

# Mechanisms for Ultrafast Generation of Coherent Phonons, Polaritons and Spin Excitations

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University of Michigan

Scientific Potential of High Repetition-Rate,  
Ultra-short Pulse ERL X-ray Source



Cornell University

VIBRATIONS (PHONONS):  
COHERENT AND SQUEEZED STATES

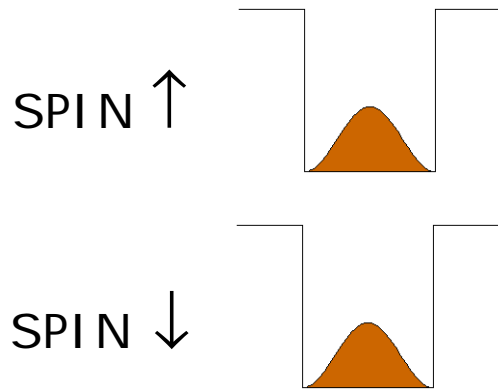
POLARITONS (LIGHT-TO COUPLED MODES)

SPIN-FLIP EXCITATIONS, MAGNONS, SPIN  
SQUEEZING, 2DEG SPIN- DENSITY  
FLUCTUATIONS

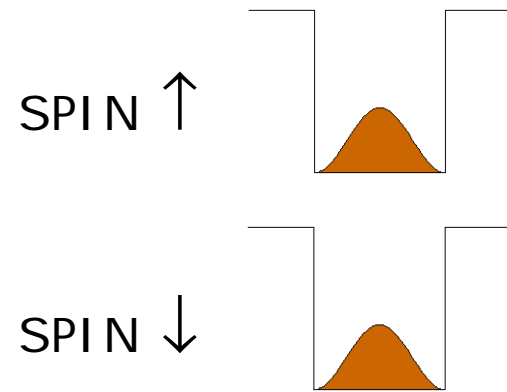
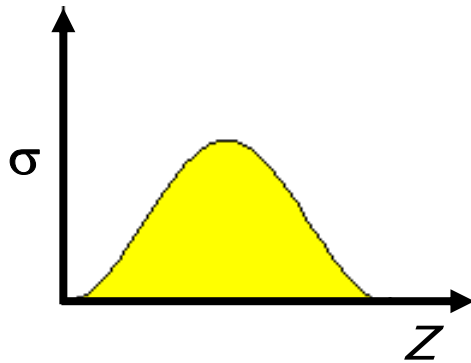
PLASMONS, 2DEG CHARGE-DENSITY  
EXCITATIONS

SUPERCONDUCTING GAP EXCITATIONS

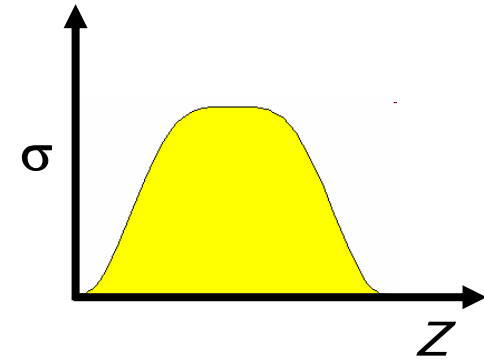
# Coherent Fluctuations: Charge-Density vs. Spin-Density



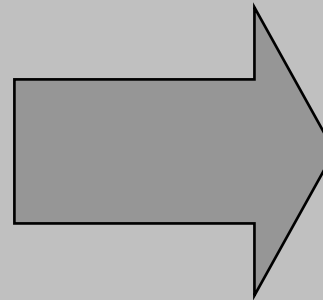
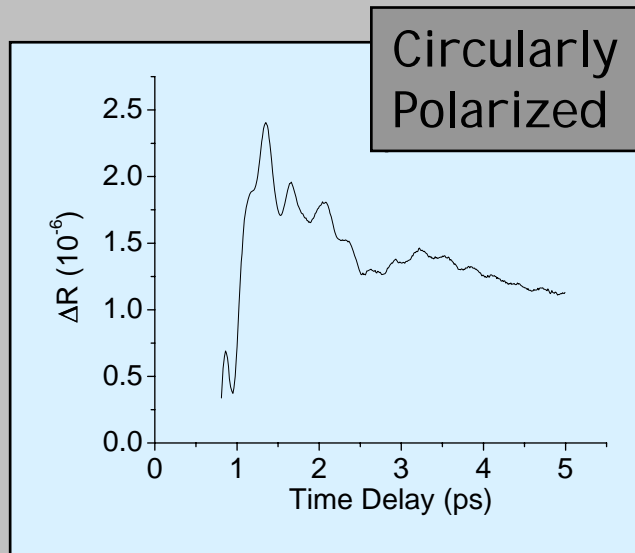
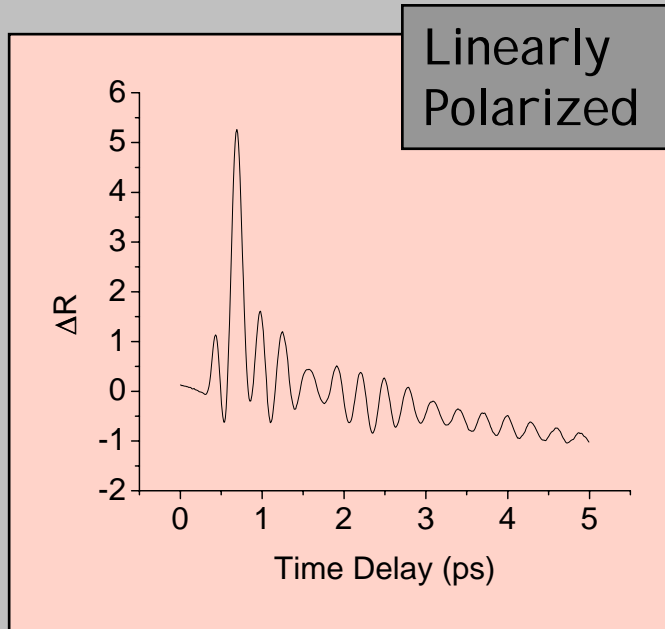
CDF



SDF

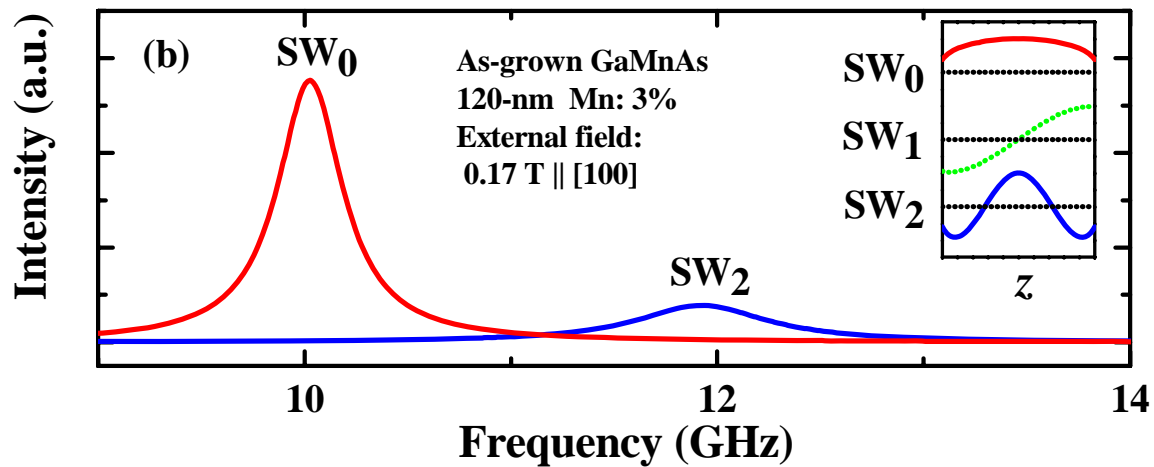
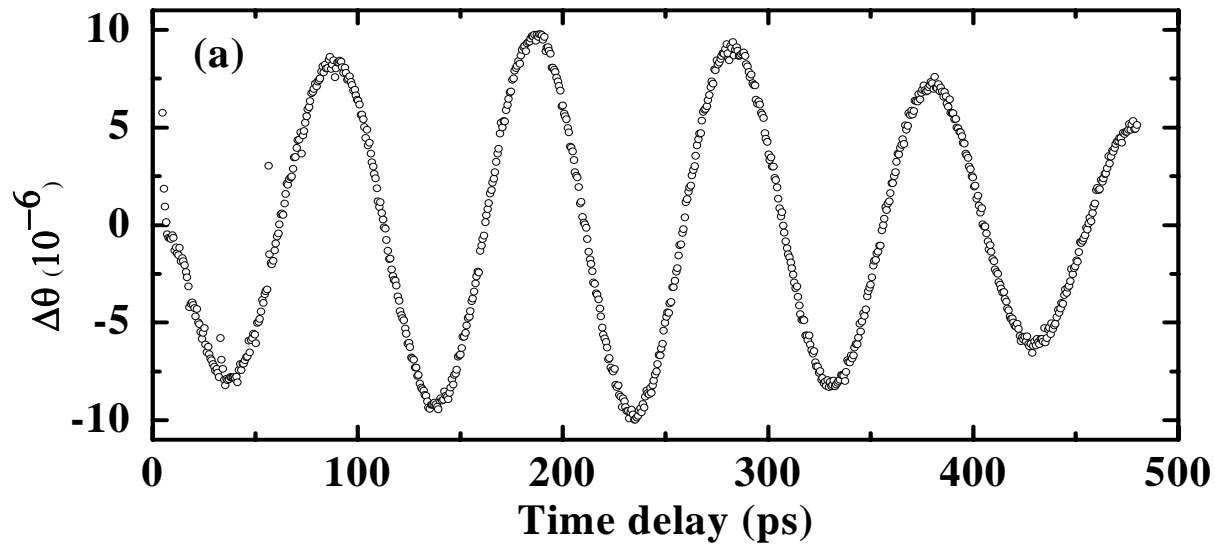


# Charge-Density vs. Spin-Density Oscillations

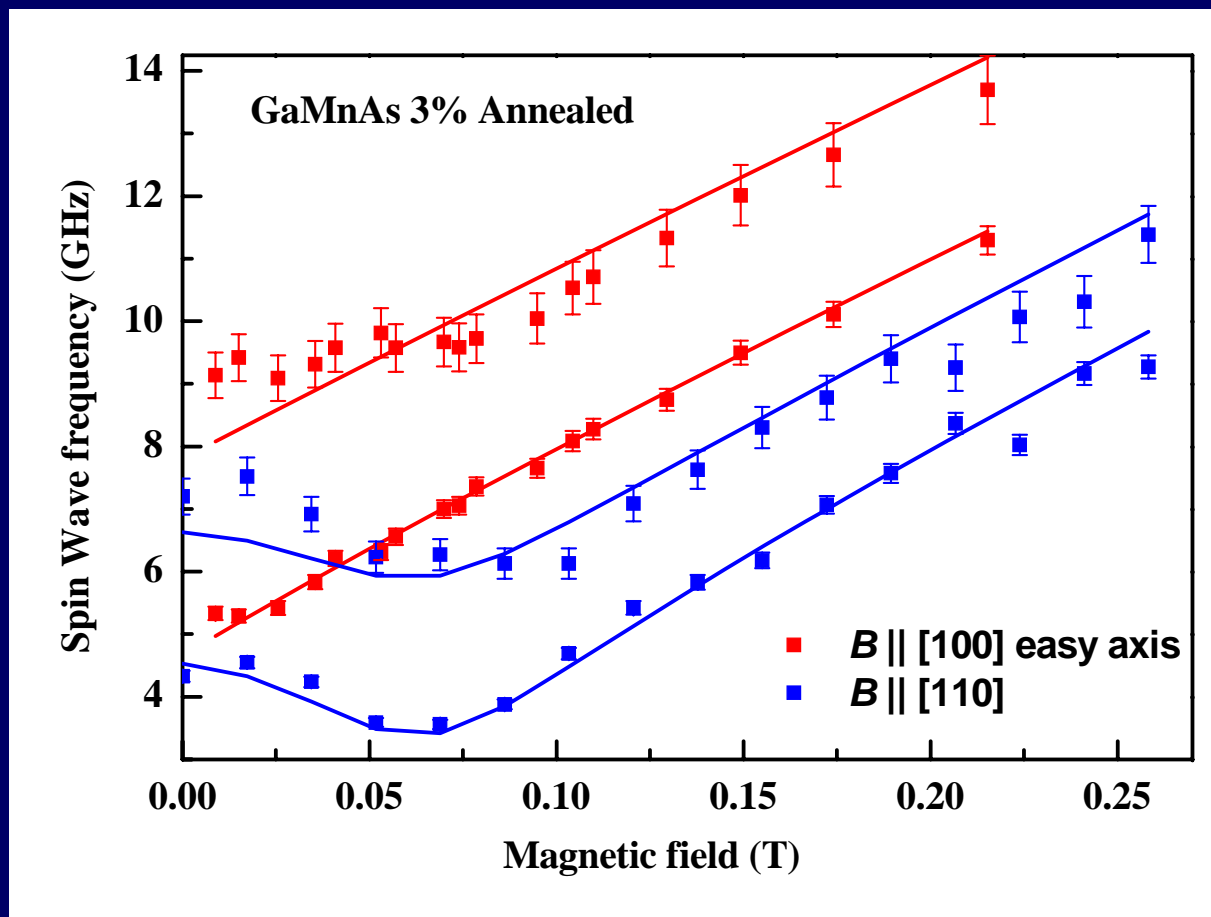


Stimulated  
Raman  
Scattering

# Ga<sub>1-x</sub>Mn<sub>x</sub>As: Time-resolved Magneto-Optical Kerr Measurements



# Dependence of the spin wave frequency on field for different directions

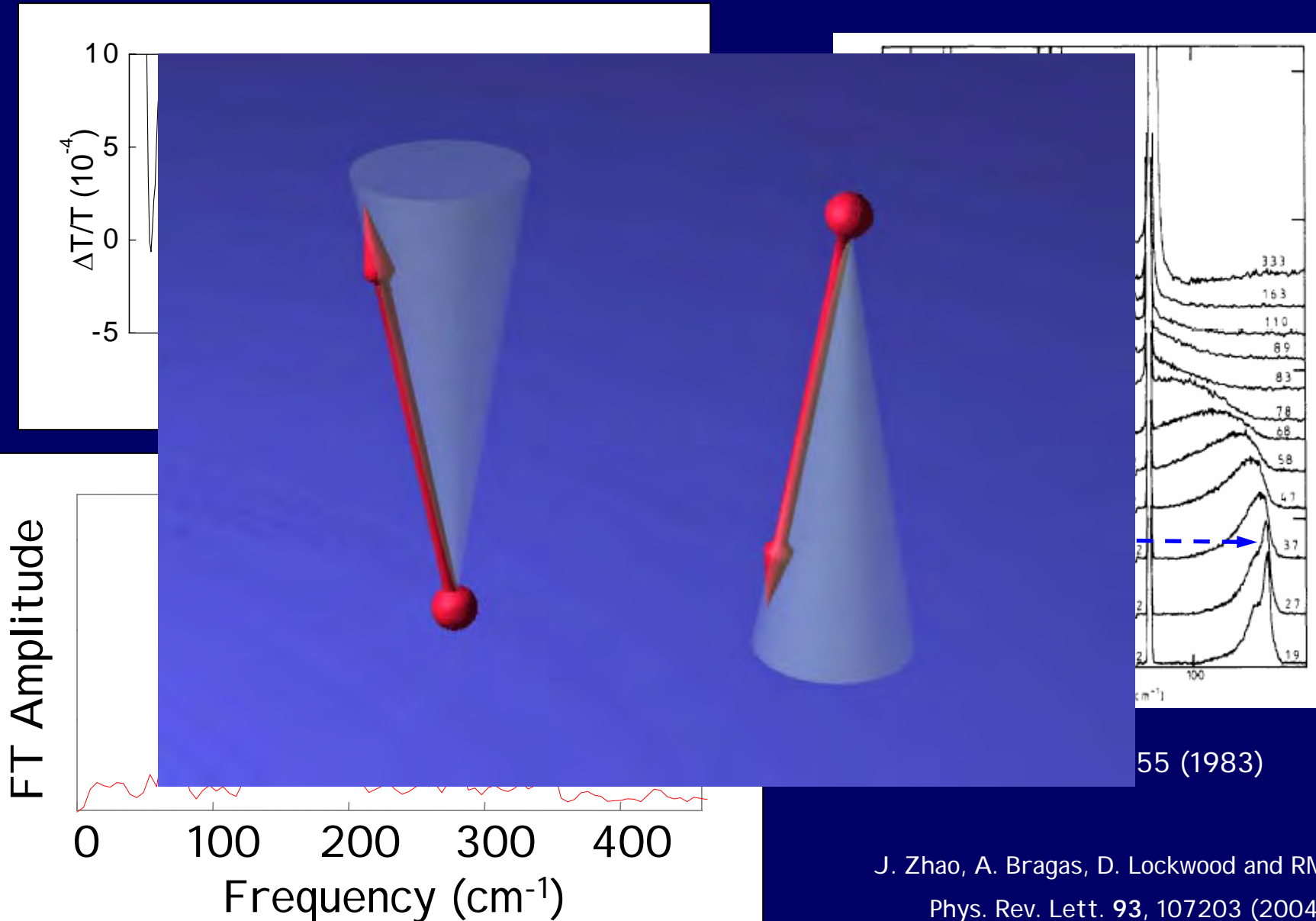


$$\omega = \gamma \sqrt{\left( H \cos(\varphi - \varphi_H) + \frac{2K_4}{m} \cos 4\varphi + Dk^2 \right) \left( H \cos(\varphi - \varphi_H) + 4\pi m + \frac{K_4}{2m} (3 + 4 \cos 4\varphi) - \frac{2K_2}{m} + Dk^2 \right)}$$

Angle between the magnetization and applied field

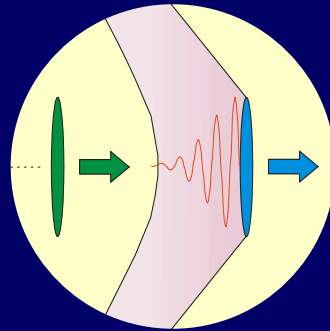
# MAGNON SQUEEZING

Spontaneous RS



J. Zhao, A. Bragas, D. Lockwood and RM  
Phys. Rev. Lett. 93, 107203 (2004)

# COHERENT POLARITONS AND CHERENKOV RADIATION

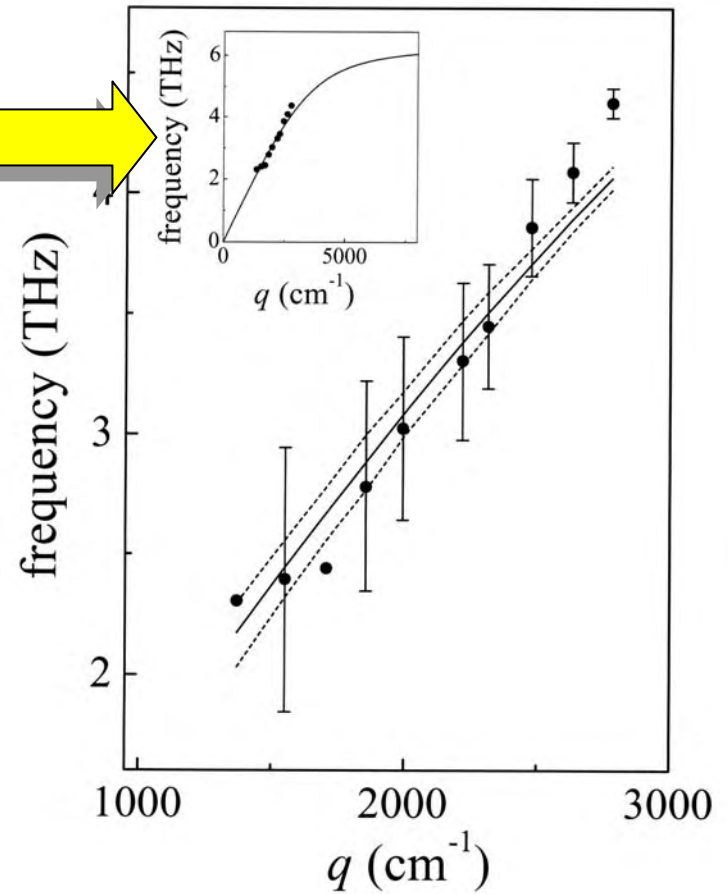
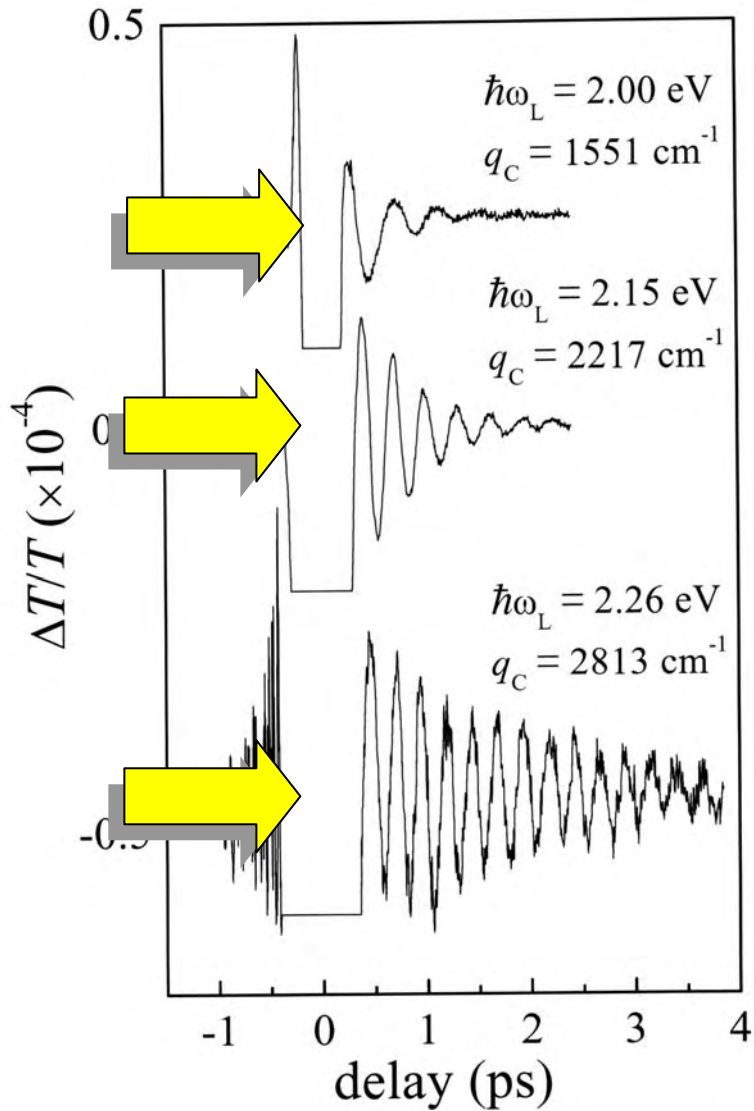


Propagation  
Effects

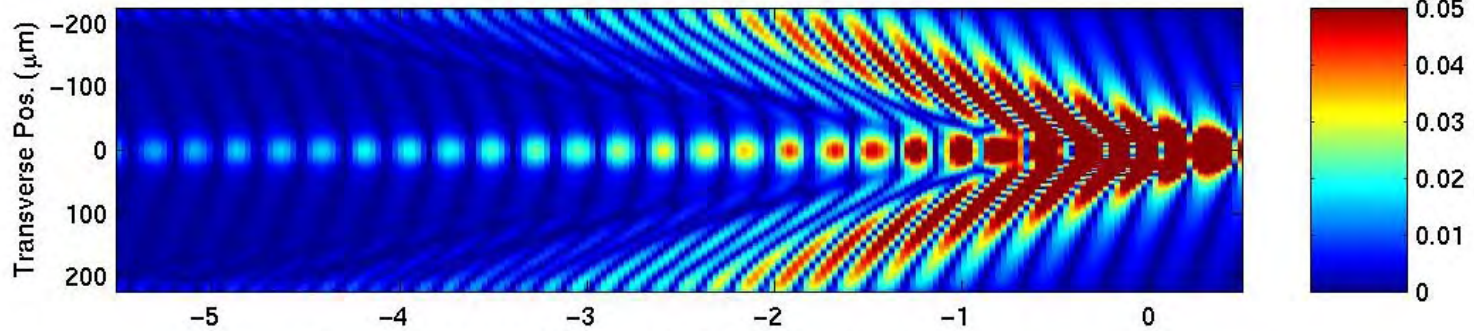
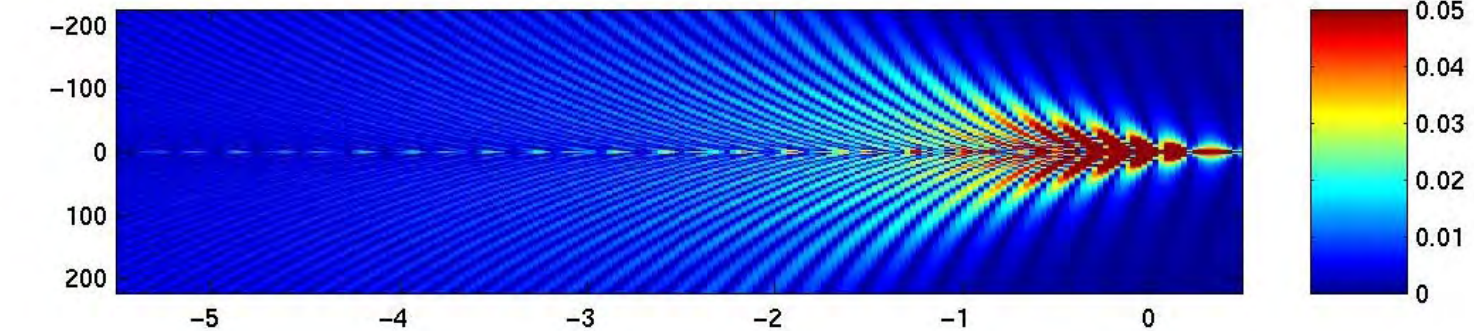


# PLANE OF DIPOLES

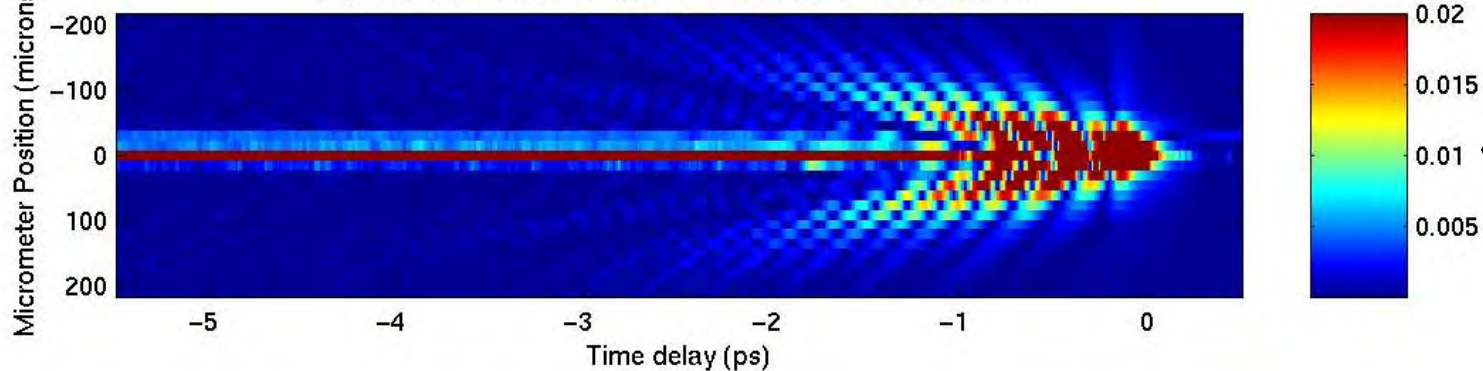
Science 291, 627 (2001)



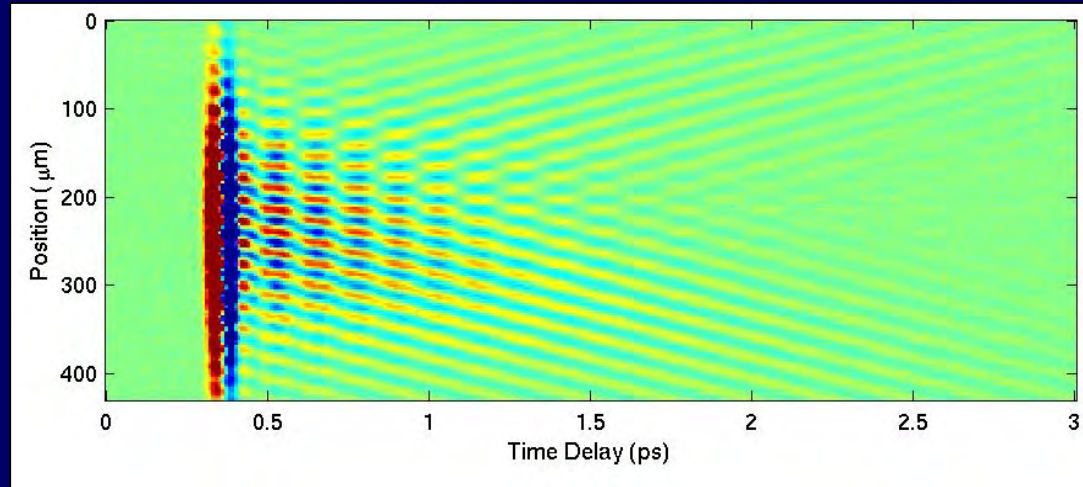
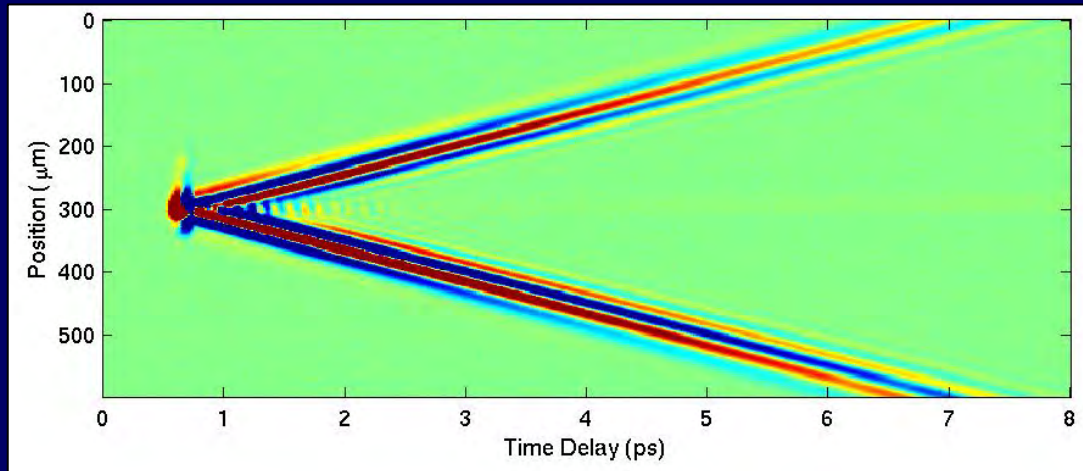
ZnSe



Experimental data, focal spot 25 microns, wavelength 810 nm



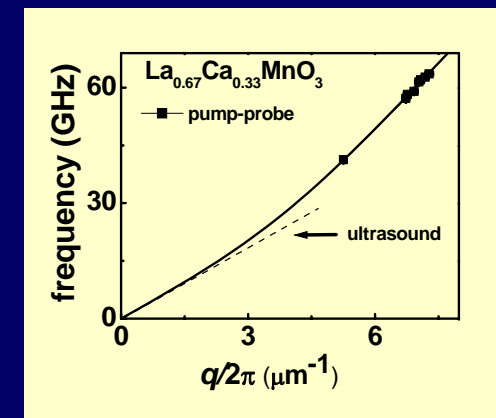
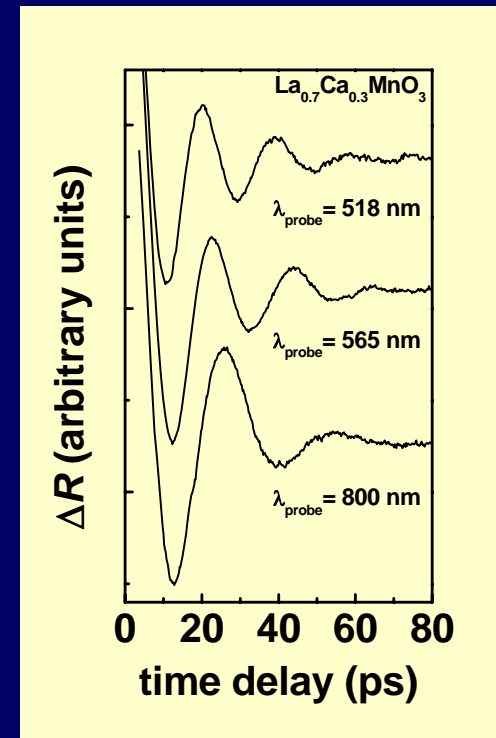
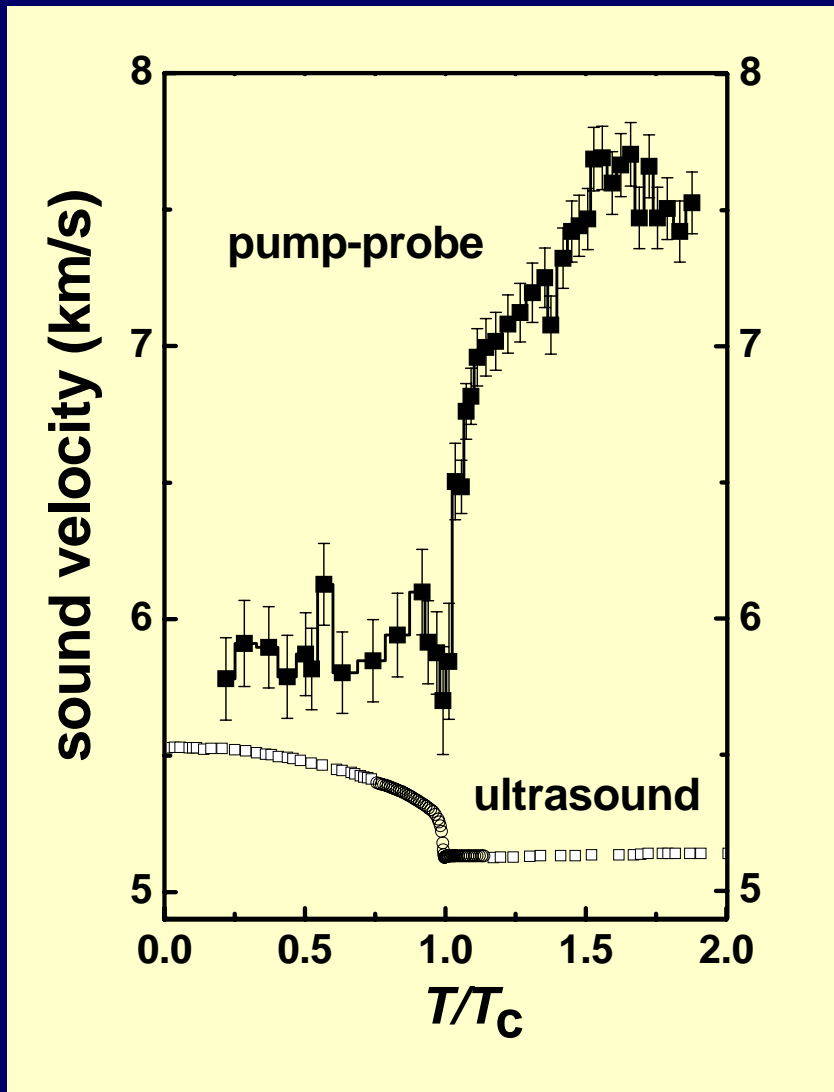
$\text{LiTaO}_3 : v = 3.07c$  (superluminal)



CHERENKOV RADIATION

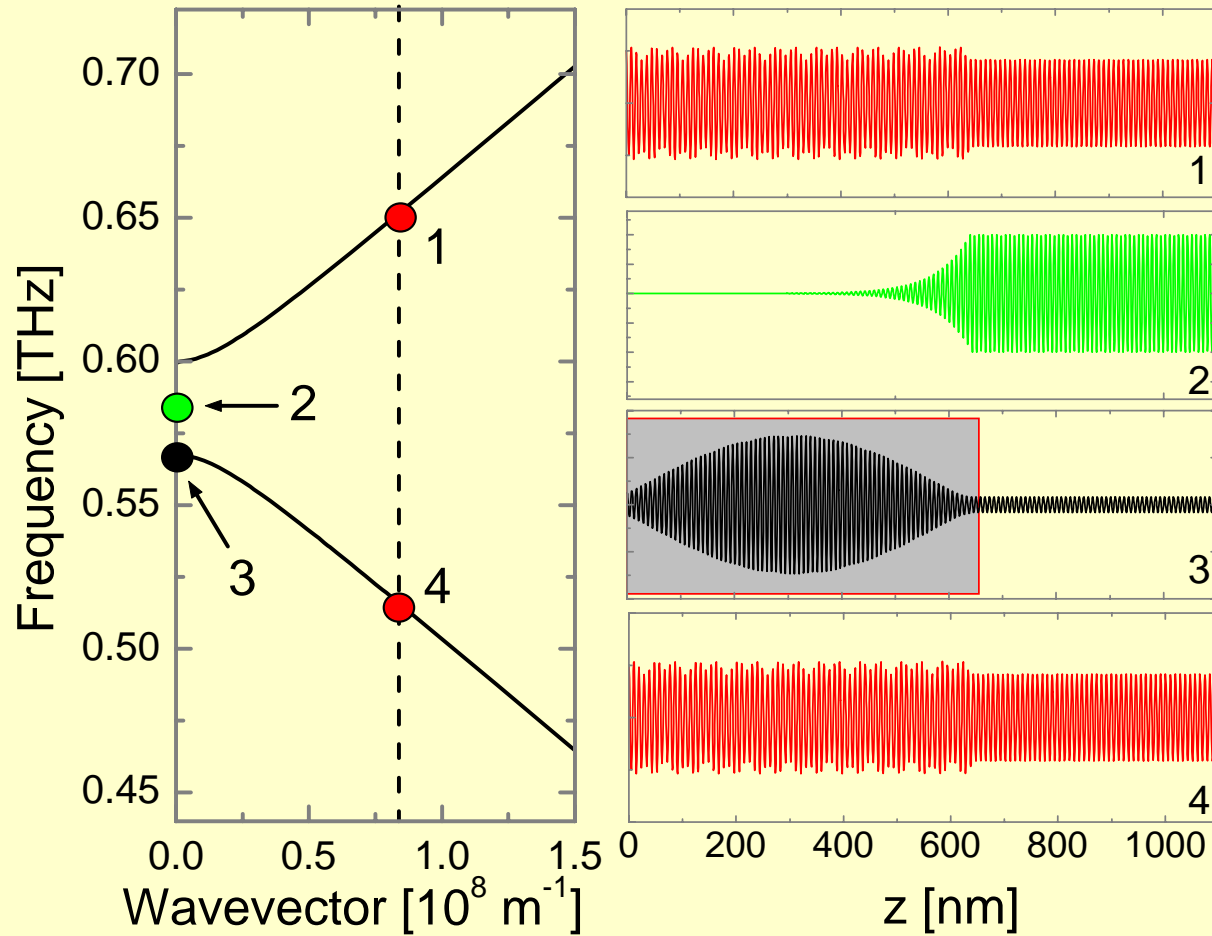
J. Wahlstrand and RM, Phys. Rev. B **68**, 054301 (2003).

# Anomalous First-to-Zero Sound Crossover in $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$

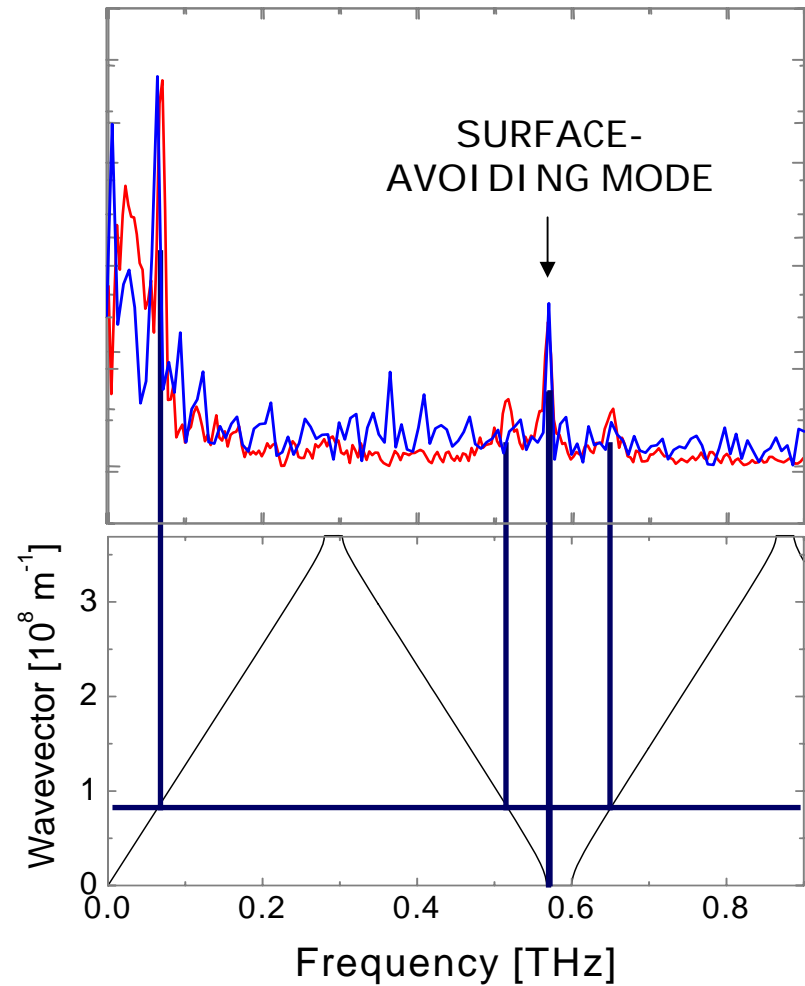
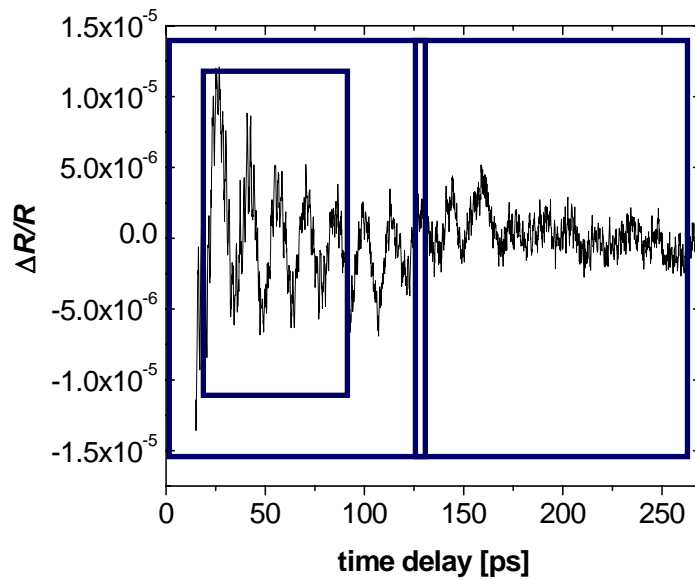
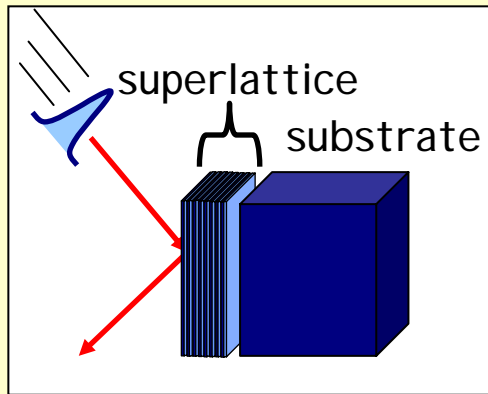


Y. Ren et al., unpublished

# Propagating and Surface Avoiding Modes

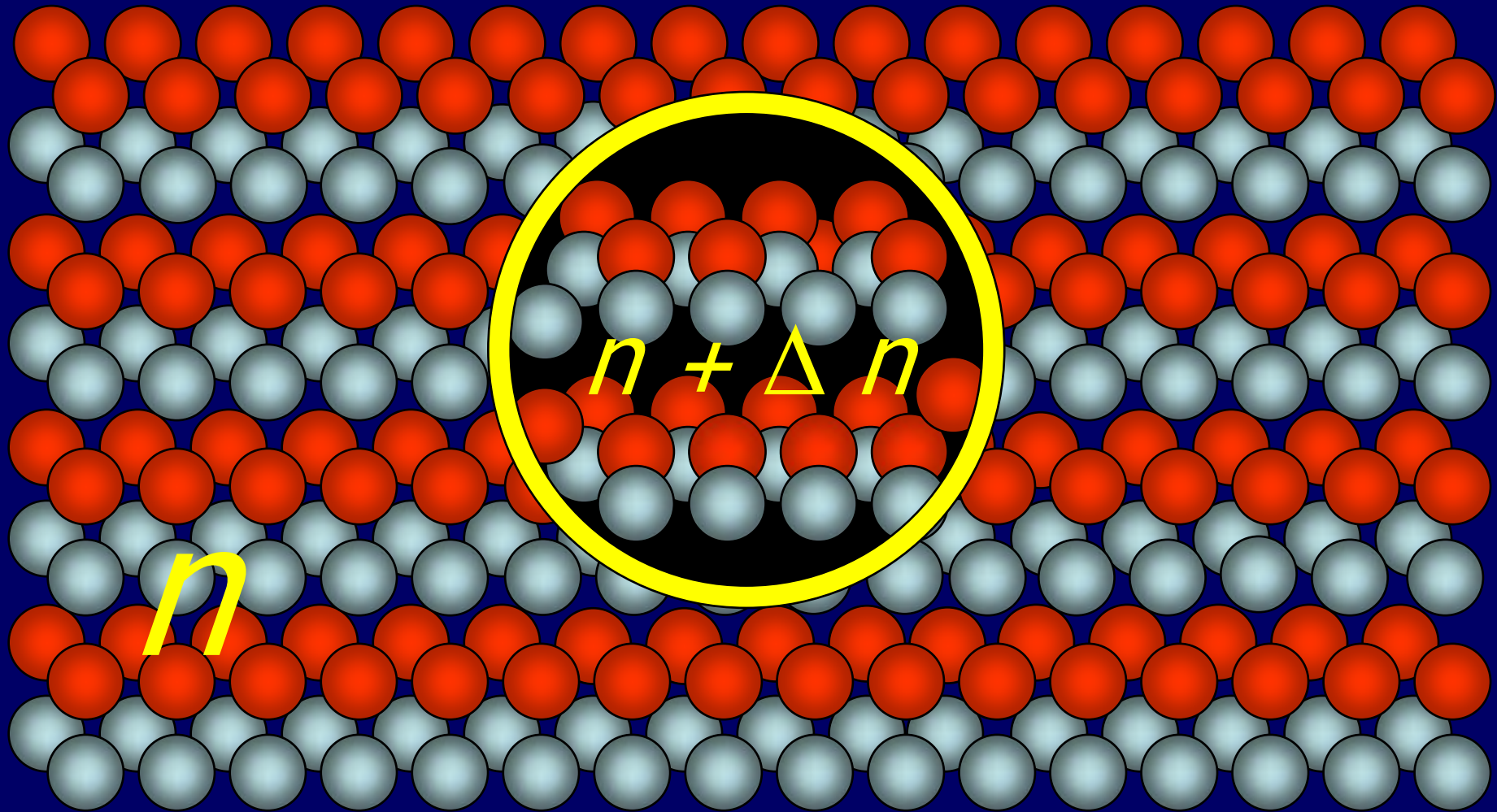


# Propagating and Surface Avoiding Modes



WHAT ARE THE  
MECHANISMS?

# WHAT ARE THE MECHANISMS? DETECTION vs. GENERATION



$$n \rightarrow n + \Delta n \quad (\Delta n \propto Q, Q^2, Q^3, \dots)$$



# LASER-INDUCED IMPULSIVE EXCITATION OF ATOMS

DISPLACEMENTS  
( $u \equiv$  ions ;  $Q \equiv$  phonons)

ELECTROMAGNETIC  
ENERGY DENSITY

$$U = \varepsilon |E(\mathbf{r}, t)|^2 / 8\pi$$

CHANGE IN  
DIELECTRIC  
RESPONSE

$$\delta\varepsilon = \sum_{im} (\partial\varepsilon / \partial u_{im}) u_{im} + \sum_{ijmn} (\partial^2\varepsilon / \partial u_{im} \partial u_{jn}) u_{im} u_{jn} + \dots$$

CHANGE IN  
ENERGY  
DENSITY

$$\delta U \approx \delta\varepsilon_{k=0} |E(\mathbf{r}, t)|^2 / 8\pi = \frac{|E(\mathbf{r}, t)|^2}{8\pi} \times$$

$$\sum_s (\partial\varepsilon / \partial Q_{s,k=0}) Q_{s,k=0} + \sum_{st,k} (\partial^2\varepsilon / \partial Q_{s,k} \partial Q_{t,-k}) Q_{s,k} Q_{t,-k} + \dots$$

FORCE  
DENSITY

$$F \propto |E^2(t)|$$

FIRST-ORDER  
IMPULSIVE FORCE

$$F \propto Q_k |E^2(t)|$$

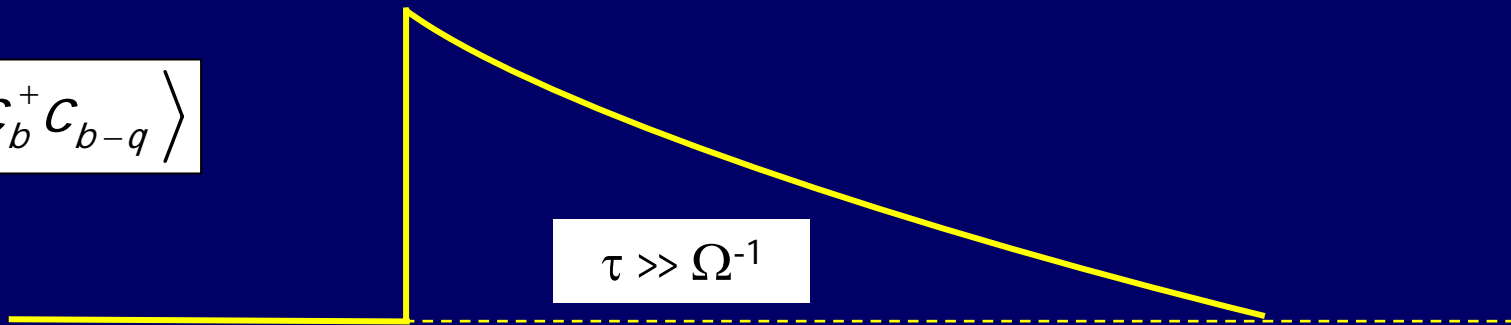
SECOND-ORDER  
IMPULSIVE CHANGE OF  
FREQUENCY

## IMPULSIVE STIMULATED RAMAN SCATTERING

# ABOVE THE GAP: DISSIPATIVE EXCITATION

$$H = \sum_b \varepsilon_b c_b^\dagger c_b + \frac{1}{2} \sum_q (P_q^2 + \Omega_q^2 Q_q^2) + \sum_{kk'} \Xi_{kk'} Q_{k-k'} c_k^\dagger c_{k'} \quad \longrightarrow \quad \ddot{Q}_q + \Omega_q^2 \langle Q_q \rangle = - \sum_k \Xi_{k,k-q} \langle c_k^\dagger c_{k-q} \rangle$$

$$F \propto \langle c_b^\dagger c_{b-q} \rangle$$



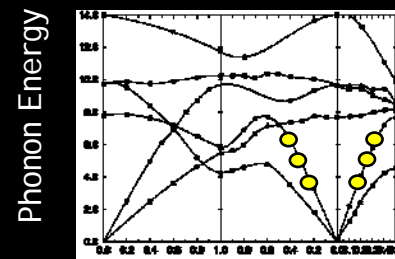
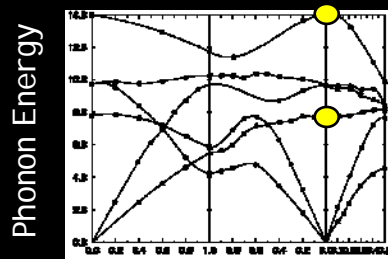
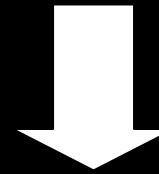
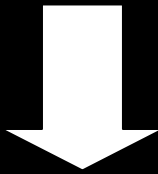
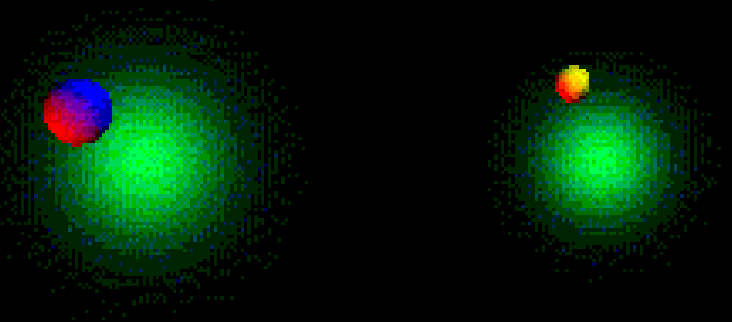
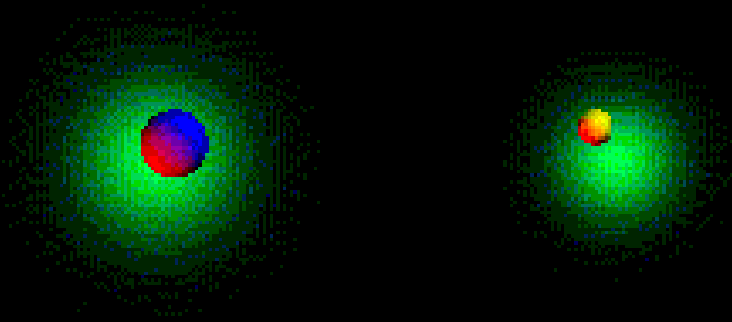
$$F(\Omega) \propto \left[ \frac{d \operatorname{Re}(\varepsilon)}{d\omega} + 2i \operatorname{Im} \varepsilon / \Omega \right] \int_{-\infty}^{+\infty} e^{i\Omega t} |E(t)|^2 dt$$

TWO RAMAN TENSORS: T. Stevens, J. Kuhl and RM

Phys. Rev. B 65, 144304 (2002)

# COHERENT PHONON FIELD

# SQUEEZED PHONON FIELD



Wavevector

Wavevector

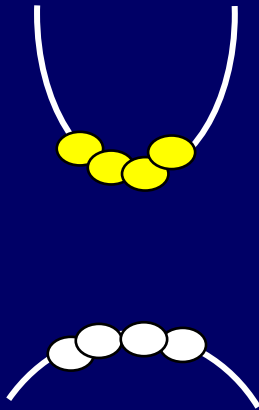
$$K=0$$

$$K_1+K_2=0$$

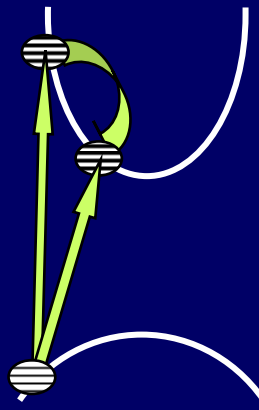
# DECP vs. COHERENT RAMAN SCATTERING

$$F_{DECP} \propto \sum_b \langle c_b^+ c_b \rangle$$

$$F_{RS} \propto \sum_b \langle c_b^+ c_{b-q} \rangle$$



incoherent  $\rightarrow$  coherent  
A-type modes

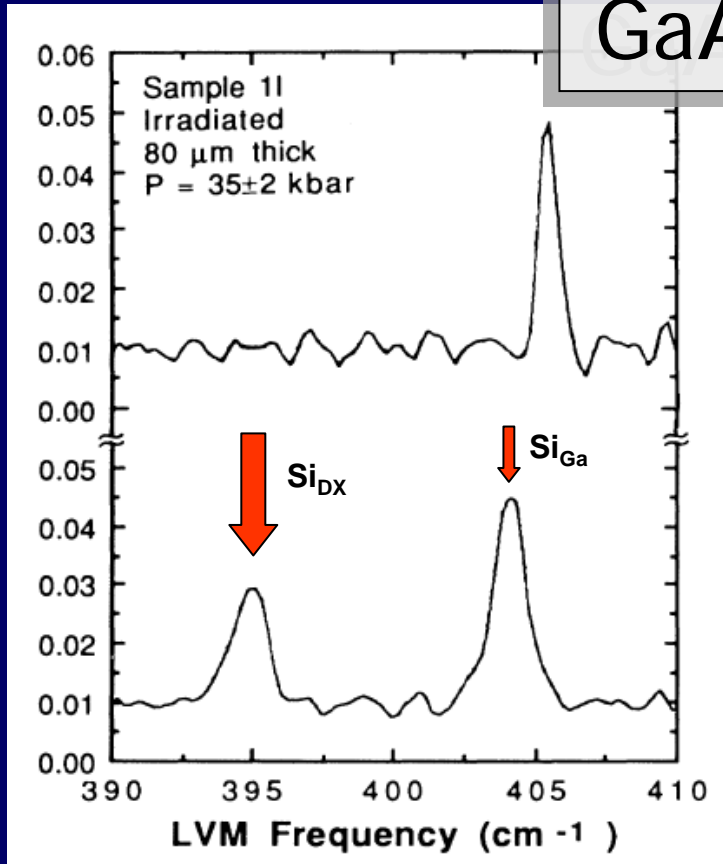


Long-lived Raman coherence  
All Raman-allowed modes

Good (?)  
ERL Idea

# Time Resolved EXAFS: Impurity Mode Identification

GaAs



ZnO

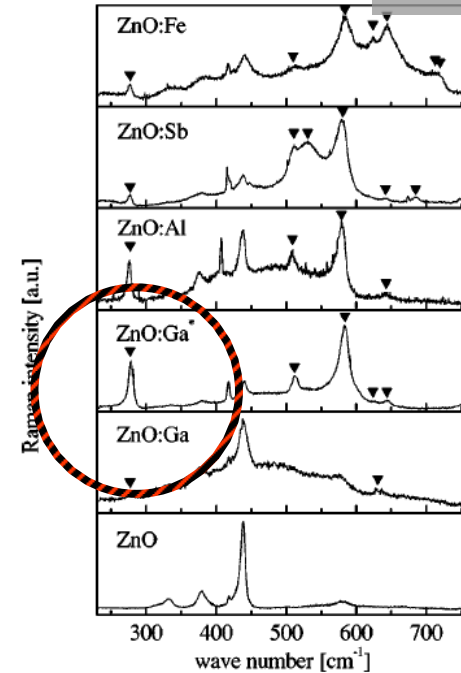


FIG. 2. Polarized micro-Raman spectra in the  $x(\gamma\gamma)x'$  scattering configuration for undoped, Ga, Al, Sb, and Fe doped ZnO thin films on sapphire. The ZnO:Ga\* film was grown in  $\text{N}_2\text{O}$  atmosphere, all other samples were grown in  $\text{O}_2$  atmosphere. Triangles denote additional modes.

VOLUME 66, NUMBER 6

PHYSICAL REVIEW LETTERS

11 FEBRUARY 1991

## Local-Vibrational-Mode Spectroscopy of $DX$ Centers in Si-Doped GaAs under Hydrostatic Pressure

J. A. Wolk,<sup>(1),(2)</sup> M. B. Kruger,<sup>(1)</sup> J. N. Heyman,<sup>(1)</sup> W. Walukiewicz,<sup>(2)</sup> R. Jeanloz,<sup>(3)</sup>  
and E. E. Haller<sup>(2),(4)</sup>

APPLIED PHYSICS LETTERS

VOLUME 83, NUMBER 10

8 SEPTEMBER 2003

## Raman scattering in ZnO thin films doped with Fe, Sb, Al, Ga, and Li

C. Bundesmann,<sup>(1)</sup> N. Ashkenov, M. Schubert, D. Spemann, T. Butz, E. M. Kaidashev,<sup>(1)</sup>  
M. Lorenz, and M. Grundmann

Silicon K-shell: 1.84 keV

Gallium K-shell: 10.4 keV

# Time Resolved EXAFS: Impurity Mode Identification

