL. The signal due to the arrival of LA phonons is marked ($t\simeq 80$ ns). This feature has been studied in great detail in previous work.[9–11] The larger peak observed at $t\simeq 120$ ns is mostly due to TA phonons and is the focus of this paper.

III. RESULTS AND DISCUSSION

A. The experimental results

Close inspection of Fig. 2 reveals that when the resonance condition is satisfied the TA component of the signal is enhanced on its rising edge and reaches a higher peak. At times $\gtrsim 500$ ns following the peak (not shown on this scale), the tail signal is reduced compared to the off-resonance case. The effect at early times following the pump pulse can be seen more clearly when the off-resonance signal is subtracted from the on-resonance signal. This effectively removes the

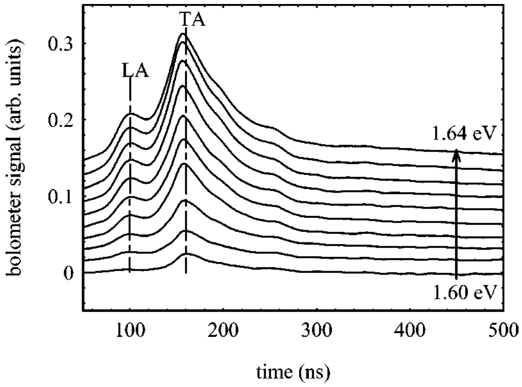

FIG. 3. Sample *A* bolometer signals after subtraction of the off-resonance signal (traces are offset for clarity). The increase of the LA and ballistic TA phonon pulses takes place over a 40 meV energy range centered on 1.62 eV, consistent with the spectral width of the ultrafast laser excitation pulse.

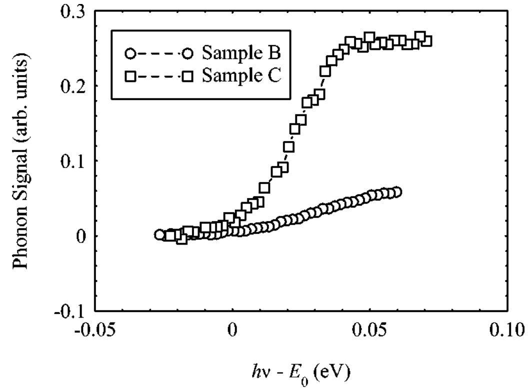

FIG. 4. Ballistic TA signal component as a function of excitation photon energy for samples *B* and *C*.

large background due to carrier relaxation in the GaAs. The background subtracted signals are shown in Fig. 3. When $h\nu \simeq E_0$, there is a strong increase in the observed TA signal. This behavior is similar to the LA mode enhancement first reported in Ref. 9. The TA peak also shifts slightly to earlier times. Note that the TA signal increase occurs over a narrow, but finite range of excitation energy consistent with the spectral width of the transform limited 100 fs laser pulses.

The appearance of the off-resonance signal is well understood. At these wavelengths, there is little optical absorption in the SL, and the phonon signal is largely due to energy relaxation by carriers photoexcited in the GaAs substrate. Since the excess energy, $h\nu - E_g$ (E_g= GaAs band-gap), is greater than the GaAs LO phonon energy ($\hbar\nu_{LO} \simeq 36$ meV), most of the excess energy is lost by LO phonon emission. These modes decay in a few picoseconds to large wave-vector LA modes which subsequently undergo anharmonic decay to large wave-vector TA phonons. Isotope mass defects, impurities, and other crystalline defects strongly scatter these large wave-vector modes, and so diffusive propagation results in delayed arrival of the phonons at the bolometer compared to the ballistic flight time. The observed signal shows a broad, delayed peak and a long tail, which is generally accepted as being characteristic of the arrival of the delay products of emission of LO phonons.[14]

The additional TA signal we observe on resonance arrives at a time corresponding to the ballistic time of flight ($\simeq 120$ ns). This suggests the origin of the signal is due to phonons of frequency ($\lesssim 1$ THz) which are not scattered in the substrate, rather than being the result of diffusive propagation. Further evidence for the ballistic nature of the on-resonance TA signal is provided by the shift of the peak to slightly earlier times as a larger proportion of the TA signal is due to ballistic phonons. The signal at the ballistic time of flight is accompanied by a reduction in the tail of the signal, probably due to the increase in optical absorption by the SL resulting in less direct excitation of the substrate.

Having discounted the possibility that the TA signal enhancement is due to diffusive propagation of decay products from the substrate, we will now move on to consider the generation processes outlined in Sec. I. Initially, we limit our discussion to sample *A*. Consider first the possibility of carrier relaxation in the GaAs QW's as being responsible for the TA enhancement. The excess energy of carriers photoexcited in the QW's is less than the GaAs LO phonon energy, and so energy relaxation is by emission of acoustic phonons. Momentum conservation considerations lead to an upper frequency cutoff for emission near perpendicular to the QW growth direction, $\nu_{max} = c_s/2w$,[15] which in this sample gives $\nu_{max} = 200$ GHz. Next consider anharmonic decay of the coherent LA superlattice modes: there are two possible decay channels, LA→LA+TA and LA→TA+TA,[16] both of which would result in sub-THz transverse phonons, since we know the LA mode is of frequency 650 GHz. Finally, if the increase in TA signal was due to optically excited TA superlattice modes leaking into propagating monochromatic phonons, their frequency would be 450 GHz.

It is clear that using sample *A* alone, we are unable to identify the origin of the signal enhancement. To determine whether the TA phonons were monochromatic we used samples *B* and *C* which contain the notch filter SL structure. These samples, as itemized previously, consist of 2 SL's separated by a GaAs spacer layer. The filter layer was designed to block the propagation of transverse phonons of a frequency 450 GHz. The generator layers were then fabricated so that in sample *B* TA superlattice modes would have a frequency of $\nu_{TA} \simeq 450$ GHz, whilst in sample *C*, $\nu_{\mathrm{TA}} \simeq 360$ GHz. Figure 4 shows the dependence of the bolometer signals on the excitation energy for samples *B* and *C*. We see that in sample *B* unlike for sample *A*, no sudden increase of the ballistic TA signal is observed when the pump photon energy is tuned onto resonance with the E1-HH1 transition in the GaAs QW's. This result provides strong evidence of the monochromatic nature of the additional TA component. The optically excited TA modes would be filtered since the generator layer frequency matches the stop band of the filter layer. However, in sample *C*, we still see an increase in the phonon signal when the resonance condition is satisfied. Since the generator layer of sample *C* was designed to a phonon frequency outside of the filter stop band, the filter should have no effect. This is indeed what is observed, proving that the filter SL is frequency selective and does not simply block all TA phonons.

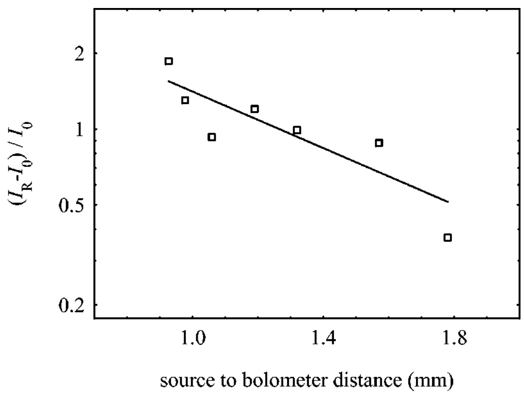

FIG. 5. $(I_R-I_0)/I_0$ for TA phonons as a function of the propagation distance along [001], measured using the wedge-shaped sample *D*. Here I_R is the signal intensity under resonant excitation and I_0 is the signal intensity for off-resonant excitation. The solid line is a best fit to the data and gives a phonon mean free path of 0.8 mm.

The notch filter experiment rules out the possibilities that the TA enhancement is due to carrier relaxation or anharmonic decay of LA SL modes because neither of these processes would result in generation of monochromatic phonons and then we would have expected samples *B* and *C* to behave in the same way. However, the possibility that the decay of the *propagating* LA modes can account for the increased TA component must be examined further. In this case, because the filter layer prevents propagation of the monochromatic LA phonons, the TA signal would also be attenuated.

To eliminate this possibility, we have studied the effect of phonon propagation distance on TA signal enhancement using sample *D*. Figure 5 shows the difference between on- and off-resonance TA signals as a function of distance between source and detector. To account for geometrical effects (i.e., the change in solid angle subtended by the detector as the propagation distance increases), we have normalized the traces to the off-resonance TA signal amplitude. In previous work using sample *D*, we observed that the LA signal intensity decreased with increasing propagation distance due to isotope scattering or LA decay.[10] If the additional TA signal was due to the arrival of LA decay products, we would observe an increase of the TA component with increasing propagation distance. Instead, we see a decrease of the TA intensity as the source-detector separation is increased, probably due to scattering in the substrate and consistent with a phonon mean free path of ≈ 0.8 mm. This rules out the decay products of propagating monochromatic LA phonons as the origin of the additional TA component.

B. The origin of the additional transverse phonon signal

We now turn our attention to discussing the possible excitation mechanisms. Having discussed and subsequently discounted the various possible causes of the additional TA signal, the one which remains is that the TA signal is due to coherent TA modes leaking into propagating phonons. The fact that these SL modes have not been previously observed in pump–probe measurements is not sufficient reason to reject this explanation. Transverse acoustic modes are not Raman active in (001) GaAs.[13] This means that generation by ISRS (the mechanism by which generation of the LA mode occurs) is not expected. Also, because the surface reflectance is proportional to the first-order Raman tensor, TA phonons will not modulate the reflectance. Our measurements using incoherent, bolometric detection show that, in most respects, the TA and LA modes behave in a very similar way under resonant excitation. However, the ratio of the on-resonance to off-resonance signal intensities is much smaller for the TA mode than for the LA. This is due to the overall TA signal being dominated by the products of carrier energy relaxation.

Since ISRS is not expected to generate TA phonons, we must consider the possibility of non-Raman processes. A possible candidate is displacive excitation of coherent phonons (DECP),[7,17] the physical basis of which is that ions are set into motion via the coupling to the photoexcited carriers or a sudden change in carrier temperature induced by absorption of the optical pulse. If the optically induced changes occur on a time scale that is very short compared to the phonon period then coherent modes are excited. However, DECP excites only the vibrational modes of A_1 symmetry, i.e., LA modes in (001) GaAs/AlAs. Brillouin scattering measurements on CdS have allowed observation of forbidden TA modes, and this has been attributed to piezoelectric coupling.[18] Due to the piezoelectric effect, the slow TA phonon carries a longitudinal electric field and it is this that couples with the light. However, as discussed in Ref. 18 this mechanism should not be effective in (001) GaAs because pure TA phonons propagating along [001] have no component of longitudinal polarization.

Another possibility is that the effects of acoustic anisotropy give rise to excitation of the TA modes. This was suggested as a possible reason for the observation of Raman scattering by TA phonons in (110) GaAs/AlAs SL's.[19] Acoustic anisotropy has also been shown to have a large effect on the mode and angular dependence of the electron-phonon coupling in GaAs QW's and heterojunctions,[20] and this has been observed in hot carrier energy relaxation measurements.[21] Acoustic anisotropy effects are very strong for TA phonons propagating close (within a few degrees) to [001] in GaAs and it is this that leads to the characteristic strong TA phonon focussing patterns.[22] In these directions, the TA modes are not ''pure'' transverse, i.e., the wave vector and polarization vector are not exactly perpendicular. The TA mode in this case carries a small longitudinal polarization component which may permit excitation of slightly off-axis TA phonons by ISRS or the piezoelectric coupling mechanism described in Ref. 18. The TA signal is expected to be weak and easily obscured by the LA mode in pump-probe measurements. It is the ability of the bolometric detection technique to resolve the phonon modes due to their differing times of flight that has allowed us to observe this enhancement of the TA mode when the condition for resonance is satisfied.

IV. CONCLUSIONS

Acoustic phonons generated by femtosecond pulsed optical pumping of a GaAs/AlAs superlattice have been detected using superconducting phonon detectors. Enhancement of the ballistic transverse acoustic phonon signal was observed when the excitation photon energy was brought into resonance with the E1-HH1 transition in the GaAs QW's. Measurements using samples which comprised a notch filter SL between the generator SL and detector showed that the TA phonons were monochromatic with frequency $\nu = c_s/d_{SL}$, corresponding to the first mini-Brillouin zone-center mode. Consideration of the various possible sources of the monochromatic TA phonons have led us to the conclusion that the additional transverse phonon signal is due to leakage of optically excited SL modes. Initially, this was unexpected since TA phonons are not Raman active in (001) GaAs. TA phonons which are not pure transverse, but carry a small component longitudinal polarization may allow generation (by ISRS) of monochromatic TA modes which propagate close to [001]. We believe these effects of acoustic anisotropy can account for our observations.

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