Nano-Ultrasonics and Nano-Acoustics

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Medical Ultrasonics

Resolution limited by Acoustic Wavelength
50MHz, 50-500 μm
Acoustic Controlled Electronic Devices by using SAWs (Surface Acoustic Waves)


Frequency
\~ 1GHz

Wavelength
Several \( \mu \text{m} \)

Acousto-electric Effect: SAW (GHz, \( \mu \text{m} \))
\rightarrow 2D, Slow response time, Micron scale resolution
Nanoultrasonics

- 3D imaging with nano resolution
- THz electronic control with high spatial accuracy (down to a nano scale)
- Require
  - (Coherent) acoustic wave with a nano wavelength
    - Generation
    - Detection
    - Synthesization
    - Propagation control
  - Based on piezoelectric semiconductor
Subjects of this lecture

- Acoustics 101.
- Previous non-piezoelectric works.
- Generation and detection of coherent acoustic phonons (nanoacoustic waves) in piezoelectric multilayers and a single layer.
- Manipulation and optical coherent control of the nanoacoustic waves.
- Study of the nanoacoustic superlattice (phononic bandgap crystal), nanoacoustic cavity, and the supersonic paradox.
- Nanoultrasonics.
- THz electronic control using nano-acoustic waves.
- Nano-acoustic waveguiding.
- Confined acoustic vibrations in nanoparticles.
Subject 1

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Longitudinal Acoustic Wave

- Strain (no unit): \( S = \frac{\partial u}{\partial z} \); u: displacement;
- Stress (force/area): \( T = CS; \) C: elastic constant (area/force);

\[
\begin{bmatrix}
X_x \\
Y_y \\
Z_z \\
Y_z \\
Z_x \\
X_y
\end{bmatrix} = \begin{bmatrix}
c_{11} & c_{12} & c_{13} & c_{14} & c_{15} & c_{16} \\
c_{21} & c_{22} & c_{23} & c_{24} & c_{25} & c_{26} \\
c_{31} & c_{32} & c_{33} & c_{34} & c_{35} & c_{36} \\
c_{41} & c_{42} & c_{43} & c_{44} & c_{45} & c_{46} \\
c_{51} & c_{52} & c_{53} & c_{54} & c_{55} & c_{56} \\
c_{61} & c_{62} & c_{63} & c_{64} & c_{65} & c_{66}
\end{bmatrix} \begin{bmatrix}
e_{xx} \\
e_{yy} \\
e_{zz} \\
e_{yz} \\
e_{zx} \\
e_{xy}
\end{bmatrix}
\]

\[ T = \begin{bmatrix} C \\ S \end{bmatrix} \]
Wave Equation of Stress $T$

(1) \[ \frac{\partial T}{\partial z} = F = \rho_{mo} \dot{u} = \rho_{mo} \dot{v} \quad \nu: \text{velocity; } \rho_{mo}: \text{density of mass}; \]

(2) \[ \frac{\partial \nu}{\partial z} = \frac{\partial S}{\partial t} \quad \text{“Conservation of mass”} \]
\[ \frac{\partial \nu}{\partial z} = \frac{\partial}{\partial z} \left( \frac{\partial u}{\partial t} \right) = \frac{\partial}{\partial t} \left( \frac{\partial u}{\partial z} \right) = \frac{\partial S}{\partial t} \]

From (1) and (2), we have wave eq. of $T$
\[ \frac{\partial^2 T}{\partial z^2} = \rho_{mo} \frac{\partial}{\partial z} \left( \frac{\partial^2 u}{\partial t^2} \right) = \rho_{mo} \frac{\partial^2 S}{\partial t^2} = \frac{\rho_{mo}}{C} \frac{\partial^2 T}{\partial t^2} \]

Assume
\[ T \propto e^{j(\omega t \pm \beta_a z)} \]

We have
\[ \beta_a = \omega \sqrt{\frac{\rho_{m0}}{c}} = \frac{\omega}{V_a}, \quad V_a = \sqrt{\frac{C}{\rho_{m0}}} \quad \text{声速} \]
Energy Density

- Elastic energy density
  \[ W_c = \frac{1}{2} TS = \frac{1}{2} CS^2 = \frac{1}{2C} T^2 \]

- Kinetic energy density
  \[ W_v = \frac{1}{2} \rho_m v^2 \]

Considering the wave propagating in the forward direction:

\[ v = \frac{\partial u}{\partial t} = \frac{\partial u}{\partial z} \frac{\partial z}{\partial t} = SV_a = \frac{V^a}{C} T \]

\[ \rho_m v v^* = \rho_m \frac{V^2}{C^2} TT^* = \frac{1}{C} TT^* \quad V_a = \sqrt{\frac{C}{\rho_m}} \]

\[ \therefore W_c = W_v \]
Acoustic Impedance

\[
Z_a \equiv \frac{T}{v} \quad \Leftrightarrow \quad Z_{EM} = \frac{E_y}{H_x}
\]

- Note when the propagation direction is reversed, the acoustic impedance is different in sign.
- Consider forward propagation

\[
Z_F = -\frac{T_F}{v_F} = \frac{C}{V_a} = \sqrt{\rho_{m0}C} = V_a\rho_{m0} \equiv Z_0
\]

Reflection of acoustic waves from interface

\[
\Gamma = \frac{Z_{02} - Z_{01}}{Z_{02} + Z_{01}}
\]

medium 1 \quad medium 2
Elastic waves in anisotropic solids

General wave equation in solids
\[ \rho \frac{\partial^2 u_i}{\partial t^2} = c_{ijkl} \frac{\partial^2 u_l}{\partial x_j \partial x_k} \]
\[ c: \text{stiffness tensor (fourth-rank)} \]

with the general solution:
\[ u_i = \dot{u}_i F(t - \frac{\tilde{n} \cdot \tilde{x}}{V}) \]

\[ \rho V^2 \dot{u}_i = c_{ijkl} n_j n_k \dot{u}_i \]  \hspace{1cm} (Christoffel’s equation)

Introducing a second-rank tensor
\[ \Gamma_{il} = c_{ijkl} n_j n_k \]

\[ \Gamma_{il} \dot{u}_l = \rho V^2 \dot{u}_i \]

\[ \left| \Gamma_{il} - \rho V^2 \delta_{il} \right| = 0 \]  \hspace{1cm} One longitudinal and two transverse modes for a specific propagation direction
Energy propagation in anisotropic solids

In general, energy velocity is not equal to the phase velocity

Slowness surface
(for a cubic structure)

*Royer and Dieulesaint, Elastic waves in Solid I
Reflection and transmission

The continuity of displacement and tractions requires that

\[
\begin{align*}
    u_i^I + \sum_R u_i^R &= \sum_T u_i^T \\
    T_i^I + \sum_R T_i^R &= \sum_T T_i^T
\end{align*}
\]

The projections of the reflected and refracted wave vectors on to the interface are equal to that of the incident waves.

*Royer and Dieulesaint, *Elastic waves in Solid I*
Nano-Acoustics

\[ V_a = f \cdot \lambda \]

\[ \lambda = 10 \text{ nm} \]
\[ V_a = 8000 \text{ m/s} \]
\[ f = 0.8 \text{ THz} \]
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- Confined acoustic vibrations in nanoparticles.
Coherent Phonon Generation and Detection by Picosecond Light Pulses

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and

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(Received 18 June 1984)

Using the picosecond pump and probe technique we have detected oscillations of photoinduced transmission and reflection in thin films of $\alpha$-As$_2$Te$_3$ and cis-polyacetylene. These oscillations are due to the generation and propagation of coherent acoustic phonons in the film. We discuss the generation and detection mechanism, and we use this effect to measure the sound velocity in a film of $\alpha$-SiO$_2$.

PACS numbers: 63.50.+x, 78.20.Hp

FIG. 1. Photinduced transmission in $\alpha$-As$_2$Te$_3$ for films of different thickness at room temperature.

FIG. 3. Experiment to measure the sound velocity in an amorphous SiO$_2$ film. Curve $a$ shows data for $\Delta T/T$. Curve $b$ is a computer fit.
The central frequency of acoustic pulses: 

\[ f_a = \frac{V_s}{2t} \]

\( V \): sound velocity 
\( t \): thickness of the metal film 

The oscillation frequency of reflection change: 

\[ f_o = \frac{2nV_m \cos \theta}{\lambda} \]

\( n \): index of refraction 
\( \theta \): incident angle in medium 
\( \lambda \): vacuum wavelength of light

Experimental Setup and Sample Structures

Pulse train (~150 fs)

Probe (400nm)

Pump (800nm)

Sample

Delay Stage

Detector

Cover glass

DI water

2-40nm gold film

Cover glass

Typical Measured Results

- For a gold thickness of 15nm, corresponding to an acoustic central frequency of ~108 GHz
- At an optical probe wavelength of 400 nm
Coherent Oscillation of Zone-Folded Phonon Modes in GaAs-AlAs Superlattices

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(Received 30 March 1994)

We have observed a coherent oscillation of zone-folded acoustic phonons in GaAs-AlAs superlattices for the first time by means of a femtosecond time-resolved pump-probe technique. The oscillatory component in the time-resolved reflection signal corresponds to the upper branch ($B_2$ symmetry) of the first-order doublet mode. Carriers were selectively excited in well layers and the $B_2$-symmetry phonon mode was selectively generated. The dephasing time of the $B_2$ mode was measured to be 70 ps from our experiments.

PACS numbers: 78.47.+p, 42.65.Re, 63.20.–c, 78.66.Fd

FIG. 1. Time derivative traces of the reflectivity change at a laser wavelength of 742 nm. (a) and (b) show the temporal traces for samples 1 and 2, respectively.

FIG. 3. Fourier power spectra of the temporal traces shown in Fig. 1. The insets show phonon dispersions of the two samples based on the Rytov model, where $\theta_{\text{max}}$ is the zone-edge wave vector $\pi/d$ and $d$ is the period of the superlattices. The open circles of the insets indicate the first-order doublet peaks obtained through Raman scattering experiments at an excitation wavelength 514.5 nm and the solid circles indicate the peak frequencies of the power spectra.
Ultrafast Acoustic Phonon Ballistics in Semiconductor Heterostructures

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(Received 16 September 1996)

Using a two-color ultrafast surface deflection spectroscopy, we demonstrate the time-resolved observation of acoustic phonon wave packets emitted from a single buried GaAs quantum well. A longitudinal acoustic phonon pulse is generated at a preselected depth within a highly confined region (≥10 nm thick) through the coherent nonequilibrium deformation potential which was previously unobserved in such structures. Subsequent detection with subpicosecond resolution at the surface resolves propagating high-wave-vector ballistic phonons and quasiballistic or “snake” phonons which subsequently merge into a quasidiffusive phonon pulse. [S0031-9007(97)03074-3]

FIG. 1. (a) Vertical structure of wafers A, B, and C. (b) Schematic of the experimental arrangement (F = filter, BS = beam splitter, T = 300 K). (c) Calculated strain pulse dynamics after the QW is permanently stressed.

FIG. 2. Typical time-resolved deflection signals from sample A: (a) Probe beam aligned to one side of the pump focus. (b) Probe aligned to the other side—the signal is inverted. (c) An infrared probe pulse is used. Nothing is seen, even in the residual, after subtracting a fit to the dominant relaxation.
Coherent Acoustic Phonons in a Semiconductor Quantum Dot

Todd D. Krauss and Frank W. Wise

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(Received 10 July 1997)

Coherent acoustic phonons in PbS quantum dots are observed using femtosecond optical techniques. This is the first observation of coherent acoustic phonons in a semiconductor quantum dot; the phonons are generated through the deformation-potential coupling to the quantum-dot exciton. The acoustic modes are weakly damped, and we also find extremely weak coupling (g ~ 0.01) to the optical modes. These conclusions have important consequences for the vibronic nature of the exciton transition in the quantum dot and its dephasing. [S0031-9007(97)04822-9]

PACS numbers: 73.20.Dx, 63.22.+m, 78.47.+p

FIG. 2. The main figure shows a transient saturated-absorption signal. The inset shows a three pulse photon echo signal as a function of delay between the first and third pulses, the delay for which the signal best exhibits the consequences of exciton-phonon coupling. The solid line is experimental data and the dotted line is a theoretical fit using the standard analysis as in Ref. [6]. The exponential decay due to recovery of the exciton population has been subtracted.
Subject 3

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Strain induced distortion of atomic positions can lead to a net dipole moment and generate polarization in a material. This effect is called the piezoelectric effect and can be exploited for strain sensors, built-in electric field and/or charge generation in devices, etc.

\[
\begin{bmatrix}
P_x \\
P_y \\
P_z
\end{bmatrix} =
\begin{bmatrix}
e_{11} & e_{12} & e_{13} & e_{14} & e_{15} & e_{16} \\
e_{21} & e_{22} & e_{23} & e_{24} & e_{25} & e_{26} \\
e_{31} & e_{32} & e_{33} & e_{34} & e_{35} & e_{36}
\end{bmatrix}
\begin{bmatrix}
\varepsilon_{xx} \\
\varepsilon_{yy} \\
\varepsilon_{zz} \\
\varepsilon_{yz} \\
\varepsilon_{xz} \\
\varepsilon_{xy}
\end{bmatrix}
\]

\( e \): piezoelectric coefficients \([\text{C/m}^2]\)
Constitution Equations for Piezoelectric Semiconductors

\[ T = C \ S - e \ E + d_e n_e + d_h n_h \]

\[ D = \varepsilon \ E + e \ S \]

\( T \): Stress, \( S = du/dz \): Strain,
\( D \): Electrical Displacement,
\( E \): Electric Field,
\( C, \varepsilon, e \): Elastic Stiffness, dielectric, and piezoelectric Constant,
\( d_e, d_h \): Deformation Potential Coupling Constant.
\( n_e, n_h \): Electron and Hole Carrier Densities.
Wave Equation for $u$

\[
\rho \frac{\partial^2 u}{\partial t^2} = \frac{\partial T}{\partial z}
\]

\[
T = C S - e E + d_e n_e + d_h n_h
\]

\[
\rho \frac{\partial^2 u}{\partial t^2} = \frac{\partial T}{\partial z} = C \frac{\partial S}{\partial z} - e \frac{\partial E}{\partial z} + d_e \frac{\partial n_e}{\partial z} + d_h \frac{\partial n_h}{\partial z}
\]

\[
S = \frac{\partial u}{\partial z}
\]

\[
\rho \frac{\partial^2 u}{\partial t^2} - C \frac{\partial^2 u}{\partial z^2} = e \frac{\partial E}{\partial z} + d_e \frac{\partial n_e}{\partial z} + d_h \frac{\partial n_h}{\partial z} = f_{pz} + f_{de}
\]
Piezoelectric multi- or single nanolayers

Equilibrium state before laser pumping

\[ T = 0, \ D = 0, \ n_e = n_h = 0 \]

\[ S_1 = (a_s - a)/a \]

Potential Wells for Electrons and Holes

No piezo-effect

With piezo-effect
Generation of NanoAcoustic Waves

- Piezoelectric field in $\text{In}_{0.1}\text{GaN}/\text{GaN}$ MQW

  \[ \Delta E_C = 0.38 \text{ V} \]
  \[ \Delta E_V = 0.12 \text{ V} \]

  GaN-barrier 4.3 nm
  InGaN-well 3.6 nm

- Built-in piezoelectric field: $\sim 350 \text{ kV/cm}$

Generation of NanoAcoustic Waves

Spatial Distribution of Photocarriers

Governing Equations with the loaded string model

\[ \rho \frac{\partial^2 u}{\partial t^2} = f = \frac{\partial T}{\partial z} \]

\[ \nabla \cdot \mathbf{D} = \rho_{sc} \Rightarrow \frac{\partial D}{\partial z} = \rho_{sc} \]

Loaded String Equation

\[ \rho \frac{\partial^2 u}{\partial t^2} - \hat{C} \frac{\partial^2 u}{\partial z^2} = f_{PZ} + f_{\text{Deform}} \]
The Driving Forces:

Piezoelectric:

$$f_{PZ} = \frac{e_{33}}{\varepsilon_3} \rho_{sc} = \frac{e_{33}e}{\varepsilon_3} (n_h - n_e)$$

Deformation Coupling:

$$f_{\text{Deform}} = d_e \frac{\partial n_e}{\partial z} + d_h \frac{\partial n_h}{\partial z}$$
Displacement Excitation of Coherent Acoustic Phonons with a New Equilibrium Configuration

New Equilibrium

Dephasing or traveling out

\[ \frac{\partial^2 u}{\partial t^2} = 0 \]

\[ \frac{\partial^2 u_{eq}}{\partial z^2} = -\frac{1}{\hat{C}} \left( f_{PZ} + f_{Deform} \right) \]

The acoustic wavelength is determined by the MQW period!

Space-Time Evolution of Coherent Acoustic Waves
QM version: Harmonic Oscillator Model

\[ \frac{\partial^2 Q}{\partial t^2} + 2\gamma \frac{\partial Q}{\partial t} + \omega_0^2 Q = \frac{F_1(t)}{m} \]

\[ Q_1(t) = \theta(t) \left[ 1 - e^{-\gamma t} \left( \cos(\Omega_q t) + \frac{\gamma}{\Omega_q} \sin(\Omega_q t) \right) \right] \]

\[ Q_1(t) = \theta(t) \left[ 1 - \cos(\Omega_q t) \right] \quad (\gamma \approx 0) \]

Cosinusoidal Oscillation!
Detection of coherent acoustic phonons (nanoacoustic waves) in piezoelectric multilayers and a single layer:

Quantum Confined Franz-Keldish Effect

\[
\left( \frac{\Delta T}{T} \right)_{NAW}(t) = \int_{-\infty}^{\infty} dz \Delta \alpha(z) = \int_{-\infty}^{\infty} dz s(z, t) \cdot F(z; \omega)
\]

strain wave function
sensitivity function

MQW-based OPT

\[ \Delta t = t_0 \quad \Delta t = t_1 \]

\[(\Delta T/T)_{NAW} \]


Time Delay (\(\Delta t\))
Optical Piezoelectric Transducer (OPT)

- Generation of nano-acoustic-waves (NAW)

Photocarriers excited in the well

Piezoelectric field being screened

Strain changed (piezoelectric effect)

Generation of NAWs

InGaN/GaN MQWs

Detection of NAW with OPT

- Strain pulses propagating through wells
- Strength of the piezoelectric field changed
- Optical absorption changed (Quantum Confined Franz-Keldysh effect)
- Detected through the transmission change of the optical probe pulse

Generation of Nano-Acoustic Waves

Femtosecond Optical Pulse

Barrier Well Barrier

Femtosecond Optical Pulse

Valence Band
THz Acoustic Sensor

\[
\left( \frac{\Delta T}{T} \right)_{NAW}
\]
Case #1
Superlattice OPT

- Well width: 12, 25, 36, 50, 62 Å
- Barrier width: 43 Å
- In composition 10%

Cosinusoidal Oscillation!

λ = 390 nm

λ = 365 nm

Pump Energy: 0.2nJ
## Effect of Period

14 Periods

<table>
<thead>
<tr>
<th></th>
<th>barrier (nm)</th>
<th>well (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>sample A</td>
<td>4.3</td>
<td>3.6</td>
</tr>
<tr>
<td>sample B</td>
<td>4.3</td>
<td>5.0</td>
</tr>
<tr>
<td>sample C</td>
<td>4.3</td>
<td>6.2</td>
</tr>
<tr>
<td>sample D</td>
<td>9.0</td>
<td>2.2</td>
</tr>
<tr>
<td>sample E</td>
<td>7.0</td>
<td>2.2</td>
</tr>
<tr>
<td>sample F</td>
<td>3.0</td>
<td>2.2</td>
</tr>
</tbody>
</table>

Unit: nm

---

### Case #1

Superlattice OPT

Differential $\Delta T/T$ (a.u.) vs. Time Delay (ps)

Sample A, B, C, D, E, F
L-Acoustic Phonon Dispersion Curve of InGaN/GaN Superlattices

\[ \omega = v q \]


sound velocity of 7350 m/s
Case #2: Single Quantum Well OPT

2.9 nm In$_{0.2}$GaN SQW OPT

Object Layer

Attenuation Layer

Broadband Acoustic Wave + Broadband Sensor

Graph showing the change in transmission ($\Delta T/T$) over time delay (ps).
Case #3: MQW Sensing of Picosecond Ultrasonic Pulses

<table>
<thead>
<tr>
<th>Material</th>
<th>Layer Type</th>
<th>Thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>transducer</td>
<td>2 nm</td>
</tr>
<tr>
<td>GaN</td>
<td>cap</td>
<td>17 nm</td>
</tr>
<tr>
<td>In$<em>{0.23}$Ga$</em>{0.77}$N</td>
<td>well</td>
<td>2.9 nm</td>
</tr>
<tr>
<td>GaN</td>
<td>barrier</td>
<td>21.7 nm</td>
</tr>
<tr>
<td>GaN</td>
<td></td>
<td>3.4 µm</td>
</tr>
<tr>
<td>Sapphire</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Transient reflectivity

FFT of Transient reflectivity

Backward Brillouin Scattering (Dynamic Fabry-Perot Effect)
Case #3: MQW Sensing of Picosecond Ultrasonic Pulses

Transient transmission changes

FFT of Transient transmission changes

Dynamic Fabry-Perot Effect

\[ \Lambda = 24.6 \text{nm} \]

Transient transmission change with a notch filter

\[ \Lambda = \frac{\Lambda}{2} \]
Case #4: Design of the Acoustic Waveform

Can we design the frequency component of the nano acoustic waves by controlling the quantum well structures?
Asymmetric Multiple Quantum Well OPT

Barrier Periodic MQWs

Well

Periodic Length

Duty Ratio = \( \frac{\text{Well Width}}{\text{Periodic Length}} \)

carrier-induced strain

\[
\begin{align*}
\text{Strain } s_3(q) & (\text{A.U.}) \\
\text{Spatial frequency } q/q_0 & \\
\text{FFT}
\end{align*}
\]
Asymmetric Multiple Quantum Well OPT

InGaN/GaN MQW, Well: 22Å Barrier: 90Å

Amplitude Ratio = \frac{\text{Second Mode Amplitude}}{\text{Fundamental Mode Amplitude}}

Duty Ratio = \frac{\text{Well Width}}{\text{Periodic Length}}
Due to the boundary condition that strain must be continuous at the interface, the strain of NAW will experience a 180-degree sign change after being reflected at a free-end interface.

**Case #5: Superlattice with a surface layer**

- InGaN/GaN MQW
- Acoustic Mirror (Air-GaN interface)
Uni-Directional OPT

InGaN well : 2.9nm
GaN barrier : 21.7nm
Surface GaN layer: 4.7 nm

FFT

Spectrum (a.u.)
Frequency (THz)

0.0 0.2 0.4 0.6 0.8 1.0 1.2 1.4

Differential ΔT/T
Time Delay (ps)

0 10 20 30 40 50

Surface Layer Effect

InGaN well: 2.9 nm
GaN barrier: 13.0 nm
Surface GaN layer: 7.9 nm

FFT


Wave Output

Time Delay (ps)

Differential $\Delta T/T$

Spectrum (a.u.)

Frequency (THz)

Cap Layer width of $\sim 0.6\lambda$

$Z_{\text{GaN}} > Z_{\text{InGaN}}$

$\sim \lambda/2$ GaN surface layer
Surface Layer Effect:

Experimental result

Simulated result $0.6\lambda$

210-degree phase change at 16.5 ps

240-degree phase change at 13 ps
Surface Effect: Surface Roughness

Surface Roughness: 1.3 nm

rms of measured surface roughness: 0.77-2.63 nm
Subject 4

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Coherent control of acoustic phonon oscillation

![Graph showing the relationship between pump and control with probe delay. The graph includes a label for the probe delay in ps and a range of ΔT/T values from 0 to 0.20. There is a note indicating the pump and control actions with corresponding values for λ = 395 nm and a material composition of 50/43 Å InGaN MQW.](image-url)
Coherent phase control

Amplitude control

$\Delta T/T$

$\lambda = 395\text{ nm}$

$62\text{Å} \text{ InGaN MQW sample}$

Trace d phase shift $+135^\circ$

Trace c phase shift $+35^\circ$

Trace b phase shift $-20^\circ$

Trace c coherent amplification

Trace b coherent cancellation
Concepts of Waveform Synthesis

Design a multiple frequency component acoustic waveform pattern

Manipulate the spectral composition of the waves by optical coherent control

Synthesize the temporal waveform
Second Harmonic Amplification via Coherent Control

22Å / 70Å
InGaN/GaN MQW
Acoustic Frequency Tuned by Optical Control Technique with SQW OPT

Femtosecond Optical Pulses with controlled time delay

Structure of OPT: 2.9 nm $\text{In}_{0.2}\text{GaN}$ SQW

Acoustic Waveform Synthesization by the Optical Control Technique with a SQW OPT

Structure of OPT: 7 nm In$_{0.2}$Ga$_{0.8}$N SQW

Subject 5

- Acoustics 101.
- Previous non-piezoelectric works.
- Generation and detection of coherent acoustic phonons (nanoacoustic waves) in piezoelectric multilayers and single layer.
- Manipulation and optical coherent control of the nanoacoustic waves
- Study of the nanoacoustic superlattice (phononic bandgap crystal), nanoacoustic cavity, and the supersonic paradox.
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- THz electronic control using nano-acoustic waves.
- Nano-acoustic waveguiding.
- Confined acoustic vibrations in nanoparticles.
Study of nanoacoustic superlattice
(1D phononic bandgap crystal)

- **Raman Scattering**
  For example, Phys. Rev. Lett. 89, 227402 (2002).

- **Superconductor Bolometer**
Measuring the Sub-THz reflection waveform of a nanoacoustic superlattice (phononic bandgap)

- 7 nm $\text{In}_{0.2}\text{GaN}$ SQW
- Optical Piezoelectric Transducer (OPT)
- Phononic Bandgap Nano-Crystal

Graph showing:
- Frequency (THz) vs. Wavevector ($\text{nm}^{-1}$)

Layer structure:
- GaN (600nm)
- GaN (240nm)
- $\text{17 X 8nm/8nm Al}_{0.7}\text{GaN/Al}_{0.2}\text{GaN}$
- Buffer Layer $\text{Al}_{0.63}\text{GaN}$ (1.2 $\mu$m)
- Sapphire (>200 $\mu$m)
Nanoacoustic wave propagation in a cavity

Frequency: 280 GHz

Time: 1 ps
Time: 90 ps
Time: 30 ps
Time: 60 ps
Time: 200 ps
Time: 175 ps
Time: 148 ps
Time: 120 ps

In$_{0.2}$GaN SQW
Superlattice (Output Coupler)
Reflection Transfer Function of the First Gap

Strain Reflectivity vs. Acoustic Phonon Frequency (THz)

Phase (radian) vs. Wavevector (nm$^{-1}$)
Superluminal Paradox

Phys. Rev. Lett. 73, 2308 (1994)
H. Winful, PRL 90, 023203 (2003)

Constant Reflection Time!!

Constant Tunneling Time!!

Velocity = Length/Time
Superluminal??
Nature of “Superluminal” Barrier Tunneling

Herbert G. Winful

Department of Electrical Engineering and Computer Science, University of Michigan, 1301 Beal Avenue, Ann Arbor, Michigan 48109-2122
(Received 27 August 2002; published 17 January 2003)

We show that the distortionless tunneling of electromagnetic pulses through a barrier is a quasistatic process in which the slowly varying envelope of the incident pulse modulates the amplitude of a standing wave. For pulses longer than the barrier width, the barrier acts as a lumped element with respect to the pulse envelope. The envelopes of the transmitted and reflected fields can adiabatically follow the incident pulse with only a small delay that originates from energy storage. The theory presented here provides a physical explanation of the tunneling process and resolves the mystery of apparent superluminality.

Solution: A pulse much shorter than the barrier thickness
Superluminal Paradox

H. Winful, PRL 90, 023203 (2003)

\[ R_{\text{bandgap}}(\omega) e^{i\phi} \]

Phase Time = \(-d\phi/d\omega\)

Phase Time is not necessarily the energy propagation time !!!

Our Measured
Phase Time = 16.4 ps
Comparison between Finite Difference Time Domain (FDTD) Simulation and Experimental Results

Propagation Round Trip Time

Phase Time of 16.4 ps?

Phase Time is not necessarily the energy propagation time (with an operational definition) !!!
Subject 6

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Nanoultrasonic Imaging

Diagram showing a sample pattern with laser pulses for excitation and detection, as well as quantum wells.
Nano seismology

2.9 nm In$_{0.2}$GaN SQW

OPT

Broadband Acoustic Pulse + Broadband Acoustic Sensor

Object Layer

Attenuation Layer

$\Delta T/T$

Time Delay (ps)

$\Delta T/T$

Time Delay (ps)
1D Nanoultrasonic Scan (A-Scan)

\[ \text{In}_{0.2}\text{GaN/ GaN MQW} \]

2.9nm/13.0nm

\[ \Delta \frac{T}{T} \]

**Acoustic Pulse with a Central Frequency 0.5 THz + 0.5THz Acoustic Sensor**

- **Objective Layer**
  - GaN (~80 nm)
- **Attenuation Layer**
  - 3 InGaN/GaN OPT

Nanoultrasonic A-Scan: Thickness of a GaN Test Layer

$\text{In}_{0.2}\text{GaN}$/ GaN MQW

2.9nm/ 13.0nm

Objective Layer
GaN (~80 nm)

3 $\text{InGaN}$/$\text{GaN}$ OPT

Attenuation Layer


Echo Time : 25.4 ps

Acoustic Velocity : 8020 m/s

Thickness of the objective layer : 84 nm

Accuracy of the Measured Thickness : <1nm
2D Nanoultrasonic Scan (B-Scan)

Objective Layer

GaN Attenuation Layer (3.4 μm)

Sapphire

OPT: 7/7nm In_{0.2}Ga_{0.8}N/GaN MQW

Sample A: SiO_2 ~30 nm
Sample B: SiO_2 ~15 nm
Sample C: SiO_2 0 nm
2D Nanoultrasonic Scan (B-Scan)

Strain reflectivity of GaN/SiO₂ : 0.58
1.5 period of the striped pattern
X-axis sampling rate: 200nm/step
2D Nanoultrasonic Scan (B-Scan)

25.6 ps (GaN: 85 nm)

12.8 ps (SiO₂ : 38 nm)
2D Nanoultrasonic Scan (B-Scan)

Echo from GaN/SiO₂

Echo from GaN/SiO₂

GaN
2D Nanoultrasonic Scan (B-Scan)

- Time Delay (ps):
  - 27.3 ps (GaN: 89 nm)
  - 6.0 ps (SiO$_2$: 18 nm)
Transverse Resolution of a 2D Nanoultrasonic Scan (B-Scan)

- 350 nm (Optical spot size with an objective with NA of 0.85; $\lambda=400\text{nm}$)

To Improve the Transverse Resolution....

- 250 nm (Optical spot size with an objective with NA of 1.3; $\lambda=400\text{nm}$)
- Oversampling and signal processing (Deconvolution)
- Near field (Surface only)
- A far field technique with transverse manipulation
2D Nanoultrasonic Scan: with a fine step size

Step Size: 50 nm

GaN

12 μm
5 μm
2D Nanoultrasonic Scan: signal processing

- Cliff-like Edge Analysis

AIR-GaN surface

![Graph showing echo signal vs time](image)
Processed 2D Nanoultrasonic Scan:
A comparison with surface-only AFM measurements

Transverse Resolution of 25 nm fitted by Gaussian spot

AFM measurement was performed by 台大應力所李世光教授、林鼎政
Carrier Screening and Saturation of the Piezoelectric Force

2D carrier Density: $10^{12}$ cm$^{-2}$

2D carrier Density: $10^{13}$ cm$^{-2}$


銘傳大學賴志明教授協助計算
Nonlinear saturation imaging for improved lateral resolution without near-field optics

Background carrier manipulation with a pre-pulse

Saturation Pulse

Pump Pulse

Quantum Wells

Laser Pulses For Excitation

Nanoacoustic Waves

Designed Distribution of Pre-injecting carriers
Nonlinear saturation imaging for improved lateral resolution without near-field optics

Acoustic Spot

- without saturation pre-pulse
- with saturation pre-pulse
Estimation of Acoustic Spot Sizes

[Diagram showing the interaction between optical pump and probe pulses and resulting EW2 ratio over different scanned positions.]
Estimation of Acoustic Spot Sizes

No Saturated Pulses

EW-1

EW-2

With Saturated Pulses

EW-1

EW-2
Estimation of Acoustic Spot Sizes

No Saturated Pulses

With Saturated Pulses
Nanoacoustic Spot Size < Optical Diffraction Limited

- Objective NA: 0.85; $\lambda=400\text{nm}$

<table>
<thead>
<tr>
<th>No Saturated Pulses</th>
<th>With Saturated Pulses</th>
</tr>
</thead>
<tbody>
<tr>
<td>300 nm</td>
<td>140 nm</td>
</tr>
</tbody>
</table>

$\lambda/2$
Subject 7

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- Confined acoustic vibrations in nanoparticles.
Acoustic propagation in waveguides

- Confinement of propagating strain energy
- Waveguide modes and acoustic dispersion

Pochhammer-Chree theory

\[
\begin{align*}
\frac{2p}{a} (q^2 + k^2) J_1(pa)J_1(qa) &- (q^2 - k^2) J_0(pa)J_1(qa) \\
-4k^2 pq J_1(pa)J_0(qa) &= 0
\end{align*}
\]

http://140.124.30.80/LUT/ugw.html
Nano-acoustic waves in nano-acoustic waveguides

- Nano-acoustic waveguides
  - semiconductor nanorods: GaN, Si, ZnO, …

Applications:
1. studies on nonlinear acoustics by dispersion engineering
2. Improvement in lateral resolution of nano-ultrasonic imagings

Material dispersion

Waveguide dispersion

Controlled by rod diameter
Subject 8

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- THz electronic control using nano-acoustic waves.
- Nano-acoustic waveguiding.
- Confined acoustic vibrations in nanoparticles.
Quantization of Energy in Low Dimensional System

- Spatial confinement result in quantization of
  - Electronic energy
    - Particle in 3D-box
  - Acoustic energy
    - Zero stress boundary condition
    - Zero strain boundary condition

![Absorption vs Wavelength graph](image-url)

- 4.3 nm CdSe
- Wavelength (nm)
- Absorption

```
300 400 500 600 700 800
0.0 0.2 0.4 0.6 0.8 1.0
```
Types of Eigenmodes

- **Free homogeneous sphere, Lamb 1882**
  - Two classes of normal modes in spherical coordinate
    - Torsional (TOR) modes
      - Divergence of displacement is zero
    - Spheroidal (SPH) modes
      - Angular momentum
        - $l=0$ breathing modes
        - $l=1$ dipolar modes
        - $l=2$ quadrapolar modes

Motional Pattern of SPH Modes

$l=0$
Pure radial mode
One node

$l=1$
Two nodes

$l=2$
Football modes
Four nodes
Interaction of Photons with Confined Acoustic Phonons

- **Selection Rules**
  - Only SPH modes can interact with photons
    - Change density and hyperpolarizability
  - Types of interaction (Just like chemical molecules)
    - Inelastic light scattering
    - Photon absorption

- These interaction can be used to reveal the excitation of confined acoustic phonons

First Observation of Confined Acoustic Modes

- MgCr$_2$O$_4$-MgAl$_2$O$_4$ nucleation in glass

Vibration Eigenmodes and Size of Microcrystallites in Glass: Observation by Very-Low-Frequency Raman Scattering

E. Duval, A. Boukenter, and B. Champagnon

Physico-Chimie des Matériaux Luminescents, Campus la Doua, Université Lyon I, 69622 Villeurbanne, France
(Received 26 February 1986)

The observation of very-low-frequency bands by Raman scattering in a nucleated cordierite glass is described. The frequency of the maximum of scattering is proportional to the inverse diameter of the particles, which are spherical spinel microcrystallites. It is shown that vibrational surface modes of particles are responsible for this Raman scattering.

PACS numbers: 61.40.+b, 68.35.Ja, 78.30.—j, 81.20.Qf

Strain free condition: due to matrix
Traces Observe (SPH, l=0) Breathing Modes
Observe Confined Acoustic Modes by Pump-probe Measurement

- 3nm PbS quantum dot

Coherent Acoustic Phonons in a Semiconductor Quantum Dot

Todd D. Krauss and Frank W. Wise
Department of Applied Physics, Cornell University, Ithaca, New York 14853
(Received 10 July 1997)

Coherent acoustic phonons in PbS quantum dots are observed using femtosecond optical techniques. This is the first observation of coherent acoustic phonons in a semiconductor quantum dot; the phonons are generated through the deformation-potential coupling to the quantum-dot exciton. The acoustic modes are weakly damped, and we also find extremely weak coupling (S ∼ 0.01) to the optical modes. These conclusions have important consequences for the vibronic nature of the exciton transition in the quantum dot and its dephasing. [S0031-9007(97)04822-9]

PACS numbers: 73.20.Dx, 63.22.+m, 78.47.+p
Pump-probe Traces

$l=0$
Observe Confined Acoustic Modes by Brillouin Scattering

Several hundred nanometer SiO$_2$ sphere

Brillouin Study of the Quantization of Acoustic Modes in Nanospheres

M. H. Kuok,*, H. S. Lim, S. C. Ng, N. N. Liu, and Z. K. Wang
Department of Physics, National University of Singapore, Singapore 117542, Republic of Singapore
(Received 3 January 2003; published 24 June 2003)

The vibrational modes in three-dimensional ordered arrays of unembedded SiO$_2$ nanospheres have been studied by Brillouin light scattering. Multiple distinct Brillouin peaks are observed whose frequencies are found to be inversely proportional to the diameter ($\approx$ 200–340 nm) of the nanospheres, in agreement with Lamb's theory. This is the first Brillouin observation of acoustic mode quantization in a nanoparticle arising from spatial confinement. The distinct spectral peaks measured afford an unambiguous assignment of seven surface and inner acoustic modes. Interestingly, the relative intensities and polarization dependence of the Brillouin spectrum do not agree with the predictions made for Raman scattering.

DOI: 10.1103/PhysRevLett.90.255502 PACS numbers: 63.22.+n, 62.30.+d, 62.65.+k, 78.35.+c

Stress-free condition
Also Observe (SPH, ℓ=2)
In Brief

- Previous works focus on the observation of (SPH, \(l=0\)) and (SPH, \(l=2\)) modes
  - Inelastic light scattering sees (SPH, \(l=0\)) and (SPH, \(l=2\)) modes
  - Pump-probe measurement sees (SPH, \(l=0\)) modes
- Study of (SPH, \(l=1\)) is rare
Selection Rules

\( l = 0 \)

Raman active

\( l = 2 \)

\( l = 1 \)

THz photon absorption

Challenges to Observe $l=1$ Modes

- Uniform size of nanosphere
  - Otherwise smeared by inhomogeneous broadening
- Large acoustic impedance contrast to matrix
  - Otherwise smeared by homogeneous broadening
- Low dielectric background absorption
  - Quadratic dependence on frequency
- Long-lived spatial charge distribution
  - For the change of dipole-moment
  - Efficient Photon to phonon coupling
Observation of (SPH, l=1) Modes

- First experimental examination
  - Fourier Transform Infrared (FTIR) Spectroscopy on TiO₂ nanopowders*
  - Suspect the required charge separation originate from the water molecule adsorption
    - Lack direct proof

- Can we make a specific charge separation on a semiconductor nanocrystal to prove the observation of (SPH, l=1)?

Murray et al. J. Nanoelectronics and optoelectronics 1, 92-98 (2006)
Resonant Absorption by \((l=1)\) Modes with Spatial Separated Charges

- \(\text{SPH} \ l=1\) modes have core-shell relative motion.

Type-II Quantum Dots

- **Type-I semiconductor junction**
  - Electrons and holes are at the same position

- **Type-II semiconductor junction**
  - Electrons and holes separate in space

- CdSe/CdTe Type-II quantum dots can make it
Expected Charge Separation Scheme

- Tellurium lone pairs serve as electron donor

Thermally or optically excited electrons will drop to the core of QD, leaving a positive ion shell
Experimental Setup for the Measurement of THz Absorption Spectra

**Bolometer**

**THz emitter**

**Objective**

**Autocorrelator**

**Grating Pair**

**Ti: Sapphire laser**
- Repetition rate: 82 MHz
- Central wavelength: 800 nm
Spectrum of THz Source

Frequency (GHz)

MMW Signal (Volt)
TEM Image of CdSe/CdTe 13nm Quantum Dots

- CdSe/CdTe Type-II Core/Shell (5.3nm/3.85nm thick)  Mean Diameter=13nm, Size variation ~20%
Photoluminescence and Absorption Spectra

- PL peaks around 1.1 \( \mu \text{m} \)
  - Recombination of CdSe electron and CdTe hole
  - Validate type-II alignment
THz Transmission Spectrum of CdSe/CdTe 13nm Quantum Dots

Original spectrum
Absorbed spectrum
Extinction Cross Section

- Calculate the absorption cross-section from transmission spectra

\[ \sigma_{ex} = -\ln(T) \frac{A}{N} \]

- Where \( T \) is the transmission, \( A \) is the excitation area, and \( N \) is the particle number in the excitation area.
THz Resonant Absorption of CdSe/CdTe 13nm Quantum Dots

- Peak 1 = 160 ± 30 GHz, Peak 2 = 250 ± 50 GHz, Peak 3 = 370 ± 70 GHz
FTIR Traces of CdSe/CdTe 10.4nm Quantum Dots

- Dielectric $\sigma_{\text{ex}} \sim 10^{-22} \text{m}^2$
- THz resonant absorption has much larger $\sigma_{\text{ex}}$ than the dielectric absorption

![Graph showing $\sigma_{\text{abs}}$ vs. Wavenumber (cm$^{-1}$)]

$33.3 \text{cm}^{-1} = 1 \text{THz}$
Sample with Different Sizes

CdSe/CdTe Type-II Core/Shell
(4.3nm/3.05nm thick)
Mean Diameter=10.4nm

CdSe/CdTe Type-II Core/Shell
(4.3nm/1.895nm thick)
Mean Diameter=8.09nm
THz Resonant Absorption of CdSe/CdTe 10.4nm Quantum Dots

- Peak 1 = 200±40 GHz, Peak 2 = 360±60 GHz
THz Resonant Absorption of CdSe/CdTe 8nm Quantum Dots

- Peak 1 = 210 ± 31 GHz, Peak 2 = 375 ± 70 GHz
Double Check with Time Domain THz Spectroscopy

- 10.1nm CdSe/CdTe QDs
- Peak 1 = 175±34GHz, Peak 2 = 322±32GHz
THz Resonant Absorption of CdSe/CdTe 11.6nm Quantum Dots

- Peak 1 = 170 ± 17 GHz, Peak 2 = 271 ± 15 GHz, Peak 3 = 333 ± 15 GHz
THz Resonant Absorption of CdSe/CdTe 8.4nm Quantum Dots

- Peak 1 = 240 ± 46 GHz
Compare with Theory

\( l=0 \) 

\[ \tan(\xi)(1-\eta^2/4) = \xi \]

\( l=1 \)

\[ 4 \frac{j_2(\xi)}{j_1(\xi)} \xi - \eta^2 + 2 \frac{j_2(\eta)}{j_1(\eta)} \eta = 0 \]

(SPH, \( l=0, n=0 \))

(SPH, \( l=1, n=1 \))

(SPH, \( l=1, n=0 \))

Frequency (GHz)

1/D (nm\(^{-1}\))
Size Dependency on $\sigma_{\text{ex}}$

- Fitting slope is 4.2, $\sigma_{\text{ex}} \sim D^4$
Charge Amount Dependence on the Particle Size

\[ \sigma_{ex} = \frac{Q^2}{b\varepsilon_0 c^*} \]

- \( Q \) : amount of charge
- \( b \) : friction constant
- \( \varepsilon_0 \) : permittivity of free space
- \( c \) : speed of light in vacuum

- \( Q \) is proportional to \( D^2 \)
  - Consistent with our assumption that the charge came from the surface donors

Use Type-II QDs in THz Imaging

- Dielectric loss at THz frequency range is small between different part of bio-tissue
- To identify the target of interest
  - Contrast agent is required
  - Type-II quantum dot is a good template

THz Transmission Imaging System

- Gunn oscillator module
- PE film
- PE fiber
- Parabolic mirror
- Golay cell
- Motion controller
- Picarin lens
- Sample under test

f=320GHz
THz Images of Nanocrystals

4.4nm CdSe  8nm CdSe/CdTe  10.4 nm CdSe/CdTe

f=320GHz
Resonant absorption
THz Transmission Images of a Dry Seahorse

- Brain cavity
- Abdominal cavity
- Spine
- Contrast agent Type-II QDs

1cm 1cm
Piezoelectricity of Nanoparticles

- If nanoparticles are piezoelectric
  - The strain of confined acoustic modes should fluctuate the dipole moment
  - Induce THz photon absorption without the help of charge separation
- Not yet demonstrated
Sample Preparation

3.4 nm

4.1 nm

4.4 nm

5.6 nm

5.8 nm

4.1 nm

4.4 nm

3.4 nm

5.6 nm

5.8 nm
Absorption Spectra of CdSe Nanocrystals

![Absorption Spectra of CdSe Nanocrystals](image-url)
Photoluminescent Spectra of CdSe Nanocrystals

![Graph showing photoluminescent spectra with wavelengths and intensities labeled]
FTIR Transmission Spectra

3.3 nm

4.1 nm

4.4 nm

5.6 nm

5.8 nm
Exctinction Cross Section Spectra

Wavenumber (cm$^{-1}$)

Extinction Cross Section (A.U.)

- 3.4nm
- 4.1nm
- 4.4nm
- 5.6nm
- 5.8nm
Eigen Frequency Equation for $l=0$ Breathing Mode

\[ \tan(\xi_o)(1 - \eta_o^2 / 4) = \xi_o \]

- where $\xi_o = \omega_o D / (2 V_L)$ and $\eta_o = \omega_o D / (2 V_T)$
- $V_L$: longitudinal sound velocity of CdSe
- $V_T$: transverse sound velocity of CdSe
- $D$: particle diameter
Compare Theory with Measured Data

- Agree well with theory
- THz photon coupled to the \( l=0 \) modes
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