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Terahertz phonon optics in GaAs/AlAs superlattice structures

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We have generated pulsed beams of ~THz monochromatic longitudinal acoustic phonons by ultrafast laser excitation of GaAs/AlAs superlattices. The phonons propagated ballistically across the GaAs substrates at low temperatures and were detected using superconducting aluminum bolometers. Between the generator superlattice and the bolometer was a second superlattice structure which acted as a reflective band-stop filter. We have made measurements of the phonon pulses in various samples for which the filter stop band was either in tune or out of tune with the generator frequency. These have enabled us to place an upper bound on the bandwidth of the monochromatic phonon beam. We have also determined the relative proportions of the detected longitudinal acoustic phonon signal that are due to monochromatic phonons and broadband phonons from carrier relaxation. We present this experiment as an example of ''phonon optics'' using propagating monochromatic phonons in the THz frequency range.

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Phonon optics is the analog of conventional optics, but for phonons instead of photons. Using appropriate structures, interference and diffraction effects and other phenomena associated with the wave nature of the phonons should be observable. The related field of phonon spectroscopy can be applied to the study of the physical properties of condensed matter. Phonon spectroscopy using acoustic phonons in the THz frequency range is particularly sensitive to the electronic and mechanical properties of nanostructures.

Over the past few decades significant progress has been made using ''heat-pulse'' phonon sources and superconducting bolometer detectors.[1] The temporal resolution is good, down to a few nanoseconds, allowing time-of-flight studies. However, the spectral resolution is very poor because heat-pulse sources generate phonons with a approximately Planckian spectrum and the bolometer is an energy detector. The heat-pulse technique in phonon spectroscopy is analogous to performing optical spectroscopy using an unfiltered tungsten lamp and thermopile. Continuous-wave (cw) and pulsed monochromatic phonon sources in the THz range, based on superconducting and normal-state tunnel junctions, have been developed and used in phonon spectroscopy.[2,3] Disadvantages with these devices are that the monochromatic component is superimposed on a large background of phonons with a broad distribution of frequencies, the temporal resolution is poor compared to bolometers, and superconducting tunnel junctions cannot be used in magnetic fields. Types of phonon detector that offer some limited spectral resolution include superconducting tunnel junctions[4] and semiconductor tunnelling structures.[5,6] Somewhat better resolution, but at a fixed frequency of 0.87 THz, can be obtained by exploiting the photoluminescence properties of chromium ions in sapphire.[7] Useful as all these methods have been, the best resolution, spectral and temporal, of phonon spectroscopy still falls far short of what is routinely achieved in conventional (photon) optics. Obviously phonon spectroscopy and phonon optics would benefit from sources of pulsed beams of monochromatic phonons having a very narrow spectral linewidth.

Recently it has been shown that coherent acoustic phonons may be generated by optical means. cw-laser-induced thermomodulation of a metal film on the surface of a crystal has been used to generate propagating beams of GHz phonons which have been utilized in phonon optics experiments.[8–10] THz coherent longitudinal acoustic (LA) phonons have been generated by resonant photoexcitation of superlattice (SL) structures.[11] The periodicity in the acoustic impedance of a SL structure gives rise to a mini Brillouin zone (BZ) into which the acoustic phonon dispersion is folded.[12] This permits optical coupling with high-frequency acoustic modes having $k \approx 0$. The coherent SL modes may be detected by measurements of the time dependence of the reflectivity; for a review see Ref. 13. In more recent experiments using GaAs/AlAs SL's,[14,15] we have shown that these phonons leak out of the SL into propagating monochromatic LA phonons which can be detected at distances up to 1 mm from the source using a bolometer.

The ability to grow by molecular beam epitaxy (MBE) SL structures with interfaces that are smooth and abrupt on the scale of a single atomic layer (much less than the typical phonon wavelength) makes SL's ideal systems in which to study phonon optics.[16] The nearest conventional optics equivalent is the study of the transmission of light through multilayer dielectric films. In this paper we describe a phonon optics experiment using pulses of ~0.6 THz LA phonons generated by impulsive stimulation of GaAs/AlAs SL's with femtosecond laser pulses. The phonons are detected by a superconducting bolometer on the back of the substrate. Between the generator SL and the bolometer is a second SL which acts as a notch filter. The transmission of acoustic phonons through SL's has previously been studied

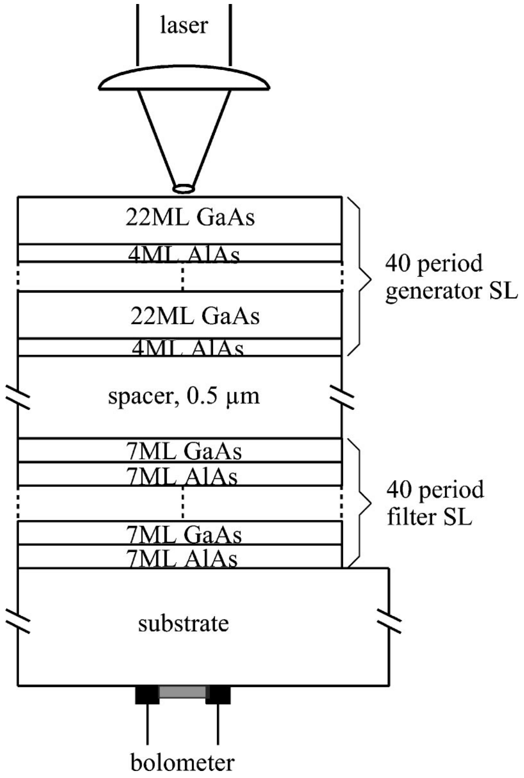

FIG. 1. The structure of sample B. Sample A has the same generator SL but no filter and sample C has a 24-ML-GaAs:8-ML-AlAs generator SL.

using superconducting tunnel junctions and heat pulses. Dips in the phonon transmission at particular frequencies corresponding to the filter stop bands were observed.[17,18] However, because the resolution was limited, these dips were weak and superimposed on a large background signal. In our experiment using a narrow-line monochromatic source we observe strong, $\sim 100\%$, attenuation of phonons in a sample where the filter stop band overlaps the SL generator frequency.

The results that we describe here are based on three samples, all grown by MBE on 0.4-mm semi-insulating GaAs substrates. The design of the SL's was optimized to have a small gap at $k \approx 0$ for the generator SL, giving a narrow linewidth of the monochromatic phonons and a wide stop band for the filter SL. Sample A consisted of a 40-period SL, each period containing 22 monolayers (ML) of GaAs and 4 ML of AlAs. This formed the generator SL; there was no filter SL in sample A. The structure of sample B is shown in Fig. 1; it consisted of a 40-period generator SL, the same as in sample A, but below it was a filter SL consisting of 40 periods each of 7 ML GaAs and 7 ML AlAs. The two SL's were separated by a 0.5-μm GaAs spacer layer. Sample C was of the same design as sample B, except that the generator SL consisted of 40 periods each of 24 ML GaAs and 8 ML AlAs. Photoluminescence (PL) and Raman spectroscopy were used to characterize the SL structures. The back surfaces of all three samples were polished and aluminum bolometers of active area 40 μm^2 were fabricated photolithographically.

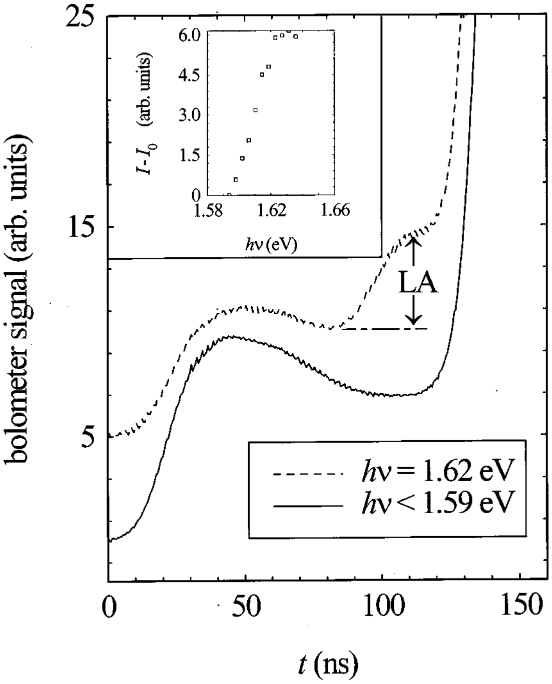

FIG. 2. Bolometer signals for sample A and for two excitation photon energies: off resonance, $h\nu < 1.59$ eV, and on resonance, $h\nu = 1.62$ eV, with the GaAs quantum well (QW) exciton energy. The inset shows the LA signal size as a function of the excitation photon energy, after subtraction of the off-resonance signal I_0.

All the measurements were carried out in an optical cryostat at the superconducting transition temperature of the bolometer, $T \simeq 2$ K. The generator SL was excited by ≈ 100-fs-long pulses from a tunable mode-locked Ti-sapphire laser focused to a spot of diameter 40 μm directly opposite the bolometer. The energy per pulse was about 2 nJ, and the pulses were picked at a frequency of 100 kHz to give the sample and bolometer time to return to thermal equilibrium in between them. A high-speed digitizer and signal averager were used to capture the bolometer signal.

Typical bolometer signals obtained using sample A are shown in Fig. 2. As shown in the inset, we measure a large increase in the LA phonon signal at laser photon energy $h\nu \approx 1.61$ eV (note that the increase actually takes place over the range 1.59–1.63 eV which is consistent with the spectral width of the laser pulse). This matches the condition for the excitation of coherent phonons in the SL; that is, the laser photon energy $h\nu$ must be equal to or larger than the ground-state exciton energy E_0 in the GaAs quantum wells. As we have shown in previous work,[14,15] the increase in LA signal is due to the leakage of the SL modes into propagating monochromatic phonons which reach the bolometer. The slower transverse acoustic (TA) mode phonon signal is largely due to phonons emitted by carriers relaxing excess energy in the GaAs. There is also a very weak "off-resonance" LA signal for $h\nu < 1.59$ eV, also due to carrier relaxation. Figure 3 shows the LA signal intensity, following subtraction of the off-resonance signal, as a function of the excitation photon energy for samples B and C. For sample C we obtain an increase in LA signal for $h\nu \geqslant E_0$, the same as

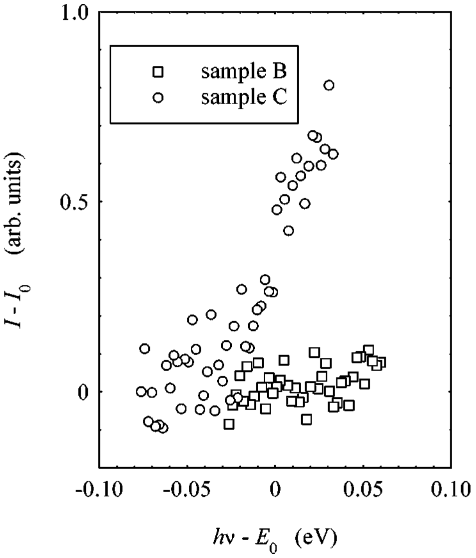

FIG. 3. LA signal size (after subtraction of off-resonance signal) as a function of excitation photon energy for samples B and C. E_0 is the GaAs QW ground-state exciton energy.

for sample A. However, no increase of the LA signal is seen with sample B. We explain these observations by considering the effect of the filter SL on the monochromatic phonons generated in samples B and C.

The acoustic dispersion relations of the SL's used are shown in Fig. 4(a). These were calculated following the method of Tamura *et al.*[12] We considered SL structures consisting of n_α ML of material α (GaAs) having phonon speed c_α and acoustic impedance Z_α and n_β ML of material β (AlAs) having phonon speed c_β and impedance Z_β. The gaps (stop bands) in the dispersion at the BZ center and at the BZ boundary occur at frequencies

$$\omega_m = m\pi \frac{1}{d_{ML}(n_\alpha/c_\alpha + n_\beta/c_\beta)},$$

where d_{ML} is the monolayer thickness ($\simeq 2.83$ Å). The widths of the stop bands are given by

$$\Delta\omega_m = \frac{2\omega_m}{m\pi}(2\gamma)^{1/2}\left|\sin\left(\pi m \frac{n_\alpha/c_\alpha}{n_\alpha/c_\alpha + n_\beta/c_\beta}\right)\right|,$$

where $\gamma = (Z_\beta - Z_\alpha)^2/2Z_\alpha Z_\beta$. Under resonant photoexcitation, coherent LA phonons are generated in the SL with a fundamental frequency corresponding to the first BZ center ($q=0$) mode—i.e., $m=2$. For the generator SL in samples A and B, this is at $\nu = \omega_2/2\pi = 0.66$ THz and in sample C at $\nu = 0.54$ THz. There will also be sidebands corresponding to $q = 2k_{laser}$ and higher order modes ($m = 4,6,\ldots$). However, all these are much weaker than the fundamental.[19,20] Figure 4(b) shows the frequency dependence of the transmission coefficient for the filter SL in samples B and C. This was calculated using the transfer matrix approach.[21] We see the filter SL has a 80-GHz-wide stop band corresponding to the first ($m=1$) mini-BZ-boundary mode centered at $\nu = 0.67$ THz. The transmission of the filter is very small inside this stop band which overlaps the generator SL frequency in sample B, but not in sample C.

We believe that the absence of any increase in the LA signal for $h\nu \geqslant E_0$ in sample B is due to the filter SL blocking the propagation of 0.66 THz phonons to the bolometer. We can discount the possibility that the filter SL blocks all LA phonons because an increase in LA signal for $h\nu \geqslant E_0$ is observed using sample C. Signal loss due to scattering of 0.66 THz phonons in the GaAs substrate can also be discounted because an increase in the LA signal is seen using sample A. One other possibility which should be considered is that the LA phonons emitted under resonant photoexcitation are due to photogenerated carriers relaxing their excess energy within the quantum wells. For this process, the phonons emitted perpendicular to the wells are mostly of low frequency up to a cutoff imposed by the condition of conservation of crystal momentum. The cutoff frequency $\nu_c = c_\alpha/2n_\alpha d_{SL} = 385$ GHz for samples A and B and 353 GHz for sample C. Both these are well clear of the filter stop band and so we can discount carrier relaxation as the source of 0.66 THz LA phonons.

In summary, we have carried out a phonon optics measurement using ~ 0.6 THz monochromatic phonons generated by resonant photoexcitation of a GaAs/AlAs SL.

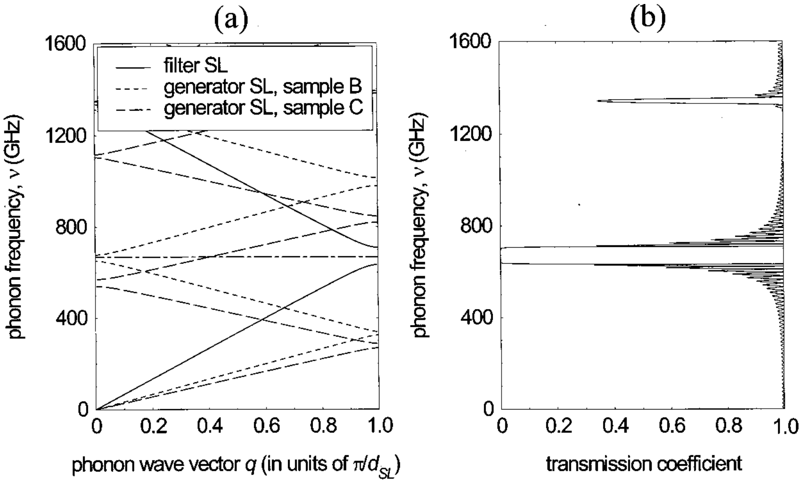

FIG. 4. (a) Folded phonon dispersion for the filter SL (solid line) and the generator SL's in samples B and C. (b) Transmission coefficient of the filter SL as a function of phonon frequency.

Fabricating a filter SL in between the generator SL and detector leads to a 100% attenuation of the phonon beam when the filter stop band matches the generator frequency. Our results lead us to conclude that the spectral linewidth of the propagating monochromatic phonon beam is less than 80 GHz (the width of the filter stop band).

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