Observation of Large Acoustic Gain in Coherent Acoustic Phonon Oscillators

Chi-Kuang Sun, Gia-Wei Chern, Kung-Hsuan Lin, and Yue-Kai Huang

Graduate Institute of Electro-Optical Engineering and Department of Electrical Engineering, National Taiwan University, Taipei 106, Taiwan R.O.C.

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Amplification of coherent acoustic phonon oscillations in InGaN multiple-quantum-wells (MQWs) has been observed. After initiating the coherent acoustic phonon oscillation in InGaN MQWs with an ultraviolet (UV) femtosecond (fs) pulse, a second UV fs pulse was introduced into the MQWs to initiate another coherent oscillation. Through an interferometric analysis based on a simple harmonic oscillator theory, a large coherent amplification (3.5 dB) of the initial acoustic phonon oscillation was revealed.

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Coherent acoustic amplification, which provides the necessary gain to realize a self-sustaining phonon source, is the key to the construction of a phonon laser. Phonon amplification can be achieved through various physical mechanisms, which have already been theoretically proposed and analyzed. Some experimental implementations of phonon amplification have also been reported. Stimulated phonon emissions have been observed in a spin-phonon system of crystals doped with paramagnetic ions [1-3] under low temperature (e.g. 1.5 K). A localized defect-induced two-level system in an amorphous media also provides the avenue to achieve a population inversion of carriers and the corresponding phonon gain. Sound amplification by the stimulated emission of phonon radiation (SASER) has been observed with a 2dB acoustic gain at 20 mK for 340 MHz waves [4]. Coherent phonon avalanches and the stimulated emission of acoustic phonons in an acoustic cavity have also been demonstrated in Cr^{3+}:Al_2O_3 crystals [5]. In all of these previous works, the phonon vibration modes were relatively weak and the acoustic gain was relatively low. With strong acoustic phonon modes, semiconductors might be an excellent candidate for large acoustic phonon amplification. Stimulated phonon emission, based on coherent phonon-assisted tunneling [6,7] and various electron-phonon [8-12] interactions in semiconductors, has been proposed. Cerenkov amplification, based on piezoelectric coupling, has been theoretically analyzed for both bulk and confined semiconductor systems [8,9]. Sound amplification utilizing electrostrictive electron-phonon coupling has been proposed [10,11]. The possibility of phonon lasers utilizing macroscopic nonlinear acoustic effects, resembling a free-electron laser, was also theoretically analyzed [12].

Recent progress in femtosecond spectroscopy technology has enabled us to generate, observe [13-18], and control [19-23] the coherent oscillations of high-frequency acoustic phonon modes in semiconductors using ultrashort optical pulses. By initiating the coherent acoustic phonon oscillation in piezoelectric/electrostrictive semiconductor multiple-quantum-wells (MQWs), it should be easier to study the acoustic amplification effect, due to the coherent nature of the initiated phonon oscillation. At the same time, piezoelectric semiconductor MQWs can act as optical transducers for coherent acoustic wave generation and detection. A large coherent acoustic phonon oscillation has been demonstrated by our group, using InGaN MQWs with a high piezoelectric field [15,17]. Here we report the evidence for coherent acoustic amplification and large acoustic gain in the InGaN MQW system.

In order to study the acoustic amplification effect in InGaN MQWs, we performed experiments with a standard transmission type pump-probe technique [15,17,21]. The MQWs consist of 14 periods of InGaN/GaN quantum wells (QWs), with either a 36 Å or a 50 Å well-width and a fixed 43 Å barrier-width [24], grown on top of a 2.5μm-thick GaN layer on a sapphire substrate. Following the formation of the MQW structure, a ~100 nm-thick $Al_{0.1}Ga_{0.9}N$ cap layer was grown on top of the sample. Because of the large piezoelectric constant along the [0001] orientation, a strain-induced piezoelectric field, on the order of 0.5 MV/cm, existed inside the QWs [23]. A room temperature absorption measurement indicated that the bandgaps of the MQWs are ~410 nm (3.02 eV) and ~422 nm (2.92 eV) for the 36 Å and 50 Å well-width samples, respectively. The output of a femtosecond (fs) Ti:Sapphire laser was frequency doubled with a BBO crystal, in order to reach a photon energy well-above the bandgap; it was then used as the optical pump/probe sources. The frequency-doubled pulses had a pulsewidth of 200 fs at a wavelength of 390 nm, as measured by a two-photon-absorption-type autocorrelation with another GaN thin film [25]. The fs pump pulses photo-excited carriers inside the quantum wells. Because of the periodic distribution of the photoexcited carriers, coherent acoustic phonon oscillation with an acoustic wavelength the same as the MQW period thickness (79 or 93 Å) was initiated, due to space-charge-screening of the piezoelectric field [17] and deformation potential couplings [26]. Thus the resultant coherent acoustic phonon oscillations modulated the MQW absorption spectrum through a quantum-confined Franz-Keldysh effect [17,27], which was detected with a weak probe pulse, by measuring the probe transmission change as a function of the probe pulse time-delay.

In order to study the amplification effect, a high intensity UV amplification pulse was directed into the same area, after the initiation of the oscillation, with a controlled time-delay relative to the first pump pulse. The experimental setup used here is exactly the same as that used in the previous coherent acoustic phonon control experiments [20,21]. This second pump (amplification) pulse will not only induce a possible amplification of the already existing coherent acoustic phonon oscillation, but also generate another coherent oscillation that interferes with the original one. This is also the same as in the previous two-pulse coherent control experiments [19-22], where a simple coherent wave interference theory is applicable for its analysis when the excitations were low [20,22]. The overall acoustic phonon oscillations in the MQWs were again detected using a weak probe pulse with variable time-delays.

Figure 1 shows an example of the measured transient transmission change due to the original pump and the following amplification pulses for the 50 Å MQW sample at a pump/probe wavelength of 390 nm. By measuring the reflected and transmitted optical

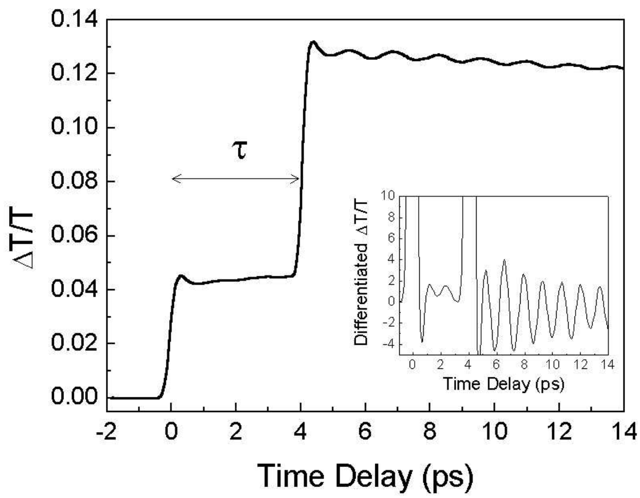

FIG. 1: Transmission change vs. probe time delay, with both pump and amplification pulses in the 50Å InGaN MQW sample. The excitation wavelength was 390 nm. The inset shows the differentiated trace, where the oscillation is more observable.

powers before and after the sample and the focused beam diameter (11 μm, determined by pinholes), the pump- and amplification-pulse-induced 2D carrier densities were estimated to be 9.0×10^{12}cm^{-2} and 11.8×10^{12}cm^{-2}. After the first pump excited the carriers at zero time-delay, a small cosinusoidal acoustic phonon oscillation with a period of 1.38 ps could be observed in the transient transmission measurements, on top of a large positive background due to the photocarrier-induced bandfilling effect. This coherent oscillation was then subjected to interference, and possibly amplified, by the following second pump pulse, with a controlled optical intensity and time-delay τ. For the example shown in Figure 1, the second pulse arrived at τ= 4.2 ps (3.04 oscillation circles) after the zero time-delay and induced another transmission background increase. By changing the time delay τ and the second pulse intensity, we can study the amplitude and phase variation of the initial oscillation as a result of coherent interference. Figures 2(a) and (b) show the amplitude and phase variations of the initial oscillation-induced probe transmission change ΔT/T as a function of the pulse delay τ (from 2.76 ps to 4.14 ps, corresponding to the 2.0 to 3.0 oscillation periods). These amplitude and phase variations were caused by the second pump pulse. Both excitation pulse intensities were kept fixed at those in Figure 1 during the experiment. In order to measure the transmission modulation due to a coherent acoustic phonon oscillation generated by the second pulse itself, we also performed all our experiments with a long time-delay τ (>70 ps). The time-delay was much longer than the acoustic phonon dephasing time (8.8 ps, the time for a coherent acoustic wave to travel out of the MQW region) and the echo time (29 ps, the time for a coherent acoustic wave to travel toward the cap-layer/air interface and be reflected back into the MQW region), so

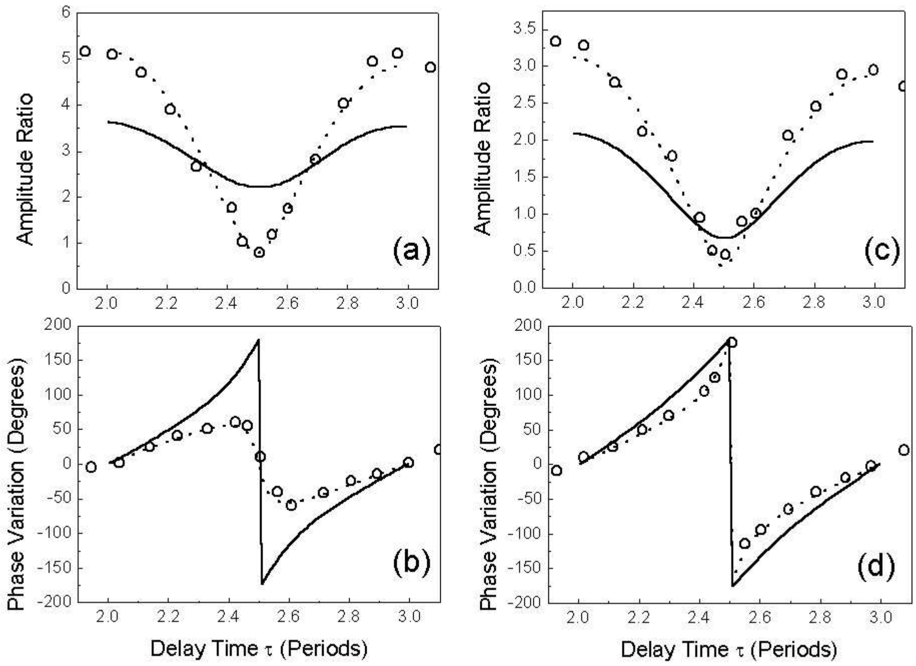

FIG. 2: ΔT/T (a) amplitude and (b) phase variations of the initial oscillation after amplification pulse excitation, as a function of the pulse delay τ (open circles). The pump/amplification pulse induced photoexcited carrier densities were $9.0/11.8\times10^{12}$cm^{-2}. Experiments were performed on the 50Å MQW sample. The solid lines indicate the expected linear interference results. The dotted lines show the fittings, assuming a ΔT/T modulation amplification G=4.0. After calibrating the ΔT/T modulation enhancement due to increased carrier density, the acoustic phonon oscillation (c) amplitude and (d) phase variations of the initial oscillation after amplification pulse excitation, as a function of pulse delay τ (open circles), was obtained. The solid lines in (c) and (d) show the expected linear acoustic interference results. Dotted lines indicate the fittings, assuming a coherent acoustic gain G=1.5 (3.5 dB).

that the residue oscillation effect induced by the first pump pulse can be minimized. Thus we obtained transmission modulation (ΔT/T) amplitudes induced by the second pump pulse which were used in the following fitting. Please note that a previous study on these MQW samples has indicated that the carrier lifetime in the studied samples is longer than 1 ns [24].

For a linear system without acoustic gain and transmission modulation nonlinearity, the final ΔT/T oscillation amplitude is expected to be a linear superposition (with interference) of the transmission modulation of the individual coherent acoustic phonon oscillations induced by two different pulses with $\frac{\Delta T}{T}(t)\Big|_Q = \frac{\Delta T}{T}(t)\Big|_{Q_1} + \frac{\Delta T}{T}(t-\tau)\Big|_{Q_2}$, where $\frac{\Delta T}{T}(t)\Big|_Q$ is the transmission modulation induced by the cosinudoidal coherent acoustic phonon oscillation amplitude Q, which will be defined below. This linear behavior has been observed

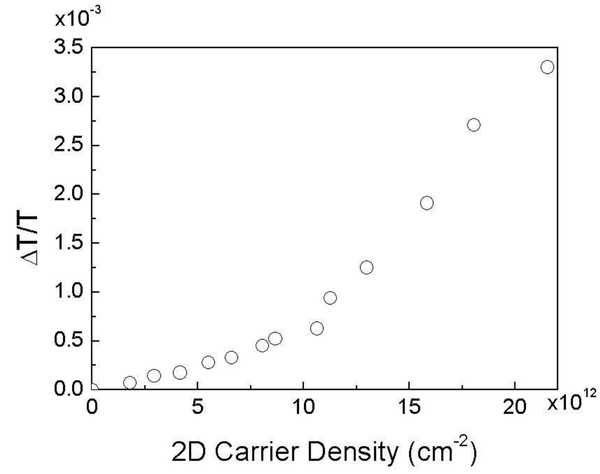

FIG. 3: Oscillation induced transmission change vs. 2D photocarrier density for the 50Å InGaN MQW sample in the single-pump pulse experiments.

in two previous experiments when the excitations were low [19,22]. However this linear model could not explain our observed experimental results when the photo-injected carrier densities are high. For a linear superposition system (shown as the solid lines in Figure 2) with the experimental conditions described above, we would expect a final ΔT/T amplitude ratio smaller than the summation of two independent ΔT/T oscillation amplitudes (due to the 8.8 ps dephasing time), with a time delay τ of 2 oscillation periods (with constructive interference). Our observed results (shown as open circles), however, can be easily explained if we assume there exist a ΔT/T modulation amplification (possibly due to acoustic gain or other effects) for the initial coherent phonon oscillation induced by the second pulse with $\frac{\Delta T}{T}(t)\Big|_Q = \frac{\Delta T}{T}(t)\Big|_{Q_1} G(t-\tau)\exp(i\phi) + \frac{\Delta T}{T}(t-\tau)\Big|_{Q_2}$ for $t > \tau$. A large signal gain, G=4.0 with $\phi = 0$, can be extracted from this specific data through a simple fitting process (dotted lines in Figure 2).

In order to investigate the dynamics of the coherent phonon oscillation itself, we have to know the relation between the measured transmission modulation and the coherent phonon oscillation. This general relation can be expressed as [28]

$$\frac{\Delta T}{T}(t) = \int_0^L F(z)\frac{\partial u}{\partial z}(z,t)dz\,, \tag{1}$$

where $u(z,t)$ represents the displacement field of the acoustic phonon and $F(z)$ is the sensitivity function relating ΔT/T to the strain field $\partial u/\partial z$. A general expression for the sensitivity function of the reflection change in a bulk medium has been derived in Ref. [28]. The sensitivity function reflects the response of the material absorption due to the propagating acoustic field. For a propagating acoustic field, the displacement field can be written as $u(z,t) = Q(t)\exp(iqz + i\theta_0)$, with q being the value of the phonon wave vector in the z direction and $Q(t)$ being the acoustic phonon oscillation amplitude. From this definition, the strain is given by $\partial u/\partial z = iqQ(t)\exp(iqz + \theta_0)$. By substituting these into Eq. (1), and taking the integral over z, we obtain the following relationship between ΔT/T and the phonon amplitude Q:

$$\frac{\Delta T}{T}(t) = \alpha(\omega_{pr}, n_b)Q(t)\,. \tag{2}$$

Here the integration over F gives the coefficient α, which depends on the center angular frequency of the probe pulse ω_{pr} and the background carrier density n_b. A previous study at low excitation energies [22] (with photo-carrier densities on the order of 10^{10} – 10^{11} cm^{-2}) showed a linear relationship between the coherent phonon amplitude and the ΔT/T amplitude. However, with high excitation energy conditions (that is with photo-carrier densities on the order of 10^{13} cm^{-2} as in our interferometric experiments), we found an increase in the ΔT/T amplitude for more photo-excited background carriers, provided the coherent phonon amplitude was the same. Figure 3 shows the oscillation induced transmission change versus 2D photocarrier density for single pump pulse experiments (without the second pump pulse) for the 50 Å MQW sample. When the 2D photocarrier density was lower than $1{\times}10^{13}$ cm^{-2}, a quasi-linear relationship between the observed transmission modulation and the photoexcited carrier density could be observed. However, when the photoexcited carrier density was high, some superlinear relationship was observed, which could be partially attributed to the dependence of α on the n_b in Eq. (2). Consequently, we must calibrate this dependence experimentally, in order to obtain the real phonon oscillation amplitude Q. We used the phonon echo reflected from the surface to calibrate this enhancement. After the initiation of the coherent phonon by the first UV pump pulse, the generated acoustic wave was propagated out of the MQW in two opposite longitudinal directions. One of these acoustic waves was reflected at the interface between the AlGaN cap layer and air. Before this reflected echo returned to the MQW, the carrier density within the MQW was increased by pumping another UV pulse. Since we can fix the amplitude of the echo phonons by the first pump pulse, we can thus study the dependence of the transmission modulation ΔT/T on the MQW carrier density by varying the intensity of the second pump pulse. In particular, with the 2D carrier densities induced by the first and second UV pulses estimated to be $9.0 \times 10^{12}cm^{-2}$ and $11.8 \times 10^{12}cm^{-2}$, which is the same condition as the interferometric trace in Figures 2 (a) and (b), an enhancement factor caused by the increased carrier density can be determined, which is around 2.6. Using this enhancement factor, one can calibrate the ΔT/T modulation amplitude for all the data points in Figure 2 and obtain information about the coherent phonon oscillation. By plotting both the magnitude ratio and the phase variation between the final and the original acoustic oscillations in Figures 2 (c) and (d), we find that we still need an acoustic gain factor in the interferomery analysis. An acoustic gain factor $G = 1.5$ (3.5 dB) and a zero phase shift $\phi = 0$ can thus be obtained (dotted lines in Figures 2 (c) and (d)). This gain factor should be attributed to the nonlinearity of the phonon itself, indicating phonon amplification induced by the second amplification pulse. Our observation also indicates the coherent nature of the acoustic amplification process, due to the zero phase shift $\phi = 0$.

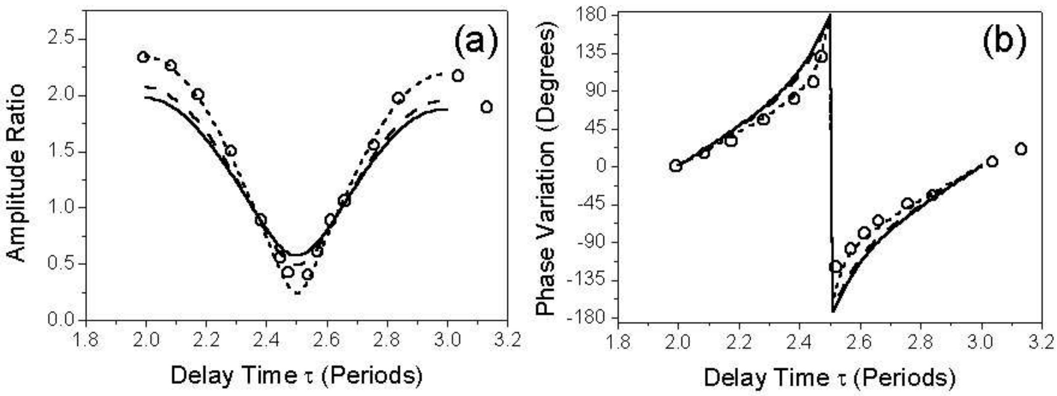

FIG. 4: ΔT/T (a) amplitude and (b) phase variations of the initial oscillation after amplification pulse excitation, as a function of the pulse delay τ (open circles). The pump/amplification pulse induced photoexcited carrier densities were $3.6/5.8\times10^{12}$cm^{-2}. Experiments were performed on the 36Å MQW sample. The solid lines show the expected linear interference results. The dashed lines indicate the fitting results, assuming no acoustic amplification but considering modulation enhancement due to increased carrier density. The dotted lines show the fitting results, assuming a coherent acoustic amplification of G=1.35, after considering the expected modulation enhancement.

Similar phonon amplification nonlinearities are also observed with relatively lower carrier densities or with other samples. Figures 4 (a) and (b) show the amplitude and the phase variations of the initial oscillation-induced ΔT/T, after the excitation of the second pulse as a function of the pulse delay τ (from 2.20 ps to 3.30 ps, corresponding to 2.0 to 3.0 oscillation periods) for the 36 Å QW sample. The experiment was performed at a wavelength of 390 nm, with the 2D carrier densities induced by the first and second UV pulses estimated to be 3.6×10^{12}cm^{-2} and 5.8×10^{12}cm^{-2}, respectively. The observed coherent phonon oscillation period was 1.1 ps [15]. Acoustic echo experiments determined an enhancement factor caused by the increased carrier density due to the second pulse of around 1.1. After considering this enhancement factor, the interferometry analysis still showed an acoustic gain factor $G = 1.35$ (2.6 dB) and a zero phase shift $\phi = 0$ (dotted lines in Figures 4 (a) and (b)). The interferometric fittings, considering only the linear response (solid lines) or only the carrier density enhancement effect (dashed lines), are also provided for comparison.

There are several possible sources for the nonlinear driving force responsible for our observed acoustic gain. After photoexcitation, there might be effective inverted populations in the relaxing electron system. Due to the small acoustic phonon energy, the transition should be intra-subband [29]. Another possible population inversion mechanism would be due to photoinduced longitudinal optical (LO) phonon emission [7,30]. Due to the large piezoelectric field within the well, Cerenkov amplification of the acoustic phonons might also be possible [8,9]. Recently a large electrostrictive coefficient for GaN was determined

[31], which could also create sound amplification in GaN [10,11].

In conclusion, coherent amplification of coherent acoustic phonon oscillations in InGaN MQWs was observed. After initiating the coherent acoustic phonon oscillation in InGaN MQWs with an UV fs pulse, a second UV fs pulse was introduced into the MQWs, to initiate another coherent oscillation. Through an interferometric analysis based on the harmonic oscillator theory, large coherent amplification of the initial acoustic phonon oscillation was observed. It is important to notice that the observed acoustic amplification occurred at room temperature, in contrast to the previous cryogenic temperature experiments [1-4], but a 3.5 dB acoustic gain was observed, which is much higher than previous reported values (2.0 dB [4]).

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